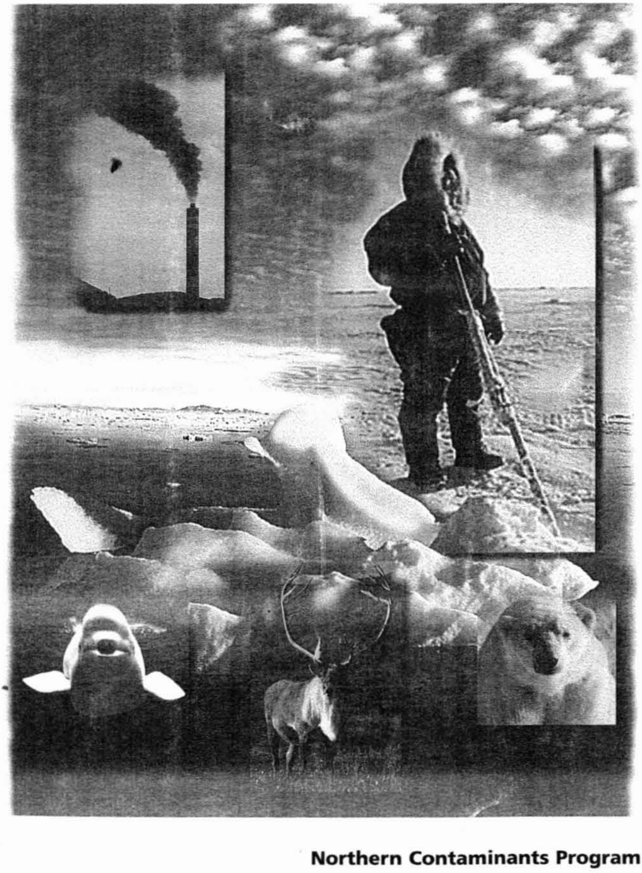
Canadian Arctic Contaminants Assessment Report



Northern Contaminants Program

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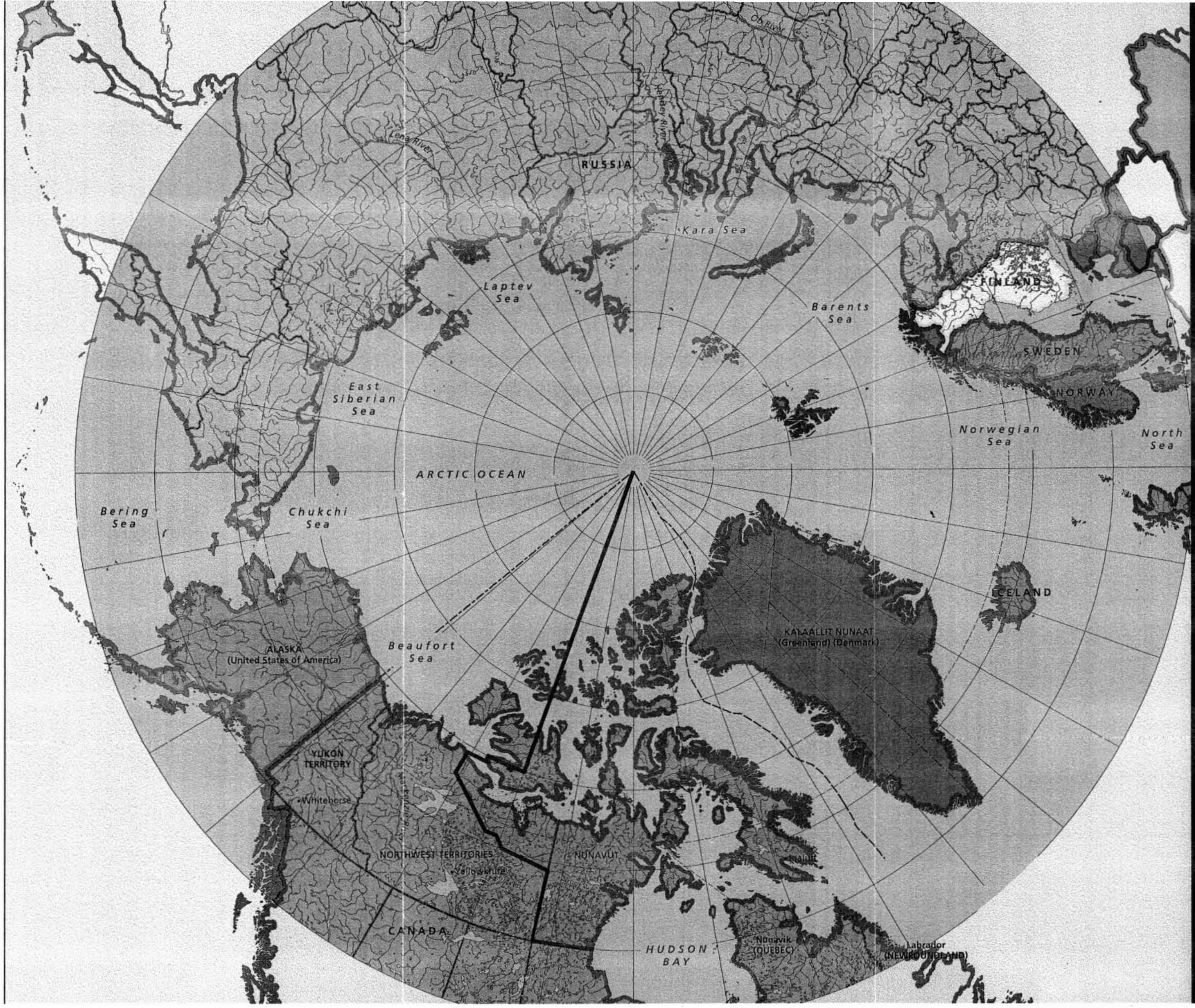
Circumpolar Map



Canadian Arctic Contaminants Assessment Report

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Working together, we will make the North a safer and more prosperous place for present and future generations.

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Executive Summary

CHAPTER 1
Introduction

Contaminants have been detected throughout Arctic ecosystems at unexpectedly high levels. Many of these contaminants bioaccumulate and reach high concentrations in animals at the top of the food chain. Northern residents, especially Aboriginal peoples, are susceptible to the potential adverse effects of contaminants, due to a dietary dependence on arctic fish and wildlife. Subsistence fishing and hunting is an integral part of Aboriginal culture.

After five years of intensive research and monitoring, a significant amount of information has been assessed under the Arctic Environmental Strategy's Northern Contaminants Program (AES-NCP). The NCP works to:

i) measure contaminant levels in the Canadian north;
ii) find out where contaminants are coming from, how they get to the north, and what happens when they arrive;
iii) assess effects of contaminants on health of the northern environment, including humans;
iv) provide information to assist Northerners' decision-making and encourage their involvement in the NCP; and
v) pursue international agreements and cooperation on controlling contamination at a global level.

The NCP has set new standards for community participation in scientific programs. Through its partnership structure the NCP has had the capacity to address arctic ecosystem contamination using approaches that ensure scientific integrity and relevance, while responding to community concerns and priorities.

The purpose of producing this report is to provide an evaluation of the current state of contamination in the Canadian Arctic environment. The NCP results constitute a large proportion of the global knowledge base on arctic contaminants. This report provides all information produced to date under this program. Its companion document, *Highlights of the Canadian Arctic Contaminants Assessment Report: A Community Reference Manual*, was prepared in consultation with the AES Aboriginal Partners and with input from community-level participants.

CHAPTER 2
Sources, Occurrence and Pathways

Contaminant Sources

- Organochlorines (OCs) have industrial and agricultural (pesticide) uses. They are also released as by-products of combustion. From the 1950s to the present, they have been intensively used over a large part of the globe. Sources of OCs within the Canadian Arctic are minor. For example, dumped PCBs are found within the immediate vicinity of DEW line sites. However, for the entire Yukon and NWT, dumped PCBs are only 1% of the total deposition of PCBs over 30 years.
- Long-range transport from other parts of the world via the atmosphere, ocean and rivers is clearly the dominant source of OCs to the Canadian Arctic.
- Ranked in order of total global usage between 1950 and 1992, the most-used persistent organochlorines were: HCHs (6.3 Megatonnes), DDT (2.6 Mt), toxaphene (1.33 Mt) and PCBs (1.2 Mt). HCHs are the most abundant OC in arctic air and water.
- Most usage of HCH has occurred in China, India and the former USSR. China and India switched to using only the pesticide-active isomer of HCH (γ-HCH or lindane) in 1983 and 1990, respectively. The annual usage of HCH globally shows a decrease in these years.
- A prime product of NCP research has been the production of a globally gridded (1 lat. x 1 long.) inventory of annual usage of the HCH isomers for 1980 and 1990. This is essential for 3-D environmental models of pathways.
- PAHs, metals and radionuclides have natural as well as anthropogenic sources. At any given location, the PAH mixture of chemicals found in the Arctic originates from anthropogenic (coal and oil combustion) and natural sources.
- For the metals of concern in the NCP, the ratios of global anthropogenic to natural emissions to the atmosphere in the mid-1980s are: 28:1 (Pb); 5.4:1 (Cd) and 1.4:1 (Hg). Atmospheric inputs of anthropogenic Cd to the Arctic Ocean are less than 1% of the ocean current inputs of natural Cd. In the terrestrial environment the deposition of anthropogenic atmospheric Cd onto vegetation may be

sufficiently isolated from the huge natural soil reservoir of Cd that it can be selectively grazed and enter the caribou-wolf food chain. Best global budget estimates of Hg sources to the atmosphere indicate three roughly equal components: oceanic degassing, natural terrestrial sources, and anthropogenic releases. There is tentative observational evidence indicating that that there has been a three-fold increase in Hg concentrations in air and in the upper ocean since pre-industrial times.

- Long-range transport of radionuclides into the Arctic is not significant. Atmospheric fallout and European reprocessing plants have been the greatest contributors to anthropogenic radionuclides observed in the arctic environment. Uranium processing plant effluent that was released into European waters in the past from the UK and France is now radiologically inactive. The main source of locally dumped radioactive waste is from the Russian continental shelves, particularly in the Kara and Barents seas. Studies show that presently there is no indication of contamination. Radioactivity will inevitably be released as containment of waste deteriorates, but how long this might take is not known. If leakage occurs, threats may be posed on local and regional scales, but it is not likely that there will be a significant threat to the general arctic environment. Ocean and ice currents do not favour the movement of waste to the North American Arctic.

Contaminant Transport Pathways

- Between 1991 to 1996, significant progress was made by the Northern Contaminants Research Program (NCP) in understanding pathways of contaminant movement in the Arctic environment. Advancement of knowledge has been made in 93% of possible pathways for OCs, 60% for PAHs, 32% for metals and 34% for radionuclides.

Atmospheric Transport

- Contaminants can be subdivided into two types according to their atmospheric pathway: "one-hop" and "multi-hop" compounds. If a substance is emitted to the atmosphere, transported and deposited to the surface never to return to the atmosphere, it is a "one-hop" compound. One-hop contaminants include heavy metals (except Hg) as well as the relatively involatile OCs (e.g. DDT) and PAHs (e.g. B[a]P). In contrast, if a compound re-enters the atmosphere after initial deposition to the Earth's surface, it is a "multi-hop" compound. Processes by which this can occur include volatilization from the Earth's surface under warmer temperatures than those in which it was deposited, sudden exposure of ocean water to the atmosphere after being covered by ice or resuspension by winds of soil dust. Most OCs, many PAHs and Hg fall into the "multi-hop" group.

Marine Transport

- Contaminants are transported to the Arctic Ocean via ocean currents. Water flows to the surface Arctic Ocean from the Atlantic Ocean through eastern Fram Strait and the Barents Sea (80%) and from the Pacific Ocean through Bering Strait (17%). Precipitation and rivers contribute the remaining 3%. Water flows out of the Arctic through the Canadian Archipelago (34%) and western Fram Strait (62%). The rest (4%) flows out in ice.
- Other oceanographic factors control the transport of contaminants within the Arctic Ocean. In winter, a polar mixed-layer is produced by the freezing of ice and concomitant exclusion of brine (approximately the top 50 m). In summer, a stratified surface layer is produced by the melting of sea ice and the addition of runoff at the margins (5–10 m deep). The influence of direct exchange of dissolved gases with the atmosphere is therefore limited to these shallow ocean depths. The near-surface water layer (30–50m) is probably the most important layer in terms of the transport of contaminants and their entry into marine biota.
- The mean residence time of water in the Arctic Ocean surface layer is 12.5 years. Beneath the surface Arctic Ocean layer (0–200 km) is the warm Atlantic layer, which circulates on time scales of the order of 30 or more years. The deep basin waters (1500–4000 m) are oldest and have the longest residence times taking in the order of centuries to replace their water.
- This research tells us where contaminants are coming from, how long they will remain in the Arctic Ocean, how they enter into the food chain, and how they flow out of the Arctic Ocean. Contaminants in Arctic Ocean water trapped under sea ice will slowly drain through the Canadian Archipelago, on a time scale of decades. Outflow of contaminated water will impact biota in the Archipelago and Baffin Bay.
- The defining characteristic of the Arctic Ocean is sea ice. Despite its small contribution to the total volume of water in the Arctic Ocean, sea ice has a potentially important role in the transport of contaminants and their entry into biota. First, sea ice provides a solid interface at the ocean's surface, and therefore, is an important habitat for a wide range of organisms from plankton to bears. Second, it collects atmospheric inputs throughout winter and spring to be released in summer into melt ponds and surface water at a time when biological productivity is at its highest. Third, much of the

ice in the Arctic is produced in shallow marginal seas during winter and exported to the interior ocean. Ice forming on shelves can incorporate contaminated sediments. This contaminated ice may then be transported thousands of kilometres undergoing little or no alteration.

Riverine Transport

- Rivers also transport contaminants to the Arctic Ocean. 78% of the total annual riverine input of water to the Arctic Ocean is via ten large rivers, nine Russian and one Canadian — the Mackenzie River. The Mackenzie River carries 75% of the total Canadian riverine water input to the Arctic Ocean. The rest is through a number of smaller rivers that discharge into the Canadian Archipelago and play an important role in the generally shallow, more productive waters of the Archipelago. An important feature of arctic rivers is extreme seasonality in waterflow, therefore suspended solid content and associated contaminants can be delivered in high levels over a short period.

Contaminants in the Arctic Atmosphere

- For the first time, a database of at least two years of routine observations of the important OCs, PAHs and Hg in the Arctic atmosphere exists for Alert, NWT; Tagish, Yukon; Cape Dorset, NWT; and Dunay Island, Russia. Ranking OCs according to their mean annual concentrations in air yields the following abundance order: ΣHCH > ΣPCB > toxaphene ≈ Σchlordane ≈ endosulfan > pentachloranisole > chloroveratroles > ΣDDT ≈ dieldrin > octachlorostyrene > methoxychlor ≈ endrin > trifluralin ≈ mirex. Many compounds (e.g. toxaphene) have a strong seasonality in their atmospheric concentrations that is likely related to the temperature dependence of phase partitioning between the atmosphere and Earth's surface.
- The composition of the complex PCB mixture of compounds differs between the North American Arctic and the Russian Arctic. Heavier more chlorinated compounds are found in Russia.
- PAHs in the Arctic peak in the winter months of December to February overlapping the Arctic haze season. In the high Arctic, the most abundant PAH compounds in air are fluorene, phenanthrene, fluoranthene and pyrene.
- Mercury concentrations at Alert, NWT are three times higher than concentrations observed in Antarctic air. Natural and anthropogenic sources of atmospheric Hg are greater in the northern hemisphere than the southern hemisphere.

Contaminants in Arctic Snowfall and Snowpack

- The most abundant OCs in arctic fresh snowfall at Alert NWT are: ΣHCH ≈ ΣPCB> ΣDDT ≈ ΣHCB ≈ ΣChlordane. Concentrations are generally much higher in the high Arctic than at the Tagish, Yukon site.
- A detailed snowpack chemistry survey in the Arctic in spring 1992 yielded the following ranking of OCs according to abundance: ΣPCBs > ΣHCHs > ΣDDT ≈ ΣHCB ≈ ΣChlordane. Thus compared to snowfall, snowpack contains relatively less ΣHCH than ΣPCB. The former is more volatile than the latter.

Contaminants in the Arctic Marine Environment

- New observations confirm that the relative abundance of OCs in arctic seawater is α-HCH > HCBs > γ-HCH ≈ CHBs (toxaphene) > ΣChlordane ≈ PCBs > DDTs.
- PAH composition indicates that anthropogenic, atmospheric PAHs are minor contributors to particle-associated PAHs in the Mackenzie River estuary but relatively more important for the surface and offshore waters of the shelf.
- Because the oceans have been contaminated globally by industrial lead (Pb), there now exists no baseline in the surface oceans against which to assess contaminant Pb inputs. However, contaminant Pb is known to have been delivered in significant quantities to the Arctic. In surface marine sediment cores of the Canada basin, it is present at higher concentrations near the surface than in deeper, pre-industrial period sediments.
- There is no recent data for mercury (Hg) concentrations in Arctic Ocean waters with which to assess its natural or contaminant geochemistry or its trends. Determining the distribution of Hg in the Arctic Ocean remains a priority for future research.
- Levels of radioactivity (^{137}Cs and ^{239,240}Pu) in nearshore marine sediments in the Canadian Arctic are typical of those measured at similar latitudes in other regions of the Arctic. ^{137}Cs inventories can be explained entirely by fallout — that is, there is no detectable excess ^{137}Cs from the former Soviet Union dumping activities. ^{137}Cs-laden ice does not appear to be a significant radiological risk.

Contaminants in the Terrestrial/ Freshwater Environment

- The relative abundance of OCs in arctic rivers is ΣHCH ≈ ΣPCB > ΣDDT ≈ ΣChlordane. Most of this is carried in the dissolved phase.

- The order of abundance of OCs in northern lake surface sediments is ΣPCB> ΣDDT> ΣHCH ≈ toxaphene ≈ ΣChlordane.
- The suspended sediment load of the Mackenzie River is the major source of PAHs to the Beaufort basin of the Arctic Ocean. The PAHs are primarily natural in origin.
- Hg is the most important metal in Arctic lakes from a toxicology viewpoint. Observations show and models confirm that of the total Hg that enters a high arctic lake about one third is retained in the sediments, half is exported downstream and the rest is lost to the atmosphere.
- The main source of toxaphene, ΣPCB and HCH to Yukon River basin lakes is atmospheric rather than local dumping. Lake Laberge receives over 90% of HCH loadings from the Yukon and Takhini rivers.
- Results of Yukon lake studies did not support the explanation that different OC concentrations in fish could be due to major physical or limnological differences among lakes.

Trends

- In parallel with trends in global usage of HCH, there has been a significant decrease in observed arctic atmospheric concentrations between 1978 and 1990. Sharp decreases occurred in the mid-1980s and in 1991.
- Global emissions of lead to the atmosphere have decreased greatly since the 1970s. This corresponds to a significant decrease observed between 1980 and 1995 in atmospheric lead levels in the Arctic. This is clearly an example of pollution controls that work.
- In Canada, historical records of contaminants in glacial ice have been reported. The record shows the appearance of PCBs in about 1960 after they were synthesized and commercially distributed. There is no trend evident after 1960. PAHs did show a trend. They peaked in the 1950s and 1960s and then settled down to a relatively constant concentration from 1970 to 1992. More measurements are needed of OCs, PAHs, Pb and Hg in northern glaciers.
- In lake sediments, there is a clear record of contaminant deposition history. Sub-arctic lakes show an onset of PCBs in the 1940s (±10 years) in reasonable agreement with synthesis and release history. In contrast, high arctic lakes show no significant PCB inputs until the 1960s (±10 years), lagging the initiation of production by about twenty years. This difference is consistent with large scale model predictions of the lag time in diffusion of PCBs released at mid-latitudes to the north and with glacial ice PCB records.
- Lake sediment records of anthropogenic lead and mercury show clearly that anthropogenic sources contribute substantially to lake inputs. Unlike synthetic organic compounds such as the OCs, these metals also have a natural geological source. The ratio of the current deposition flux to the natural, pre-industrial flux in numerous lakes sampled ranges from 1.1 to 7 for Hg and from 1.1 to 10 for Pb.

CHAPTER 3
Ecosytem Uptake and Effects

Overall Assessment of Contaminants in Freshwater Environments

- Mercury is the one contaminant consistently exceeding guideline limits in fish for subsistence consumption or commercial sale. Lake trout and northern pike in the Canadian Shield lakes of the NWT and northern Québec generally have the most elevated levels.
- Toxaphene is the major OC contaminant in all fish analysed. Highest toxaphene levels are generally seen in fish that are strictly piscivorous such as lake trout and burbot in Lake Laberge and burbot in the east arm of Great Slave Lake. Toxaphene, as well as mercury and PCB concentrations tend to increase with increasing fish size.
- Chlorinated dioxins and -furans have been determined in fishes from the Slave River, and in Great Slave Lake and some Yukon lakes. Levels of 2,3,7,8-TCDD are low (typically <1 $pg \cdot g^{-1}$) in comparison with levels in fish sampled near bleached kraft mills or to species in the lower Great Lakes.
- The geographical coverage of contaminant measurements is very good in the Yukon, where most major subsistence fisheries have been surveyed, but is much less detailed in NWT and northern Québec. Contaminant analysis of freshwater fish in the Nunavut region of NWT and the Nunavik area of northern Québec is limited mainly to arctic char and is insufficient to assess spatial trends. Further sampling of other subsistence fishes in Nunavut and Nunavik should be a priority in the future.

Overall Assessment of Contaminants in Terrestrial Environments

- PCBs and cadmium are the most prominent contaminants in terrestrial mammals and waterfowl species that have been analysed. Geographic coverage of major species such as caribou and mink (within NWT), as well as waterfowl and game birds, is quite complete. Contaminant data are available for a greater number of terrestrial mammals in the Yukon compared with NWT.

- Relatively high levels (10-60 $\mu g \cdot g^{-1}$) of cadmium are observed in kidney and liver of caribou from the Yukon, the NWT and northern Québec. Cadmium is higher in the western Arctic/Yukon herds. High levels of cadmium did not appear to be particularly associated with industry (mining) or populated areas.
- Current levels of radionuclides (^{137}Cs) in caribou muscle are 4- to more than 10-times lower than during the 1960s following atmospheric weapons testing. Levels of ^{137}Cs show a pronounced geographic trend with higher concentrations in northern Québec caribou herds. The levels of ^{137}Cs are much lower than observed in reindeer in northern Europe after the Chernobyl accident in 1986.
- Caribou receive relatively high doses of natural radiation relative to other mammals in the Canadian environment due mainly to ^{210}Po. Highest levels of ^{210}Pb were found in bone.
- PCB levels in terrestrial mammals show some clear geographic trends. Mink in southern NWT (the Great Slave Lake region) had higher PCB concentrations than those from the Mackenzie delta. A significant west to east increase in PCDD/Fs, ΣPCBs, HCB and ΣHCH was found in caribou with the highest mean levels in Cape Dorset and Lake Harbour herds and the lowest in the Inuvik herd.
- ΣPCB and ΣDDT were higher in piscivorous and molluscivorous birds from the eastern Arctic (Hudson Bay, N. Québec and Baffin Island) than in comparable species in the western Arctic. In the case of birds, the east-west trends in OCs may reflect migratory patterns and winter feeding locations rather than regional contamination differences.
- Levels of PCDDs and PCDFs in caribou were very low (<1 $pg \cdot g^{-1}$ for total PCDD/Fs at most sites). Most of the TCDD TEQs in caribou were attributable to non-ortho and mono-ortho PCB congeners. The TCDD TEQ levels observed in caribou fat can probably be considered to be background concentrations because they are as low or lower than those reported in fat of domestic animals in Canada.
- Significant contamination by PCBs and lead of soils and vascular plants is observed in the immediate vicinity and within a 20 km radius of abandoned and recently active DEW line sites in the Canadian Arctic.
- There is evidence for transfer of PCBs from plants to lemmings at former DEW line radar sites. This raises the possibility that some terrestrial mammals and birds could be contaminated because of feeding, even infrequently, on resident plants or animals at these locations.
- There was no evidence that large mammals such as caribou living in the general area of the DEW line sites had elevated levels of PCBs, however, it should be noted that the studies were not designed specifically to address this question.

Overall Assessment of Contaminants in the Marine Environment

- Geographic coverage of levels of persistent OCs and metals in marine mammal and seabird populations in the Canadian Arctic is very good. Most stocks/populations of major species (e.g. beluga, ringed seals, walrus, polar bears, thick-billed murres, glaucous gull) have been studied. Little or no data is available on contaminants in minor species such as harbour seals, harp seals.
- There is much less information on contaminant levels and very limited geographic coverage for marine fish and invertebrates.
- There is evidence of PCB and DDT contamination of the nearshore marine environments at some locations in the Canadian Arctic due to pollution from military radar facilities and other activity. The detailed study of PCBs in nearshore areas of Cambridge Bay illustrates how PCBs can be transported from dump sites to the immediate marine environment. But these studies also show that the PCB contamination is quite localized when considered on a broad regional scale.
- The major OC contaminants in marine biota were toxaphene and PCBs. DDT and chlordane related contaminants are also important especially in polar bears and seals. Toxaphene predominates in lower food web organisms and does not biomagnify to the same extent as PCBs or some chlordane components.
- Slightly higher concentrations of PCBs are observed in ringed seals and polar bears from Hudson Bay compared to populations in the western or central Arctic. These west to east spatial trends are difficult to identify within the Canadian Arctic, but become obvious when examining the circumpolar data for these species. This trend parallels observations for PCBs and other OCs in caribou and waterfowl.
- Glaucous gulls from Prince Leopold Island and Brown Island in the central Arctic had much higher OC levels (by 3 to 10 times) than populations from the western Arctic. Different feeding habits on the breeding grounds as well as winter feeding areas for the eastern and western glaucous gulls are the possible explanations for these differences.
- The remarkable variation of OCs in walrus, in which about 35% of the 53 samples analysed had elevated concentrations of PCBs (<1 $\mu g \cdot g^{-1}$), provides an example of the influence of diet on contaminant levels in top predators. The reason

for the elevated levels is almost certainly from predation on seals. Local contamination was ruled out because levels of all OCs were elevated in each animal from Inukjuak, and elevated levels were also found in animals from other areas of the eastern Canadian Arctic.

- Contamination of marine biota by heavy metals shows consistent large regional differences. Polar bears, ringed seals and beluga all have higher mercury contamination in the western Arctic. In polar bears, levels of mercury and selenium were highest in samples from areas bordering the Beaufort Sea. High mercury appears to be related to elevated natural levels associated with the sedimentary geology of the western Arctic.
- The spatial trend for cadmium was opposite to that for mercury, i.e. higher in the eastern Arctic than the western Arctic for belugas and the rate of accumulation of cadmium in the liver was higher in the eastern Arctic. Again the geographical trend is probably related to the different mineralogy of the eastern Arctic.
- Cadmium levels in kidney and liver of Canadian arctic marine mammals are as high or higher than those observed in the same or similar species in more temperate waters, but are similar to concentrations found in some terrestrial animals such as caribou and moose. Cadmium concentrations are also elevated in muscle of beluga, ringed seals and adult sea birds.
- Relatively high levels of mercury and methylmercury are found in skin or muktuk of beluga. Approximately 20% of the total mercury in the skin is lost annually through molting.
- Additional studies of metal and OC contaminants in muktuk need to be made in eastern Canadian marine populations because information is limited. These studies should take into account any variations, between individuals or communities, in methods of preparing muktuk, which might affect fat content of this food.
- Comparisons with other locations: PCB concentrations in juvenile ringed seals (1 year old) sampled in the 1980s from the Baltic averaged 17 $\mu g \cdot g^{-1}$ — about 10 to 20 times higher than levels in ringed seals from the Canadian Arctic. Male beluga from southern Hudson Bay (Sanikiluaq) sampled in 1994 had mean PCB concentrations of 6.8 $\mu g \cdot g^{-1}$ compared with levels of 79.2 $\mu g \cdot g^{-1}$ in male beluga sampled in 1993-94 from the St. Lawrence estuary. Belugas from the St. Lawrence estuary had higher lead and mercury concentrations in kidney and liver tissues than arctic beluga.

Temporal Trends

- Long-term studies (20 years) of OCs in ringed seal blubber at Holman Island and in sea birds at Prince Leopold Island in Lancaster Sound show declining concentrations from the 1970s to 1980s then a more recent leveling-off (mid-1980s to mid-1990s). The decline and leveling-off in concentrations of ΣDDT and PCBs in marine biota are consistent with the general trend of OC concentrations that has been observed in marine mammals and sea birds from eastern Canadian waters and in lake trout in Lake Ontario.
- The decline of ΣDDT in peregrine falcons was greater than observed for arctic sea birds. It may reflect declining concentrations in their wintering grounds. Recent reviews have shown a clear trend of decreasing PCB and OC pesticide contamination of the American peregrine falcon from the late 1960s to the 1990s.
- There is much less information on temporal trends of other OCs (such as toxaphene, chlordane, and chlorobenzenes) in marine biota. Limited data for the 1980s to early 1990s suggest that there has been no significant decline in the concentrations of these other OCs in marine mammals or sea birds during this period.
- Higher mean concentrations and higher rates of accumulation of mercury were found in ringed seals and beluga in more recent (1993–94) samples than in earlier collections (1981–83 in eastern Arctic, 1972–73 in western Arctic). The results are consistent with observations of increasing anthropogenic mercury deposition in lake and marine sediments (especially in the eastern Arctic).
- Cadmium concentrations in the same animals (eastern Arctic only) showed no change over a 10-year period.
- No temporal trend data are available for metals in sea birds or polar bears or other Arctic marine biota other than ringed seals and beluga.
- There is very limited temporal trend information on OCs or metals in the terrestrial and freshwater environments. For example, no temporal trend information for OCs or metals appears to be available for caribou or mink.
- Declining concentrations of toxaphene and other OCs were observed in burbot liver from the Mackenzie River at Fort Good Hope. Comparison of OC concentrations in round and lake whitefish from Lake Laberge over an 18-year interval showed a significant decline in ΣDDT, dieldrin and ΣHCH but increases in ΣPCBs, ΣCHL and toxaphene.
- At present, the temporal trend data are too limited to be strongly predictive for most OCs and metals because they are based on two or at most three sampling times. By comparison, temporal trend data for contaminants in Lake Ontario lake trout and in various species from the Baltic and from Lake Storinveld in northern Sweden are available yearly for a 15- to 20-year period.

- There is clearly a need for well-designed temporal trend studies utilizing tissues from tissue banks created by the Northern Contaminants Program at selected, well-characterized locations.

Biological Effects

- With the possible exception of peregrine falcons, contaminant levels or biochemical indicators of effects have not been linked to effects on arctic animals at the individual or population level.
- The species with the most significant risk of exposure to PCBs and OC pesticides may be the polar bear which appears to have elevated mixed function oxidase (MFO) activity (a liver detoxification enzyme).
- Similar positive correlations of MFO activity with non-ortho PCBs levels were observed in beluga that were starving due to ice entrapment. Arctic animals with relatively low levels of contaminants compared with temperate species may be vulnerable if they have to mobilize lipid deposits during fasting or starvation.
- Concentrations of TCDD TEQs in arctic ringed seal or beluga blubber are three to five times lower than those associated with impaired immune function (depressed leucocyte activity) in harbour seals.
- Concentrations of organochlorine contaminants in arctic beluga such as PCBs are 10- to 20-fold lower than in St. Lawrence beluga where preliminary evidence of a link for immune system dysfunction due to high PCB exposure (relative to Hudson Bay animals as controls) has been developed.
- Concentrations of PCBs associated with poor reproductive success of captive harbour seals are also about 10- to 20-fold higher than concentrations in blubber lipids of ringed seals.
- Females and their offspring may be most vulnerable during mobilization of these lipophilic contaminants for lactation because it occurs at a crucial point in the growth and development of the young. Overall, the MFO enzyme data for polar bear and beluga suggest that even the relatively low levels of contaminants present in arctic animals may not be without biological effects, especially during years of poor feeding.
- Other studies on fish and ringed seals have not shown the strong correlations between MFO enzyme activity and PCB levels, nor have they shown the presence of effects related to toxaphene exposure. As is usually the case with arctic animals, the lack of experimental dosage/response data continues to limit the ability to interpret concentrations observed in the animals.
- Arctic animals have relatively high body burdens of heavy metals and radionuclides compared to similar or related species in temperate regions. But they also may be adapted to relatively high exposure because of the importance of natural sources of these contaminants.
- Potential effects of the high doses of metals such as cadmium on caribou, beluga and narwhal are not clear. Cadmium was found to be predominantly associated with metallothionein in narwhal liver, suggesting it was in a relatively nontoxic form. The strong correlations of metallothionein in sea birds and marine mammals show that they are responding to heavy metal exposure.
- An evaluation using criteria established to protect fish-eating wildlife suggests that there is not a large margin of safety for Arctic marine or freshwater piscivores, especially if TCDD TEQs are used for the assessment. Using these criteria, carnivores such as polar bears would be at risk due to consumption of ringed seal tissues.

CHAPTER 4
Human Health

- Traditional foods are widely consumed within northern communities, the key food resources being large ungulates, marine mammals and fish. Consequently, potential exposure of Aboriginal Northerners to contaminants in country food is a widespread and typical condition right across the Canadian Arctic, including northern Québec.
- Traditional foods are an integral component to good health among Aboriginal people. Reduced country food consumption in northern Native populations has been associated with decreased physical activity, obesity, dental caries, anemia, lowered resistance to infection, and diabetes. Traditional food is as much the substance of Aboriginal Northerners' social well-being as it is that of their physical health. Harvesting, sharing, processing, and consuming traditional food is an important sociocultural process. Consequently, the contamination of country food raises problems which go far beyond the usual confines of public health, which cannot be resolved by health advisories or food substitutions.
- The geographic and socio-economic location of Aboriginal Northerners makes them particularly susceptible to the negative implications of country food contamination. Country food is economically essential for most Aboriginal Northerners.

Human Contaminant Exposure and Risk Assessment

Organochlorines

Levels of organochlorine contaminants in human tissue in the Arctic are considerably higher than those in southern Canadians, reflecting the greater consumption of high trophic level species for food.

- The OCs of primary health concern at this time for Inuit adults are PCBs, toxaphene and chlordane. Exposures in the eastern region are higher than in the western region. For Dene/Métis, exposure to OCs is in general below a level of concern. However, levels of chlordane and toxaphene exposure are elevated in some individuals and are a cause for concern if individual exposures are elevated on a regular basis.
- The developing fetus and breast-fed infant are likely to be more sensitive to the effects of OCs than adults and are the age group at greatest risk in the Arctic. Fetus/infant intakes of dioxins and furans, PCBs, toxaphene and HCB through human milk/cord blood are of primary concern. The concentrations of organochlorines in mothers milk of Inuit from Nunavik, northern Quebec are 2 to 10 times higher than the levels in a southern non-Aboriginal population.
- A proportion of Inuit newborn cord blood samples exceed the level of concern for PCBs. The Inuit cord blood samples from Nunavik have PCB and DDE levels that are double those seen in NWT Inuit. The difference between Inuit from northern Québec and Inuit from the western NWT deserves further study and may be due to amount and type of country food consumption or differences in the sample population (e.g. age of mothers, number of children or number of children breastfed). Cord blood samples from Inuit populations have higher contaminant concentrations and a greater proportion of detected analytes of almost all OCs than in Dene/Métis or non-aboriginal populations. This is not unexpected as Inuit consume more marine based traditional foods which contain higher OC levels.
- The human health risks of exposure to OCs are uncertain. Research associates dioxin, furan and PCB levels in human milk with subtle immunological and neurodevelopmental disorders.
- In consideration of the uncertainty of the effects of OCs, the extensive knowledge of the benefits of breast-feeding are a strong rationale for Inuit women to continue to nurse unless told otherwise by their health care provider. There are many recognized advantages to breast-feeding to infants and to mothers, including improved nutrition, increased resistance to infection, protection against allergy, better parent-child relationships, and possibly a degree of protection of the mother against breast cancer. In the Arctic, alternatives to breast-feeding, such as infant formula, can be difficult to obtain (due to availability and affordability) and can pose difficulties with respect to the maintenance of hygiene in cases where the water supply is compromised.
- As a comparison to the Canadian studies, elevated concentrations of some OCs have also been detected in fat tissue from Greenland Inuit. Levels of 2,3,7,8-TCDD TEQs (dioxins/furans/coplanar PCBs) in human milk samples from the Netherlands were found to be very similar to the levels in Inuit.

PAHs

- Despite considerable potential dietary exposure to PAHs in smoked food, there is little risk to health from this route of intake.

Metals

Lead

- Current levels of lead in the Arctic do not pose a significant threat to health and, based on declining emissions of lead globally, are not likely to pose a threat to health. Many adult and infant blood lead levels are below Canadian and WHO guidance values. However, because of the toxic effects of lead on children, cord blood and infant blood monitoring should continue to ensure local or regional lead levels are not increasing. It is worth noting that the Nunavik value has been shown to be in the range of cord blood lead associated with the threshold for premature labour.

Cadmium

- Several studies have indicated cadmium intakes by non-smokers in the Arctic, whether Inuit or Dene/Métis, are low and similar to intakes reported in Southern Canada. One study has shown some Inuit may exceed the TDI for cadmium. Cadmium levels in most of the traditional foods are fairly low. Therefore, the major route of cadmium exposure for most people is through cigarette smoking. Organ meats (liver, kidney) can contain significant amounts of cadmium. Therefore oral exposure becomes substantial for people who consume organ meats frequently (once a week year round). They may have higher risk of chronic cadmium toxicity, particularly if they also smoke. A recent risk assessment for cadmium indicates that some people, especially the elderly and diabetics, may be affected at lower levels than those previously considered safe.
- Smokers have 20 to 30 higher mean levels of blood cadmium than non-smokers regardless of where they live in Canada or the Arctic. These intake levels exceed the current WHO TDI value several fold and are not related to consumption of traditional food.

Mercury

- Methylmercury is a potent neurotoxin and the most toxic form of mercury in the environment. Human exposure in the Arctic is almost exclusively through food consumption, especially fish and marine mammals. Blood mercury levels are highest in the north and eastern Inuit communities. Blood mercury levels are higher for Inuit than Dene.

- A number of blood mercury measurements from native peoples (adults and newborns) reported from 1970 to 1992 are above the 5% risk range proposed by the World Health Organization for onset of neurological parasthesia or neonatal neurological damage. Recently data has become available under the AES program which indicates that some Arctic aboriginal communities may have lower levels of mercury.
- Continued hair monitoring and community-by-community dietary survey results will guide communities in their decisions on what, if any, selective food intake reductions are warranted.

Radionuclides

- The amounts of radionuclides in the Arctic environment are generally about the same as, or lower than, levels found in the temperate zone. Of all radionuclides, natural and of anthropogenic origin, ^{210}Pb and ^{210}Po (which are natural) make the greatest contribution to human radiation doses in the Arctic. Both occur in nature as airborne particles and tend to settle out on vegetation (i.e. lichens) and get eaten by caribou. Residents in Arctic communities may be receiving up to 10 mSv per year compared to a normal background of about 2 mSv. This has likely been occurring in the Arctic for several thousand years. Of the anthropogenic radionuclides, the two main isotopes of radiocesium (^{137}Cs and ^{134}Cs) are considered to be of greatest concern in the Arctic environment. Levels of radiocesium have gone down from about 450 Bq·kg^{-1} in 1965 to roughly 10 Bq·kg^{-1} in 1990.

Assessing and Managing Risk

- Risk determination for contaminants in country food involves a consideration of the type and amount of food consumed and the sociocultural, nutritional, economic, and spiritual benefits associated with country foods. Risk management decisions must involve the community and must take all aspects into account to arrive at a option that will be the most protective and least detrimental to the communities.
- Advisories related to country food if not released with proper communication and consultation can result in individual estimations of risk which are often based on untested assumptions and are often wrong, leading to harmful and undesirable social and economic results. This may result in the abandonment of subsistence to such a degree that the social integrity of the community is seriously compromised. As such, risk management decisions must be carefully considered and must be implemented in ways that minimize the risk of adverse effects. They should also always consider options which minimize the extent to which the nutritional and sociocultural aspects of aboriginal societies, of which country foods play a significant role, are compromised.
- Transfer of accurate and complete information via good communication may help limit the social and cultural effects resulting from the presence of contaminants in traditional food. In Arctic communities, communication is most effective when it is interpersonal and face to face, the preferred mode of communication of most Aboriginal people. It should be a two-way flow of information where the opportunity for feedback is maximized. The best studies and the best solutions to local contaminant problems are developed with and by the community.
- Risk management decisions must continue to be developed with communities to reduce exposures and to sustain traditional ways, especially the consumption of traditional food. Regardless of the decision taken, some health risks associated with exposure to contaminants may remain. In the Arctic, these risks are outweighed by the benefits of continued consumption. This uncertainty over risks and benefits often poses a large and confusing public, moral and political dilemma.

CHAPTER 5

From Research to Action — Using the Program Results

- In the short term, NCP results should provide people with an understanding of the issue of contaminants in the Arctic. In the long-term, it is hoped that NCP results will contribute to political actions that will ultimately result in significant reductions in the quantities of contaminants reaching the Arctic.
- Much of the strength of the NCP is derived from the partnership approach that forms the basis of its management process. The NCP is managed by DIAND on a partnership basis with a series of interrelated committees: the AES Partners Committee, the Science Managers Committee on Contaminants in Northern Ecosystems and Native Diets, the Technical Committee on Contaminants in Northern Ecosystems and Native Diets, the Yukon Contaminants Committee, the NWT Policy Advisory and Technical Committees on Arctic Contaminants.
- The Aboriginal Partners while increasing their internal capacity to address contaminants issues, also work to find ways of improving the NCP's communication and working relationship with its primary client group, consumers of country foods. This is an ongoing activity with one-to-one

contacts, workshops, newsletters and fostering of local contaminants committees, local environment committees, or regional program officers.

- The NCP is committed to optimizing the working relationships between NCP scientists and Northerners. The NCP Document "Researcher Guidelines for Planning Communications and Community Participation" sets out a series of requirements for researchers. Funding is contingent on demonstration of adherence to these guidelines. In the case of research on human health, the level of community participation in the planning and management of a project is necessarily very high.
- A number of NCP projects have incorporated traditional knowledge. There is a growing recognition that traditional knowledge can contribute to program planning.
- The NCP has, under the leadership of the northern Aboriginal organizations, increased its emphasis on communicating program results to Northerners and improving the two-way exchange of information between the program and people in the communities. Activities have focused on building capacity for informed decision-making and participation in research at the community level. Feedback from Northerners has had an important influence on the evolution of NCP priorities and activities.
- In addition to responding to community level information, NCP results have been used to substantiate a need for action under domestic and international policy. Nationally, NCP data have strengthened, accelerated and substantiated domestic policies such as the federal governments Toxic Substances Management Policy and other legislation on toxic substances.
- The NCP has been a driving force in the call for international action to control releases of persistent organic pollutants (POPs). NCP data have substantiated the need for action on a global scale, and NCP participants have played a leadership role in forwarding the issue of POPs on the international agenda. Progress in obtaining controls is evident, with the commencement of negotiation of protocols on POPs and Heavy Metals under the United Nations Economic Commission for Europe's Convention on Long-Range Transboundary Air Pollutants and the resolution by the United Nations Environment Programme to begin negotiations on a global, legally binding instrument on POPs.

Chapter 1: Introduction

Canadian Arctic Contaminants
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CHAPTER 1 Introduction

Table of Contents

1.1 Contaminants in the Arctic – Reason for Concern

The Canadian Arctic and sub-Arctic regions comprise the area north of 60° latitude, extending from the Beaufort Sea in the west to Davis Strait in the east, and from north of Ellesmere Island in the north to Hudson Bay in the south (see map insert). This vast region, encompassing 40% of Canada's land area, has one of the lowest population densities in the world — one person per 100 square kilometres (Bone 1992). The total population is approximately 85 000, including more than 40 000 Aboriginal people comprising Indian, Métis and Inuit (INAC 1996).

The Arctic was once considered pristine because of its remoteness and sparse population. However, over the last 50 years, the North has been exposed to contaminants originating from local sources (such as mining) and from distant industrial and agricultural regions of the world. These persistent contaminants have been detected throughout arctic ecosystems, including air, surface seawater, suspended sediments, snow (Barrie *et al.* 1992, Hargrave *et al.* 1988, Gregor *et al.* 1995), fish, marine mammals, sea birds (Muir *et al.* 1992, Lockhart *et al.* 1992), and terrestrial plants and animals (Thomas *et al.* 1992).

Contaminants such as persistent organic contaminants (POPs), heavy metals, and radionuclides enter the Arctic through long-range transport on air and water currents, with the atmosphere being the primary pathway. There are also some local sources of contaminants, such as metals and PCBs, that have been associated with mining and military sites in the Arctic. Many of the contaminants of concern in the Arctic are POP pesticides and industrial chemicals which are no longer used in Canada, and which have in many cases been banned or restricted for use in most of the developed world. They continue, however, to be used by many developing nations. Therefore reduction of POP contamination in the Canadian Arctic can only be achieved through action on a global scale.

Arctic ecosystems are particularly vulnerable to contaminants. The highly lipophilic and persistent nature of many of these chemicals results in their accumulation in the lipid-rich tissues of arctic organisms, reaching highest concentrations in the long-lived animals at the top of the food chain (Lockhart 1995, Barrie *et al.* 1992). Unexpectedly high levels have been found in some arctic biota, especially those such as fish, seals and whales, that are significant in the diets of many arctic residents (Kinloch *et al.* 1992). Chemical contamination of these traditionally harvested foods provides a critical path of contaminant transfer to human consumers (Kinloch *et al.* 1992), particularly northern Aboriginal peoples. Elevated levels of contaminants have been found in certain tissues and species of fish and wildlife in specific areas of the Northwest Territories (NWT) and the Yukon. Subsequent human health advisories that were issued resulted in the banning or restricted consumption of these traditional foods.

These findings raise concern because of the potential human health implications arising from the dependence of many northern Native peoples on traditionally harvested foods and their position as high food chain consumers. As much as 91% of native households in the NWT consume traditionally harvested meat and fish, and 22% have reported that all their meat and fish are obtained entirely through harvest (GNWT 1990). The replacement value of traditionally harvested foods and other renewable resources in the NWT is approximately $60 million per year (1990 dollars) (Brett Elkin, pers. comm.). There are tremendous nutritional, socio-economic and cultural impacts to northern Aboriginal peoples, if traditionally harvested foods are reduced or eliminated as a food source.

Because of their dietary dependence on arctic fish and wildlife, northern residents, especially northern Aboriginal peoples, are susceptible to the exposure to and potential adverse effects of contaminants. In instances of accidental or chronic exposure to high levels of some contaminants, toxicological effects including reproductive disorders, immune dysfunction, developmental abnormalities and reduced learning abilities, have been detected. This raises concern for the health of the arctic ecosystem and its peoples. However, the extent to which these and other effects result from the relatively low levels being found in Arctic biota, compared with other areas of Canada and the world, remains to be determined.

Scientists have been monitoring contaminants in the Arctic since the 1970s; however, concern over this issue heightened in the late 1980s with some disturbing findings:

- Increasing evidence that Aboriginal traditional foods contained contaminants raised questions about resulting contaminant levels in northern Aboriginal peoples. Thus, in 1985–87 PCBs were measured in the blood of Inuit from the community of Broughton Island, NWT, known to have a relatively high per capita intake of traditionally harvested foods. Results showed that blood PCBs exceeded tolerable levels, set by Health Canada, in 63% of the females and males under 15 years of age, in 39% of females aged 15–44, in 6% of males

15 years and older, and in 29% of women 45 years and older. This study also revealed elevated organic mercury levels in some individuals (Kinloch *et al.* 1992).

- A study in the late 1980s showed that PCB levels in the milk of Inuit women from the east coast of Hudson Bay in Nunavik (northern Québec) were approximately five times higher than in women of southern Canada. The elevated levels in these Inuit were attributed to their position at the top of the arctic marine food chain (Dewailly *et al.* 1989).

1.2 Contaminants of Concern, their Sources and Transport to the Arctic

There are three main classes of contaminants that are of concern for ecosystem and human health in the Arctic: persistent organic pollutants, heavy metals and radionuclides. More detailed information on these contaminants of concern can be found in Chapter 2, section 2.1.2 and Appendix I.

1.2.1 Persistent Organic Pollutants

Persistent organic pollutants (POPs) are organic compounds, including industrial chemicals and byproducts (e.g., PCBs, HCB, dioxins and furans), chlorinated pesticides (e.g., DDT, toxaphene, chlordane, HCH, aldrin and dieldrin) and polyaromatic hydrocarbons (PAHs). Local sources of these contaminants in the Arctic are rare (e.g., Distant Early Warning (DEW) line radar sites). The majority of POPs in the far north originate from hemispheric or even global sources, reaching the Arctic via the atmosphere, ocean currents and north-flowing rivers (Voldner and Li 1995, Iwata *et al.* 1993, Barrie *et al.* 1992, Hargrave *et al.* 1988). POPs have characteristics that favour their long-range transport, including low water solubility, the tendency to remain for long periods of time in the atmosphere in a gas phase or on particles, and high resistance to biological or chemical degradation (Barrie *et al.* 1992).

Many of the pesticides, such as toxaphene and HCH, that reach the north are applied to agricultural regions far removed from the Arctic. Though the uses of many pesticides with demonstrated toxic characteristics have been banned in North America and western Europe, they are still applied in less developed countries and continue to revolatilize from formerly treated soil in regions of banned use (Voldner and Li 1995, Barrie *et al.* 1992). Use of PCBs (e.g., flame retardants, dielectric fluids) has also been banned in western countries, however, these chemicals are still entering the Arctic as a result of continued use in some countries, revolatilization from areas of past use (Gregor *et al.* 1995, Voldner and Li 1995, Goldberg 1991), and abandoned military sites. Other industrial compounds reaching the Arctic include dioxins and furans, released during the chlorine bleaching process in the pulp and paper industry and during the production of pesticides.

Due to the chemical nature of POPs, they may be systematically transferred via the atmosphere from warmer to colder areas, accumulating in the Arctic. This process is called the "cold condensation" effect. This means that any release to the environment represents a potential global source, with the Arctic representing an ultimate sink for many atmospherically transported substances. Therefore, reduction of the POPs contamination in the Canadian Arctic can only be achieved through action on a global scale.

Levels of POPs build up over time in biota, a phenomenon known as bioaccumulation. Organochlorines also biomagnify, that is, they increase in concentration up the food chain (Muir *et al.* 1992). Once in the body, excretion of organochlorines is very limited, except through nursing female mammals to their offspring (Norheim *et al.* 1992). Levels in polar bear cubs have been found to be twice those in their mothers (Norstrom and Muir 1994). This raises concern about potential effects on young mammals in their vulnerable developmental stages.

There is limited data that directly links POPs to adverse effects in wildlife and humans. Evidence suggests that potential effects include reproductive and immune disorders, developmental abnormalities and cancer (Norstrom and Muir 1994, Barrie *et al.* 1992, Kinloch *et al.* 1992). The extent to which these and/or other effects result from the low levels being found in Arctic biota remains to be determined (Lockhart 1995).

1.2.2 Heavy Metals

The heavy metals of greatest concern in the Arctic are cadmium (Cd), mercury (Hg) and lead (Pb) (Pacyna 1995). Originating from both anthropogenic (e.g., mining, smelting, fossil fuel burning, waste incineration) and natural (local geology, volcanoes, degassing in aquatic environments) sources, these contaminants have been found in all components of the Arctic ecosystem (Barrie *et al.* 1992, Muir *et al.* 1992, Melnikov 1991). With chronic exposure, heavy metals bioaccumulate in biota. Potential effects of these metals include reproductive disorders, kidney and liver dysfunction, immune suppression and increased blood pressure (Scheuhammer 1987, Luoma *et al.* 1995).

The primary long-range transport pathway to the Arctic for heavy metals is the atmosphere. Global anthropogenic sources of mercury include: zinc, copper and lead smelting; burning of fossil fuels; chlorine production in chlor-alkali plants; production and use of agricultural pesticides; and waste incineration. Anthropogenic releases of cadmium include coal combustion, copper-nickel-zinc production, waste incineration and cement manufacture. Lead is emitted in automobile exhaust where leaded gasoline is still being used, and during non-ferrous metal production and fossil fuel combustion. Natural sources of cadmium and lead are volcanoes and windblown dust (Barrie *et al.* 1992, Nriagu and Pacyna 1988).

1.2.3 Radionuclides

A host of radionuclides have been found throughout the Canadian Arctic, including strontium 90 (^{90}Sr), cesium 137 (^{137}Cs), cesium 134 (^{134}Cs), plutonium 238 (^{238}Pu) and plutonium 239 and 240 (^{239,240}Pu) (Walton 1995, Barrie *et al.* 1992). Anthropogenic sources of radionuclides of significance to the Arctic environment include nuclear weapons testing, the Chernobyl accident, nuclear powered satellites burning up in the atmosphere upon re-entry, waste disposal into oceans, and discharges from nuclear power plants, waste management and reprocessing plants (NRPA 1995, Walton 1995, Barrie *et al.* 1992). The presence of radionuclide contaminants in the Arctic is a concern. Radionuclides are well-known for their carcinogenic effects. ^{137}Cs and ^{90}Sr are persistent, concentrate in food chains and have high radiotoxicity (Barrie *et al.* 1992).

Anthropogenic sources of radionuclides reach the Arctic via the atmosphere, ocean currents and north-flowing rivers (Walton 1995, Barrie *et al.* 1992). Atmospheric weapons testing represents the largest source of artificial radionuclides (including ^{90}Sr and ^{137}Cs) to the Arctic. Following restrictions on nuclear weapons testing, adopted in the 1960s, levels of radionuclides in Arctic air declined to near natural levels in the late 1980s (Barrie *et al.* 1992).

The 1986 explosion of the Chernobyl nuclear power plant released large amounts of fission products into the atmosphere. Those of health concern that reached the Arctic included ^{137}Cs and ^{134}Cs and to a lesser extent ^{90}Sr. The burn-up of nuclear satellites (e.g., the Soviet Cosmos 954 over the Northwest Territories in 1978) release a host of radionuclides such as plutonium 238, ^{90}Sr, and ^{137}Cs (Barrie *et al.* 1992).

There are significant natural geological sources of polonium, however, the arctic environment, particularly the terrestrial food chain, has been exposed to these levels for thousands of years.

1.3 Canadian Action

1.3.1 Canadian Action since the 1970s

Canada has been assessing the problem and taking action on contaminants in the Arctic for more than 25 years:

- early 1970s — specialists began measuring long-range transported atmospheric contaminants in Canada's North
- 1985 — the Department of Indian Affairs and Northern Development responded to Northerners' concerns about widespread ecosystem contamination and risks to human health by forming an intergovernmental Technical Committee on Contaminants in Northern Ecosystems and Native Diets
- 1989 — the Technical Committee on Arctic Contaminants expanded to include the five northern Aboriginal partners, and developed a five-year research and monitoring framework known as the Northern Contaminants Program's Strategic Action Plan
- 1991 — the Northern Contaminants Program became part of the Green Plan and a component of the new Arctic Environmental Strategy. In addition to the Technical Committee, a Science Managers Committee (which included the five Aboriginal partners) was struck to oversee the program's policy and funding issues
- 1991 — Canada signed the Declaration on the Protection of the Arctic Environment with the seven other Arctic circumpolar nations (Denmark/Greenland, Finland, Iceland, Norway, Russia, Sweden and the United States) in Rovaniemi, Finland which adopted the Arctic Environmental Protection Strategy (AEPS).
- 1991 — the Task Force on Persistent Organic Pollutants, led by Canada and Sweden, was established under the United Nations Economic Commission for Europe's Convention on Long-Range Transboundary Air Pollution
- 1992 — scientists published a benchmark study on the state of knowledge of contaminants in the Canadian Arctic in *The Science of the Total Environment*, an international research journal
- 1993 — Canada signed the Nuuk Declaration with the other Arctic circumpolar countries and reconfirmed its commitment to protection of the arctic environment as originally established in 1991 under the AEPS.
- 1993 — the Science Managers and Technical Committees on Northern Contaminants updated the five-year Strategic Action Plan
- 1994 — implementation of the program's Guidelines for Responsible Research, developed by the Aboriginal partners
- 1995 — Canada co-hosted an international experts meeting with the Republic of the Philippines on the global control of Persistent Organic Pollutants, in Vancouver, B.C.
- 1996 — Canada hosted the eight circumpolar nations under the Arctic Environmental Protection Strategy
- 1996 — the Arctic Council was established among the eight Arctic circumpolar countries in Ottawa. The Arctic Council institutionalizes the Arctic Environmental Protection Strategy
- 1997 — negotiations began, led by Canada, on a legally binding protocol to control POPs under the United Nations Economic Commission for Europe's Convention on Long-Range Transboundary Air Pollution
- 1997 — the Canadian Arctic Contaminants Assessment Report was published, summarizing all the information and data collected since 1991 and providing an evaluation of the state of contamination in the Canadian Arctic and sub-Arctic regions
- 1997 — a Ministerial Conference will take place in June in Tromsø, Norway involving the eight Arctic circumpolar countries as part of the newly formed Arctic Council. The Arctic Assessment Report and the State of the Arctic Environment Report produced by the eight Arctic countries under the Arctic Monitoring and Assessing Programme (AMAP) were released.

1.3.2 The Arctic Environmental Strategy/Northern Contaminants Program (AES/NCP)

In 1991, Canada established the Arctic Environmental Strategy's Northern Contaminants Program (NCP) in response to concerns over contaminants in northern traditionally harvested foods as a result of cooperative studies undertaken in the mid- to late-1980s by a number of federal and territorial government departments. The Program is assessing the risk to northern ecosystems and human health from the long-range transport of persistent pollutants into the Arctic. Therefore, the Program's key objective is "to reduce and, where possible, eliminate contaminants in traditionally harvested (country) foods while providing information that assists informed decision making by individuals and communities in their food use." Consequently, the Program has obtained information on the occurrence, abundance, sources, pathways, fate, spatial and temporal trends and effects of POPs, metals and radionuclides in the arctic ecosystem. As well, information is collected to assess the implications to human health. All this information is being used to communicate/educate northern communities regarding health aspects of their traditionally harvested food diets and to allow Canada to pursue action and negotiate contaminant controls in international fora.

The NCP is directed by a management committee and a technical committee, both of which are chaired by the Department of Indian Affairs and Northern Development, and which include representatives from the five northern Aboriginal Organizations (Inuit Tapirisat of Canada, Inuit Circumpolar Conference, Dene Nation, Métis Nation and the Council for Yukon First Nations), the Yukon and Northwest Territorial Governments, four federal departments (Environment, Fisheries and Oceans, Health, and Indian Affairs and Northern Development), the Centre for Indigenous Peoples' Nutrition and Environment (at McGill University) and the Québec Ministry of Community and Public Health. There are two regional contaminants committees in the Yukon and the Northwest Territories that support these national committees. A further description of these committees and the management structure of the NCP is provided in Chapter 5.

A state of knowledge report was produced and published in July 1992 in a special issue of the journal *Science of the Total Environment*, which served as a benchmark for contaminants information in the Canadian Arctic and which was used to rationalize the current NCP. The development of the strategic research plan and priorities for the Northern Contaminants Program were based on an ecosystem approach, focusing on three main categories: i) sources, pathways and fate; ii) ecosystem uptake and effects; and iii) human health. Other priority work areas that complement and/or are supported by the scientific research are a) communications, education and community-based strategies, and b) initiatives to promote international action and control of these persistent contaminants due to the transboundary nature of the issue. This report is structured around these main topics as outlined in Chapters 2–5.

Results from the NCP sampling programs have been used in human health risk assessments for contaminants in traditionally harvested food species. The use of NCP data in the health risk assessment process follows the protocol agreed upon by all NCP participants (federal, territorial and Aboriginal). Evaluation, validation and communication of health risk assessment results, including the integration of benefits considerations, is done by the two Territorial Contaminants Committees. Exposure, effects and nutrition data, as well as health risk assessments, are required in order to provide northerners with information for decisions regarding traditionally harvested food consumption. The information generated by the program is only useful to communities if it is available to them in a format that is relevant to their interests and needs. This holds true at every stage of the research process, from project planning and implementation to reporting of results and follow-up. Therefore, efforts to develop community dialogue processes and tools are increasing, as is recognition of the importance of community participation. A requirement for researchers to involve communities in their work was instituted in 1994 through the Guidelines for Responsible Research. Initiatives such as the Métis Nation's contaminants curriculum, Inuit Tapirisat of Canada's research on communicating about contaminants in traditionally harvested foods, and materials such as a layperson's overview booklet on Contaminants in Northern Canada, a variety of fact sheets and newsletters, and videos such as "Environmental Contaminants in the North" developed by GNWT Health and Social Services, form the beginning of an inventory of resources that support NCP communications. The strategic plan for coordination of NCP communication continues to evolve and form an important guide for much of the work to be undertaken in the coming years.

1.3.3 Quality Assurance

A quality assurance and quality control (QA) program was established to assess the performance of the laboratories carrying out contaminant analyses under the Northern Contaminants Program and the intercomparability of data. The majority of data included in this report have been analysed by laboratories that participated in these QA exercises and therefore these data are comparable. Details of this QA program are described in Appendix II. Any data in this report that originates from laboratories that have not participated in these QA activities should be viewed with caution.

1.4 International Action

Given the weight of evidence indicating that the majority of contaminants in the North originate from regions outside the Arctic, Canada recognizes that to address this issue requires international action. Thus, Canada is leading a number of international initiatives concerning contaminants. Findings from Canada's Northern Contaminants Program provide strong substantiation for Canada to take a lead role in seeking action to control these contaminants of concern in the international arena. The following provides a brief overview of some of the international initiatives with which Canada is involved. A more detailed review is found in Chapter 5.

1.4.1 Arctic Monitoring and Assessment Programme (AMAP)

At Rovaniemi, Finland in June 1991, Ministers from all eight Arctic countries signed the Declaration on the Protection of the Arctic Environment, which adopted the Arctic Environmental Protection Strategy (AEPS). The AEPS addresses the shared environmental concerns of the eight countries. It reviews the nature of Arctic environmental problems, assesses the existing institutional and legislative arrangements for dealing with them, and finally, identifies specific actions that should be implemented. One of these actions commits the eight governments to establish a comprehensive Arctic Monitoring and Assessment Programme (AMAP).

The mandate of AMAP is to monitor the levels of, and assess the effects of, anthropogenic pollutants in all components of the Arctic environment. It was envisaged that AMAP should as far as possible be built upon existing national and international programs, which are to be developed into a circumpolar framework. Initial priority is to be given on a circumpolar basis to POPs, selected heavy metals and radionuclides, and on a regional basis (Fennoscandia) to acidification.

To implement AMAP, a Working Group has been established comprising delegates from each of the participating nations (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden, United States), observing countries (Germany, Netherlands, Poland and the United Kingdom), and organizations of indigenous peoples (Inuit Circumpolar Conference, Nordic Saami Council and the Russian Association of Peoples of the North). The Working Group is chaired by Canada and vice-chaired by Sweden. Norway has provided a Secretariat, based in Oslo.

The Monitoring Programme for AMAP was completed in 1993, providing direction for monitoring activities for the Atmosphere, the Marine, Freshwater and Terrestrial environments, and Human Health. The requirements of the Monitoring Programme are fulfilled by the sum of the member countries' Implementation Plans. Canada's Implementation Plan is comprised largely of the projects being conducted under the Arctic Environmental Strategy's Northern Contaminants Program.

Output from AMAP will take the form of comprehensive assessment reports for the eight circumpolar Ministers, the first of which will be produced in June 1997. The first assessment will have two products: 1) the AMAP Assessment Report (AAR), which will be a comprehensive, technical and scientifically presented assessment of all validated data available on the status of the arctic environment relative to the AMAP mandate. This fully referenced document will provide the scientific basis and validation for the statements made in 2) the State of the Arctic Environment Report (SOAER), a summary of the AAR intended for an educated general public.

1.4.2 Canada/Russia Program on Scientific and Technical Cooperation in the Arctic and the North

In October 1991, an agreement was reached between Canada and Russia for cooperation on contaminants in the Arctic. Two major projects were achieved. Firstly, an air monitoring station was established at Dunay Island in the Russian Arctic, which is a replicate of the three existing stations in the Canadian Arctic. This new station has contributed to efforts to set up a circumpolar network for air monitoring. The second achievement was the exchange of environmental samples between Russian and Canadian laboratories for analytical intercalibration purposes. Future efforts will be directed toward establishing a second Russian Arctic air monitoring station on the coast west of the Ural mountains, and carrying out further analytical intercalibration exchanges with laboratories in Murmansk (Academy of Sciences), St. Petersburg (AARI), Moscow (IGCE), and possibly elsewhere.

1.4.3 United Nations Economic Commission for Europe (UN ECE) Convention on Long-Range Transboundary Air Pollution (LRTAP) Preparatory Working Groups on Persistent Organic Pollutants (POPs) and Heavy Metals

A Task Force on POPs led by Canada and Sweden was established in 1991 under the United Nations Economic Commission for Europe (UN ECE) Convention on Long-range Transboundary Air Pollution (LRTAP). The Convention is a legally binding instrument that has been ratified by 38 countries (including European countries, Canada and the US), and it currently has specific protocols addressing acid rain pollutants (sulphur and nitrogen oxides), as well as greenhouse gases such as volatile organic compounds. In April 1994, the POP Task Force completed its work and concluded that it had provided adequate substantiation to support the preparation of a POP protocol under the Convention. The UN ECE Working Groups on Technology and on Effects accepted these findings in July 1994. In December 1994, the Executive Body to the Convention concurred and established a new "Preparatory Working Group on POPs" led by Canada (through DIAND) with a mandate to assist the Working Group on Strategies with the preparation of a legally binding POP protocol. A decision was made by the Executive Body in December 1995 to develop a draft POPs protocol. Negotiation of the protocol begin in 1997 and it is expected that a legally binding POPs protocol will be in place by 1998. Canada, through a DIAND representative from the NCP, chairs the UN ECE Working Group on POPs, which is responsible for drafting negotiating text for the POPs protocol. Environment Canada is leading the negotiations for the Canadian government. All of the POPs of current concern in the Arctic are on the current "priority substance list," which currently includes 16 to 18 substances.

On a parallel track with the UN ECE POPs' efforts has been a similar initiative for heavy metals. A Task Force on heavy metals, led by the Czech Republic, was established in 1989 under the UN ECE LRTAP Convention. The Task Force completed its report in February 1994, concluding the need for a protocol to control the long-range transport and deposition of heavy metals. The UN ECE Working Groups on Technology and on Effects met in July 1994, noted concerns related in particular to the significance of natural sources and forwarded the Task Force report to the Executive Body. In December 1994, the Executive Body to the Convention established a new "Preparatory Working Group on Heavy Metals" with a mandate to assist the Working Group on Strategies with the preparation of a heavy metals protocol for submission to the UN ECE LRTAP Executive Body and to elaborate further the substantiation with respect to the relative strength of anthropogenic versus natural sources. This new Working Group is being led by Germany and negotiations will begin in 1997 to discuss a legally binding protocol for heavy metals.

1.4.4 Global Initiatives

An International Experts Meeting on POPs was held in Vancouver, Canada in June 1995. Canada co-hosted this meeting with the Republic of the Philippines, following an announcement to the United Nations Commission on Sustainable Development. The meeting produced a Conference Statement (agreed upon by participants from forty countries) that identified key components and opportunities for effective global action.

The Governing Council to the United Nations Environment Programme (UNEP) initiated an evaluation and assessment of environmental and socioeconomic considerations that should be considered in a future global legally binding instrument dealing with POPs. Preparatory work is being carried out by an *ad hoc* working group (chaired by Canada). A report to UNEP and the World Health Assembly was made in 1997 through the Intergovernmental Forum on Chemical Safety. Negotiations on a legally binding global agreement on POPs are expected to take place between 1998 and 2000, modelled after the UN ECE POPs protocol described above.

1.5 Importance of Conducting an Assessment of Contaminants in the Arctic

The purpose of producing this report is to provide an evaluation of the current state of contamination in the Canadian Arctic environment including the status and trends of contaminant levels and a comparison to other areas of the world. These trends help to determine the magnitude, geographic extent, source and duration of the problem.

After six years of intensive research and monitoring involving over 100 projects, a significant amount of information has been amassed under the Arctic Environmental Strategy's Northern Contaminants Program. This report combines all the information produced to date under this program, including the identification of contaminant sources, their transport pathways and distribution in the Arctic ecosystem, potential effects of contaminants on the health of the Arctic ecosystem including humans, assessment implications for human health in the context of northern populations utilizing local traditionally harvested foods, and the utilization of program results by northerners and the international community. Other sources of information that contributed to this assessment included all published and unpublished information related to contaminants in Canada's Arctic and Traditional Ecological Knowledge.

Through their subsistence lifestyles, Aboriginal hunters have developed a wealth of experience and understanding of the ecosystems in which they live. As hunters, they occupy the same ecosystem as their prey, and have traditionally spent many hours observing and talking about fish and wildlife. Experienced hunters are acute observers of changes in animals, and can recognize trends in the occurrence of some conditions over time. This field of knowledge is commonly known as Traditional Ecological Knowledge (TEK), and includes empirical knowledge gained through experience and observation. This is consistent with western science, where experience and observation form the basis of modern empirical research. Science differs only in that it has developed a method by which these experiences and observations can be systematically documented and verified. This report attempts to integrate these two systems to increase information on the occurrence of changes in fish and wildlife health. However, much remains to be done in this area, hopefully leading to increased trust and cooperation between Aboriginal hunters and scientists.

Over the last two years, more than 100 contributors have been involved in the writing of this report, representing federal, territorial and provincial departments, northern Aboriginal organizations, and universities. The companion document to this report entitled *Report on Contaminants in the Canadian North*, was prepared in consultation with the AES Aboriginal Partners and with input from community-level participants.

This report consists of the following stand alone chapters:

Chapter 2 – *Sources, Occurence and Pathways* presents results identifying contaminant sources, and their transport pathways and fate in Canada's Arctic environment.

Chapter 3 – *Ecosystem Uptake and Effects* assesses contaminants levels in fish and wildlife in the marine, freshwater and terrestrial arctic environments and their potential effects on the health of arctic ecosystems.

Chapter 4 – *Human Health* assesses the potential effects of contaminants on human health and assesses implications for human health in the context of northern populations utilizing traditionally harvested foods.

Chapter 5 – *From Research to Action: Using the Program's Results* outlines the various communication related activities that have been undertaken to provide information that assists informed decision making by individuals and communities in their food use, NCP management structures, other national activities related to arctic contaminants, and an outline of the progress that has been made under international action to control priority contaminants through agreements and cooperation with other countries and the circumpolar community.

Finally, Chapter 6 – *Conclusions* identifies the conclusions and key knowledge gaps that will assist in defining future research needs that meet the objectives of the Northern Contaminants Program.

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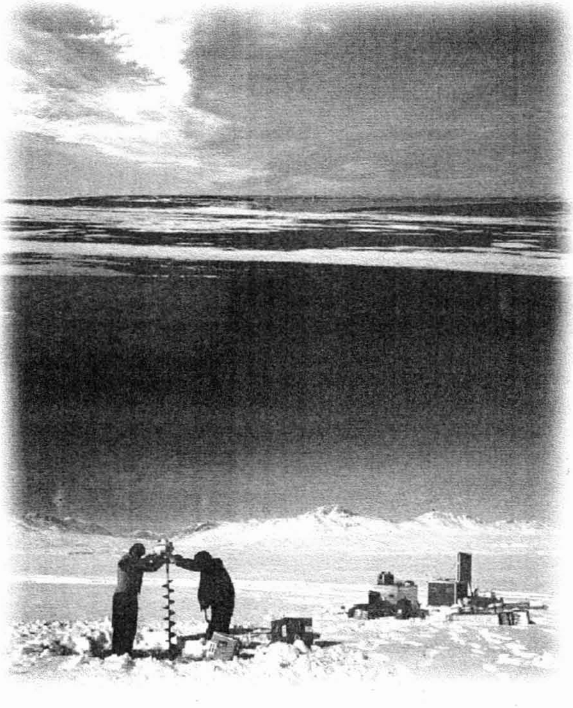
Chapter 2: Sources, Occurrence and Pathways



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CHAPTER 2 Sources, Occurrence and Pathways

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CHAPTER 2

Sources, Occurrence and Pathways

2.1 Introduction

The Arctic is no longer a pristine environment untouched by anthropogenic contaminants such as persistent organic substances, heavy metals, acids or radionuclides. On the contrary, it is a remote region that is surrounded by populated continents to which it is well connected by the currents of atmosphere and ocean. In 1991, in the formative years of the Canadian Northern Contaminants Program (NCP), a comprehensive review of the state of knowledge about contaminants in the north was undertaken. Sources, occurrence and pathways (Barrie *et al.* 1992), marine ecosystem contamination (Muir *et al.* 1992), terrestrial ecosystem contamination (Thomas *et al.* 1992), freshwater contamination (Lockhart *et al.* 1992) and risk/benefits of northern diets (Kinloch *et al.* 1992) were the topics discussed. The review laid the foundation for the NCP, a six-year research program from 1991 to 1997. A more recent review examined contaminants in the Arctic marine environment (Macdonald and Brewers 1996). This current paper summarizes progress up to 1996 in the understanding of sources, occurrence and pathways of contaminants in the Arctic and associated environmental issues.

2.1.1 Global Cycling and the Arctic System Pathways

Canada's Arctic and sub-Arctic areas occupy a vast region north of 60°N latitude, encompassing the Beaufort Sea in the west to Baffin Bay and Davis Strait in the east and from Ellesmere Island to Hudson Bay in the south (Figure 1.1). "The Arctic System," as defined here, is the area north of 60°N latitude (Figure 1.1). Previously, a multi-compartment box model (Figure 1.2) was introduced to provide a framework to describe the complex pathways linking the Arctic to the atmosphere and adjacent oceans (Barrie *et al.* 1992). This model schematically represented the atmosphere, land (terrestrial/freshwater), surface ocean, deep ocean and sediments. In addition, two interface-compartments have been added between the atmosphere and the Earth's surface representing: i) sea ice and, ii) snowpack or lake/ice. The Arctic Ocean has been separated into three compartments: two ocean layers, from

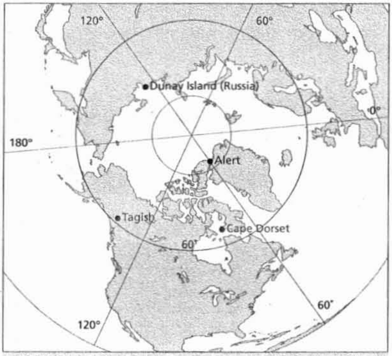


FIGURE 1.1

The "Arctic System" region as defined in this review is north of 60°N latitude. Also shown are the locations of toxics air monitoring sites (A–Alert, T–Tagish, C–Cape Dorset, and D–Dunay Island, Russia).

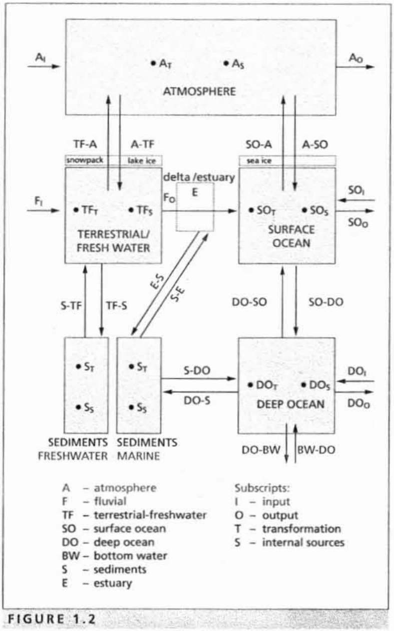


FIGURE 1.2

A box model of the "Arctic System" components and pathways of contaminants (see Table 1.1 for a summary of progress in understanding pathways depicted above).

approximately 0–200 m and from 200–800 m, and a sediment compartment. These compartments are based on hydrological differences in water layers, and the premise that the majority of marine life, and therefore biological processes such as biomagnification, occur in the top ocean layer.

Also shown in the schematic are the pathways of contaminants. Four types of pathways for contaminants are specified in Figure 1.2:

(i) transport into or out of the Arctic (subscripted with I and O) through air and water,
(ii) chemical or biological transformation within the system (subscript T),
(iii) anthropogenic sources of contaminants within the system (subscripted S), and
(iv) exchange between compartments within the system (hyphenated).

The exchange processes may involve transport, chemical reactions and physical processes as contaminants move across the interface. A summary of the current state of knowledge and how it has changed in five years (mainly as a result of the NCP) is provided in Table 1.1. Arrows are used to indicate those pathways that are better understood as a result of NCP research since 1991.

The influx of hemispherically or globally transported contaminants is not unique to the Canadian Arctic region. The terms "global distillation," "cold condensation" and "global fractionation," which refer to the way chemicals move to the poles and condense with varying effectiveness, represent a process that has been articulated by others (Rappe 1974, Dunbar 1973) and popularized by Goldberg (1975). The degree to which any one chemical begins and completes its journey and the time frame over which it occurs depends on its physico-chemical properties and the prevailing atmospheric conditions. There may be an erroneous impression that there is a tendency for bulk movement of discharged chemicals from low latitudes to high latitudes with

TABLE 1.1

Summary of level of knowledge in 1996 of pathways shown in Figure 1.2 for each group of contaminants. Changes in knowledge are denoted by an arrow.

Pathways	Industrial OCs⁺			PAHs*			Metals			Radionuclides		
	91		96	91		96	91		96	91		96
A_I	0	→	1	0	→	1			1	2		2
A_O	0	→	1	0	→	1	1		1	2		2
A_T	0	→	1	0	→	1	2		2	2		2
A_S	1		1	0	→	1	1		1	1		1
TF-A	0	→	1	0		0	0		0	1		1
A-TF	1	→	2	1	→	2	1		1	1		1
F_I	0	→	1	0		0	0		0	0		0
TF_T	0	→	1	0		0	1		1	2		2
TF_S	0	→	1	0		0	0	→	1	1		1
F_O	0	→	1	0	→	1	1		1	1		1
SO_I	1	→	2	0		0	1		1	1		1
SO_T	0	→	1	0		0	2		2	2		2
SO_S	n/a		n/a	0	→	1	n/a		n/a	0	→	2
SO_O	1	→	2	0		0	1		1	1		1
SO-A	0	→	1	0		0	0		0	n/a		n/a
A-SO	1	→	2	1		1	1		1	1		1
S0-DO	0	→	1	0	→	1	0	→	1	0	→	1
DO-SO	0	→	1	0	→	1	0	→	1	0	→	1
$D0_I$	0	→	1	0	→	1	1		1	0	→	1
DO_S	n/a			0		0	0	→	1	0	→	2
DO_T	0	→	1	0	→	1	2		2	2		2
DO_O	0	→	1	0		0	1		1	0	→	1
S-E	0	→	1	0	→	2	0		0	0		0
E-S	0	→	1	0	→	1	0	→	1	0	→	1
S-TF	0	→	1	0	→	1	0	→	1	1		1
TF-S	0	→	1	0		0	0	→	1	1		1
S-DO	0	→	1	0	→	1	0		0	0		0
DO-S	0	→	1	0	→	1	0	→	1	0	→	1
S_T	0	→	1	0	→	1	0	→	1	0	→	1
S_S	n/a		n/a	0	→	1	n/a		n/a	0	→	1

0 = essentially no measurements available and/or processes understood to make estimates
1 = some information available for crude estimates
2 = much information available for quantitative estimates
n/a not applicable, organochlorine pathway does not exist or is negligible
+ = organochlorines
** = polyaromatic hydrocarbons*

condensation of an appreciable fraction of the discharged material in the Arctic. This is analogous to the deposition of water in icefields at high latitudes, or the seasonal CO_2 "icecap" formation on Mars. In reality, there is never condensation of a pure chemical under saturation conditions, nor is a major fraction of the total emissions conveyed to the Arctic. However, the small fraction that is conveyed to the Arctic results in surprisingly high contaminant concentrations that can exceed prevailing levels in more temperate regions that are closer to discharges.

Evidence of global cycling of contaminants has been documented extensively in temperate latitudes. For example, the transport and deposition of pesticides, which have been discontinued in North America, have been documented in the Great Lakes region (Scholtz *et al.* 1993). Polychlorinated biphenyls (PCBs) have been detected in Lake Superior with the source determined to be atmospheric deposition of contaminants from distant sources (Swackhamer *et al.* 1988). However, there are several characteristics of the Arctic region that make the presence of these compounds of special concern:

i) these contaminants persist in the arctic environment for long periods of time,
ii) northern Aboriginal people depend on a high fat diet of country foods, which in turn tend to accumulate large quantities of some of these contaminants, and
iii) northern residents have not used or directly benefited from much of the activities associated with production and or increased use of these compounds.

- *From 1991 to 1996, significant progress has been made by the Northern Contaminants Research Program (NCP) in understanding pathways of contaminant movement in the Arctic environment. Advancement of knowledge has been made in 93% of possible pathways for OCs, 60% for PAHs, 32% for metals and 34% for radionuclides.*

2.1.2 Contaminants of Concern

Only in the last century have human activities significantly affected remote environments of the globe. These global impacts have been largely due to the increase in emissions to the atmosphere as a result of the industrial revolution. Nevertheless, the presence of many industrial contaminants in the Arctic and the Antarctic only became significant during the decade following the Second World War. First notice of anthropogenic contamination on a broad scale in the Arctic was taken in the 1950s by pilots reporting arctic haze (Mitchell 1956, Rahn and McCaffrey 1980). This contamination has since been fully recognized as a manifestation of long-range transport of pollutants to the Arctic. Arctic haze also plays a role in climate by altering the energy balance of the Arctic atmosphere and ocean (Blanchet 1991).

To appreciate the problems caused by contaminants requires an awareness of their distinctive characteristics. In general, OCs, polyaromatic hydrocarbons (PAHs) and heavy metals have been produced and released to the environment in large quantities. When released to the environment, they are long-lived and widely dispersed. OCs are distinct in that most are human-made and do not have natural sources. In the 1991 review, six classes of contaminants were discussed. In this review, we focus on only five classes, deleting acids from the original list (Table 1.2). Although important locally in other Arctic regions, acids consisting mainly of sulphur compounds are not present at sufficient levels in the Canadian Arctic to constitute a toxic threat (Barrie 1986, Barrie *et al.* 1992). However, they are of environmental importance to climate in the region.

2.1.2.1 Organochlorines (OCs)

Organochlorines (OCs) comprise compounds in groups 1 and 2 of Table 1.2. Since the 1970s, it has been recognized that they are reaching the Arctic and accumulating in ecosystems. Body burden measurements in marine mammals and fish have shown that arctic food chains contain relatively elevated concentrations of certain organic chemicals. Local or regional sources cannot explain the presence of many of the compounds, especially the OC pesticides that are used outside the Arctic.

One of the main groups of contaminants is the pesticides. The first generation of pesticides, based on arsenic, copper and sulphur, were predominant in the late 1800s to 1940s but were not widely used and consequently have not appeared as a global concern. However, the second generation of pesticides were OCs that have very different physico-chemical properties from the first generation pesticides. They are chemically persistent and semi-volatile. Semi-volatile means that they have a low vapour pressure that is sufficiently high to cause appreciable evaporation over a period of weeks to years which enables them to cycle between gaseous and condensed phases in the environment. The presence of chlorine in the molecule tends to increase persistence. As well, the low cost and efficacy of these compounds has resulted in their intensive use over a large part of the globe, including tropical regions, from the 1950s to until the present. Their use in warm climates enhanced their global mobility. While some use of these compounds continues, their production and use is in decline. New-generation (i.e. third-generation) pesticides, largely nitrogen- and phosphorus-based, differ from OC pesticides in that they: generally have a shorter environmental half-life; are likely to be used in greater quantities in the future; their toxic mode

TABLE 1.2

Arctic environmental contaminants of concern in 1996.

1. Chlorinated industrial organic compounds
    - Chlorobenzenes (CBZs)
    - Polychlorinated biphenyls (PCBs)
    - Dioxins/Furans (TCDD/Fs)
2. Organic Pesticides
    - Chlorobornanes (CHBs)
    - Hexachlorocyclohexanes (HCHs)
    - Chlordane
    - DDT/DDE
    - Aldrin/dieldrin
    - Endosulphan
    - Other current use pesticides (trifluralin, etc)
3. Polyaromatic hydrocarbons (PAHs)
    - e.g. benzo[a]pyrene (B[a]P)
4. Metals
    - Cadmium (Cd)
    - Lead (Pb)
    - Mercury (hg)
5. Radionuclides
    - Cesium (^{137}Cs)
    - Plutonium ($^{239,240}Pu$)
    - Iodine (^{129}I)
    - Strontium (^{90}Sr)

of action is more specific (i.e. more effect on target species than on the Arctic community); and their toxicity is better understood. PCBs are an example of an industrial OC. They were previously used as hydraulic and transformer fluids, but production and use has been largely discontinued in Western countries. However, dumping in the environment and their persistence makes them an ever-present chemical group. Dioxins and furans are another class of industrial OCs. They were never deliberately produced, but rather, formed as by-products in other processes (e.g., incineration). Widely released, they have toxicological effects in the environment at extremely low concentrations. Chlorobenzenes (CBZs) originate from chemical solvents and reagents and as by-products of hexachlorobenzene production and old electrode processes.

2.1.2.2 Polyaromatic Hydrocarbons (PAHs)

Polyaromatic hydrocarbons (PAHs) are widely distributed in the environment (structure shown in Figure 1.3) where they have the potential to form carcinogenic and mutagenic diols and epoxides that react with DNA (Zedeck 1980). They can be used as conservative tracers of atmospheric and oceanographic transportation of anthropogenic contaminants because of their relative environmental stability (McVeety and Hites 1988, Yunker *et al.* 1996). In contrast to the OCs, PAHs are produced in large quantities by natural processes as well as by anthropogenic activities. Natural sources include: natural fires (grass, trees, peat); natural losses or seeps of petroleum or coal deposits; diagenetic production in soils and sediments — bacteria working on complex organic carbon compounds — perylene is certainly a good marine example of this process; and aromatization of higher plant terpenes (i.e. retene, simonellite, pimanthrene and cadalene). PAHs are an important component of both crude and refined oil and are produced during the incomplete combustion of coal, wood, and oil (Daisey *et al.* 1981, Thomas *et al.* 1986, Steinhauer and Boehm 1992, Lockhart *et al.* 1992, Yunker *et al.* 1993). Slow thermal and bacterial degradation of plant and animal remains in lake and marine sediments is the source of highly alkylated PAHs that are found in crude and refined oil (Yunker and Macdonald 1995). These PAHs often have greater adverse effects on biota (Payne *et al.* 1988, Hellou *et al.* 1995). PAHs are semi-volatile and tend to become attached to particles, especially those of high molecular weight. Measurements of arctic air (Patton *et al.* 1991, Halsall *et al.* 1997), snow (Welch *et al.* 1991, Gregor *et al.* 1995b) and ice (Kawamura *et al.* 1994, Masclet and Hoyau 1994) have clearly shown that these combustion PAHs are transported by the atmosphere to the Arctic from temperate industrial regions of the globe. Release of anthropogenic PAHs has increased dramatically during the past century as a result of increased combustion of fossil fuels. In this century, shifts from coal to liquid fuels during the 1950s have probably reduced, or at least altered, PAH emissions. Ice core records on Ellesmere Island reflect this (see Section 2.5.2). At any given location, the PAH mixture originates from both natural and anthropogenic sources. In the environment, parent PAHs (PAHs with unsubstituted rings) are usually accompanied by a series of alkyl-substituted homologues. For example, alkyl-substituted derivatives of the parent PAH phenanthrene include compounds like pimanthrene (C_2, or two carbon substituents) and retene (C_4) (see Figure 1.3).

2.1.2.3 Metals

Metals present in the Earth's crust are mobilized by volcanoes, winds and a variety of anthropogenic applications (mining, smelting and waste incineration). Heavy metals of concern to the environment have natural sources but their abundance and distribution has been greatly impacted by anthropogenic activities. Estimates of global releases of a number of metals to the atmosphere from a variety of sources are presented in Table 1.3 (Nriagu 1991). Except for chromium and selenium, the anthropogenic component exceeds that from natural sources. For NCP metals of concern, the global anthropogenic emissions in the mid-1980s exceeded global natural emissions for Pb, Cd, and Hg by a factor of 28, 5.4 and 1.4, respectively (Table 1.3).

These metals are in a number of lists of metals of concern (e.g., Canada-US Great Lakes Water Quality Agreement, Annex 1). Cd is known to accumulate in the kidney and liver of terrestrial and marine mammals and other species (see Chapter 3). Mercury in its methylated form is well-known as a bioaccumulating compound (Wagemann *et al.* 1996).

Atmospheric Hg, mainly in the form of gas phase elemental Hg, is increasing globally at about 1% yr^{-1} (Fitzgerald 1995, EPRI 1994). It is also increasing in sediments laid down in high Arctic lakes during the post-industrial era (Lockhart *et al.* 1995). The main anthropogenic sources of Hg include: chlor-alkali plants, waste incineration, coal combustion and metal production. Natural emissions to the atmosphere from the Earth's crust can occur via the biosphere and oceans. Best estimates of the relative importance of anthropogenic sources to the global total input to the atmosphere ranges from 5% to 75% (EPRI 1994).

2.1.2.4 Anthropogenic Radionuclides

Although radionuclides are natural as well as anthropogenic in origin, their incidence in the environment increased through large scale releases from:

(i) development and testing of thermonuclear devices,
(ii) processing of nuclear fuels and
(iii) the improper disposal of nuclear reactors and spent fuels.

The significance of radioactivity is in its ability to damage DNA by ionizing radiation. From the radiological health perspective, ^{137}Cs is the most important anthropogenic radionuclide because it makes the greatest contribution to dose in the Canadian Arctic (Macdonald and Brewers 1996). Natural radionuclides (i.e. Radon decay products) contribute more than 90% to radiation dose for native northern residents; of the remaining 10%, more than half of the dose comes from ^{137}Cs and the remainder from all other anthropogenic radionuclides (Beak 1995). The predominant anthropogenic radionuclides measured in the Arctic are: ^{99}Tc, ^{129}I, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, and ^{241}Am. To these can be added short-lived fission and activation products, which are now mostly below normal limits of detection.

TABLE 1.3

Estimated natural and anthropogenic global emissions (t·a^{-1}) of trace metals to the atmosphere (Nriagu 1991). Those of concern in the NCP are highlighted.

	Natural	Anthropogenic	Anthropogenic: Natural
Antimony	2.6	3.5	1.3
Arsenic	12	19	1.6
Cadmium	**1.4**	**7.6**	**5.4**
Chromium	43	31	0.72
Copper	28	35	1.2
Lead	**12**	**332**	**28**
Mercury	**2.5**	**3.6**	**1.4**
Molybdenum	3	6.3	2.1
Nickel	29	52	1.8
Selenium	10	5.1	0.51
Vanadium	28	86	3.1
Zinc	45	132	2.9

Of the various known sources, atmospheric fallout and the European reprocessing plants have been the greatest contributors to anthropogenic radionuclides observed in the arctic environment. Uranium processing plant effluent that was released into European waters in the past from the UK and France is now radiologically inactive. The 1986 accident at Chernobyl did not distribute much radioactivity to the Arctic via the atmosphere, however, a large proportion of the radionuclides initially deposited in the Baltic Sea and North Sea will eventually reach the Arctic Ocean via oceanic currents. In 1993, the Yablokov report

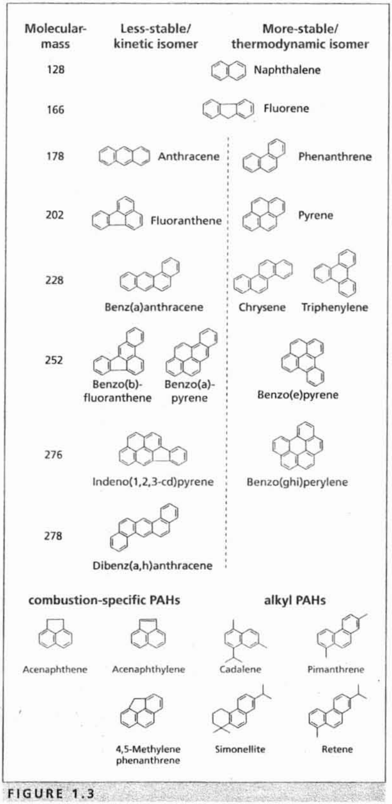


FIGURE 1.3

Structures of PAH compounds. Parent PAHs are shown separated into two groups to differentiate between less stable (kinetic — left-hand side) and more stable (thermodynamic — right-hand side) PAH isomers. Combustion-specific PAHs and higher plant terpenes are also shown.

(Yablokov *et al.* 1993, Mount *et al.* 1994) raised concerns about locally dumped radioactive wastes in the shallow continental shelf regions of the Russian Arctic. Studies show there is presently no indication of contamination and while radioactivity will inevitably be released as waste containment deteriorates, the threat would be on a local and regional scale. Currently, there is no indication of significant inputs of radionuclides to the Arctic Ocean from Russian rivers. From environmental and human perspectives, it should be emphasized that anthropogenic radionuclides must be viewed in the context of a natural background comprising cosmic radiation and radiation from naturally occurring radionuclides such as ^{210}Po, ^{232}Th and ^{222}Rn.

- *The following contaminants were the focus of research under the NCP: persistent organochlorines (OCs), polyaromatic hydrocarbons (PAHs), the metals lead (Pb), cadmium (Cd) and mercury (Hg) and radionuclides. Body burden measurements in marine mammals and fish have shown that arctic food chains contain relatively elevated concentrations of certain OCs. From the 1950s to the present, they have been intensively used over a large part of the globe. PAHs, metals and radionuclides have natural as well as anthropogenic sources. At any given location, the PAH mixture of chemicals found in the Arctic originates from anthropogenic (coal and oil combustion) and natural sources. For the metals of concern in the NCP, the ratio of global anthropogenic to natural emissions to the atmosphere in the mid 1980s are: 28:1 (Pb); 5.4:1 (Cd) and 1.4:1 (Hg). Atmospheric fallout and European reprocessing plants have been the greatest contributors to anthropogenic radionuclides observed in the arctic environment.*

2.1.3 Organization of this Chapter

Research material in this chapter is organized into five sections subsequent to this introduction. The multi-compartmental model of the northern ecosystem (Figure 1.2) is used as a conceptual framework for the organization of results.

First, Section 2.2, we discuss the sources of contaminants and their transport to the Arctic System (pathways A_L, A_S, F_L, TF_S, SO_L, SO_S, DO_L, DO_S), which is defined as the region north of 60°N latitude. Under sources, we discuss what has been learned in this program about global emissions to the environment that affect the Arctic, as well as insights gained about emissions within the Arctic. Transport to — and within — the arctic system involves atmospheric, riverine and marine currents. Because of the pervading influence of partitioning of contaminants between gas (atmosphere), liquid (rain, rivers, oceans) and solid phase (snow, snowpack, lake and marine ice, suspended particles, soil, vegetation) components of the Arctic System, a special section has been included on progress made in phase-partitioning properties of Arctic contaminants.

Next, in Section 2.3, we present results from research on the occurrence and internal pathways of contaminants in the compartments of the Arctic System (boxes in Figure 1.2). The main compartments studied are: atmosphere, terrestrial/freshwater, marine, and sediments. Important intermediate compartments that are seasonally very active, namely, snowpack/lake ice, sea ice, and delta-estuaries are also included.

Third, Section 2.4 reviews research results on the processes of inter-compartmental exchange (arrows between boxes in Figure 1.2). This section uses the concentrations reported in Section 2.3 and estimates of the flow of air, water or sediment between compartments (technically called "flux") to estimate these pathways. To some readers, the separation of concentration observations in the previous section from the flux calculations may be confusing. However, it is consistent with the system components and pathways detailed in Figure 1.2.

Fourth, Section 2.5 deals with contaminant trends in the Arctic. Much progress has been made in measuring temporal changes of contaminants in the Arctic, both within and outside the NCP. Glacial ice and freshwater sediments have yielded an historical record of OC, PAH and Hg trends that were not available previously. Current atmospheric observations, when coupled with earlier work, have also revealed important temporal trends. There is obviously a connection between contaminant occurrence data in Section 2.3 and trends data in Section 2.5.

Fifth, Section 2.6 is a special topics section. Created to address extant questions concerning northern contaminants, and anticipated future problems, this section synthesizes information presented in previous sections from an issues perspective.

Finally, Section 2.7 summarizes NCP research on the occurrence, pathways, and fate of contaminants in the Canadian Arctic. In addition, this section highlights the successes of the NCP and provides direction for future research endeavours.

2.2 Sources and Transport of Contaminants to the Arctic System

2.2.1 Circulation and Transport

2.2.1.1 Air

The atmospheric pathway of a contaminant is governed by the spatial distribution of its sources globally, air circulation patterns, exchange with the Earth's surface, and chemical transformation. Atmospheric winds deliver contaminants to the north. The lower tropospheric circulation of the northern polar region (Figure 2.1a) is dominated in winter (January) by high pressure systems over the continents and low pressure systems over the northern Pacific and Atlantic Oceans. In particular, the intense Siberian high pressure cell tends to force air on its western side northward into the Arctic. The high pressure ridge over North America tends to drive air out of the Arctic southward. The mean flow in winter is therefore out of Eurasia into the Arctic and out of the Arctic into southern North America. Of course, some air is also exchanged with the south when low pressure vortices along the Arctic front mix warm southern air with cold northern air in a large-scale turbulent eddy.

In summer (Figure 2.1b), the continental high pressure cells disappear and the oceanic low pressure cells weaken, particularly in the north Pacific. Northward transport from mid-latitudes decreases accordingly. As altitude increases, the mean circulation in the lower atmosphere depicted in Figure 2.1 gives way to a more circular clockwise flow around the pole prompted by polar low pressures prominent in both summer and winter. The winter Siberian high pressure cell disappears at an altitude of 3 km and higher.

Three types of flow regimes are involved in the exchange of mass or heat between polar and extrapolar regions: mean meridional circulation (MMC), standing eddies (SE), and transient eddies (TE). To illustrate qualitatively that these can all contribute substantially to north-south mass exchange, consider the seasonal distribution of energy exchange for each flow regime calculated by Nakamura and Oort (1988) for the Arctic and Antarctic (Figure 2.2). Exchange of heat is least in summer and greatest in winter. Furthermore, standing eddies such as the Siberian high pressure cell (marked by a source of surface winds in Figure 2.1a) tend to play a more prominent role in

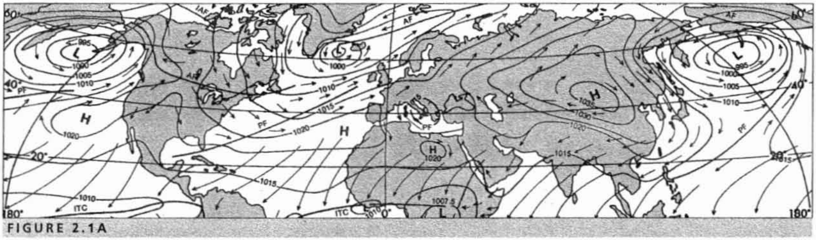


FIGURE 2.1A

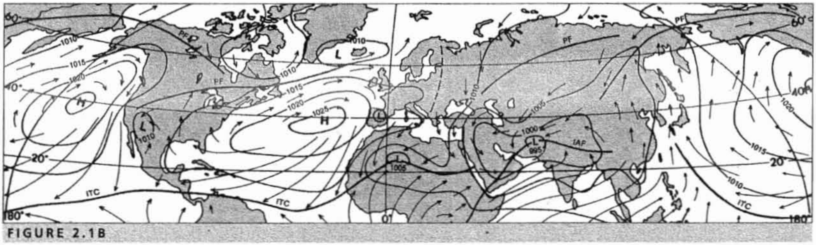


FIGURE 2.1B

Mean surface pressure maps in the Northern Hemisphere (from Liljequist 1970) representing the mean circulation of the lower atmosphere in (A) January and (B) July. Thin solid lines show surface pressure in kPa. Long arrows indicate steady winds. Short arrows indicate unsteady winds. Long lines show frontal zones and convergence zones.

the northern hemispheric winter than in summer or than in the Antarctic at any time of year.

Total eddy exchange (SE+TE) is stronger than mean meridional circulation (MMC) at all times in the Northern Hemisphere, but not during winter in the Southern Hemisphere when MMC dominates.

Exchange of contaminants between the atmosphere and the Earth's surface is complex involving dry deposition of particles, gas exchange and precipitation scavenging. Removal by precipitation depends on gas/particle partitioning, solubility in water and adsorption to snow. Also, for certain contaminants, B[a]P for example, chemical transformation in the atmosphere by reaction with OH, NO_3, Br or Cl is important in some cases (e.g. for B[a]P).

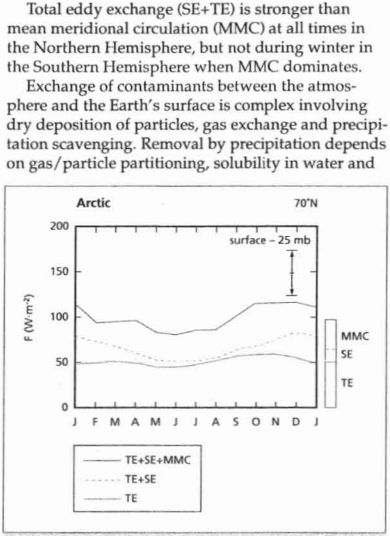


FIGURE 2.2A

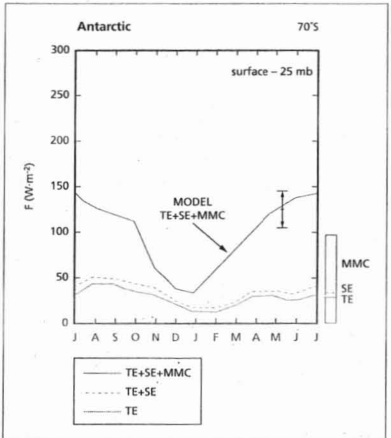


FIGURE 2.2B

Annual variation in poleward energy flux (F) for the atmosphere from the earth's surface to 25 hPa as effected by transient eddies (TE), standing eddies (SE) and mean meridional circulation (MMC) for (A) the Arctic, and for (B) the Antarctic (Nakamura and Oort 1988).

Atmospheric transport pathways can be subdivided into two types: "one-hop" pathways and "multi-hop" pathways. If a compound is emitted to the atmosphere, transported and deposited to the Earth's surface, never to return to the atmosphere (i.e. "one-hop"), the source region of this contaminant could be determined by knowledge of its initial source distribution, atmospheric circulation and its lifetime in the atmosphere (governed by removal processes). One-hop contaminants include acids, heavy metals (with the exception of mercury), involatile OCs (e.g. DDT) and less volatile PAHs (e.g. B[a]P). The pathways of these constituents follow that of arctic haze from mid-latitudinal sources into the Arctic (Barrie 1986, 1993, 1995). They are marked by: (i) longer atmospheric residence times in winter (~20–30 days) when precipitation is at a minimum than in summer (~3–7 days) when precipitation is a maximum, and (ii) stronger south-to-north transport into the Arctic from Eurasia in winter than in summer (Figure 2.1). However, if a compound re-enters the atmosphere after initial deposition to the Earth's surface, it can move through the environment by taking multiple atmospheric hops. Processes by which this can occur include volatilization from the Earth's surface under warmer temperatures than when the contaminant was deposited, sudden exposure of ocean water to the atmosphere after being covered by ice, or resuspension of dust or snow by winds. In this case, defining the source region of a contaminant is not simply a function of the processes of atmospheric transport, circulation and removal, but also of surface processes that control its re-entry into the atmosphere after initial deposition. Most OCs and many PAHs fall under the "multi-hop" contaminants. Complex multi-compartmental environmental models that account for all system components (Figure 1.2) are needed to understand the complex pathways of these compounds. Models developed under the NCP and elsewhere are reviewed in Section 2.2.4.

- *Contaminants can be subdivided into two types according to their atmospheric pathway: "one-hop" and "multi-hop" compounds. If a substance is emitted to the atmosphere, transported and deposited to the surface never to return to the atmosphere, it is a "one-hop" compound. Its source region for the Arctic is defined by its lifetime in the atmosphere (governed by removal processes), its distribution of emissions to the global atmosphere, and atmospheric circulation. One-hop contaminants include heavy metals (except Hg) as well as the relatively involatile OCs (e.g. DDT) and PAHs (e.g. B[a]P). In contrast, if a compound re-enters the atmosphere after initial deposition to the Earth's surface, it is* *a "multi-hop" compound. Processes by which this can occur include volatilization from the Earth's surface under warmer temperatures than which it was deposited, sudden exposure of ocean water to the atmosphere after being covered by ice or resuspension by winds of soil dust. The source region of "multi-hop" compounds in the Arctic is not only defined by the processes of atmospheric transport and removal but also by surface processes that control its re-entry into the atmosphere. Most OCs, many PAHs and Hg fall into the "multi-hop" group.*

2.2.1.2 Marine

The Arctic Ocean is often called a "mediterranean sea" because it is surrounded by land, interacting with the oceans to the south only through restricted passages. The major inflowing ocean waters pass from the North Atlantic through Fram Strait and the Barents Sea and from the North Pacific through Bering Strait (Figures 2.3 and 2.4). In the context of contaminant transport, the surface ocean currents in approximately the top 200 m of the ocean (Figure 1.2) will generally be most important. These surface currents take from years to decades to transport water from industrialized, temperate coastal regions to the Arctic Ocean.

The flow paths of surface ocean currents and the time scale for them to arrive at the entrance to the Arctic Ocean are best known for the North Atlantic. This understanding derives primarily from the releases of contaminant radionuclides to the sea from European nuclear fuel re-processing plants at Dounreay (Scotland), Cap de la Hague (France) and Windscale/Sellafield (England) — Sellafield being the most important, historically and currently. These radionuclide releases, while sufficient to provide sensitive water-mass tracers, have produced radioactivity levels orders of magnitude less than those of natural radionuclides.

The predominant organizing system of surface currents in the North Atlantic is the Gulf Stream, whose source is in the Gulf of Mexico (Figure 2.3). This system transports large quantities of heat from near the equator to the European coast, thus moderating climate there. Persistent, soluble contaminants that do not tend to become scavenged by particles (i.e. the so-called "conservative" contaminants) entering the sea from the eastern seaboard of North America will likewise be transported by this system, with considerable dilution, to the coast of Europe.

Surface water arriving off the coast of Europe continues to move northward as illustrated by radionuclide tracers (Figure 2.4, Aarkrog *et al.* 1987, Dahlgaard 1995). Marine transport of ^{137}Cs from the Irish Sea to the entrance of the Arctic Ocean takes about five to seven years following a current path along the Norwegian coast (Figure 2.3, Aarkrog *et al.* 1987). The water then enters the Barents and Kara seas and the Arctic Ocean through the Fram Strait.

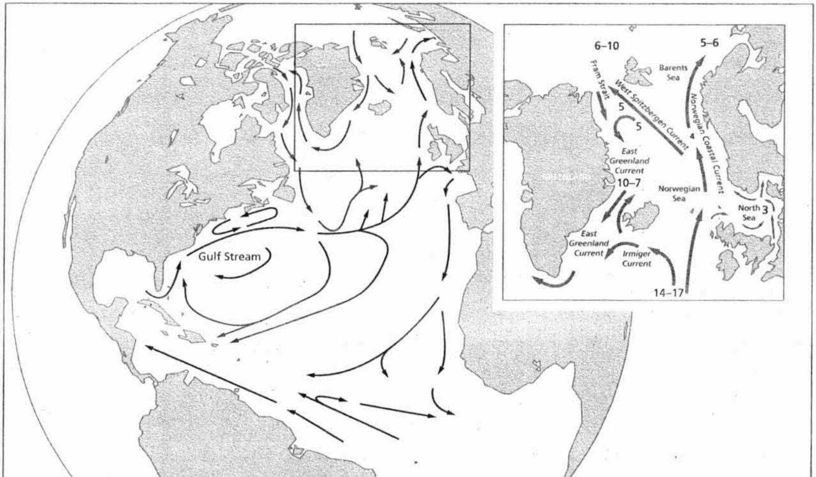


FIGURE 2.3

The main surface currents for the North Atlantic Ocean transporting water toward the Arctic (after Tchernia 1980, Dahlgaard 1995).

Some of the northward flowing water is returned to the North Atlantic in a subsurface flow along eastern Greenland; however, a substantial proportion of the ^{137}Cs previously discharged from Sellafield — currently about 10 to 15 PBq — now resides in the Arctic Ocean (Aarkrog 1993). During transport, the contaminants are dispersed and diluted. Concentration changes have been used to estimate dilution on the way to the Arctic Ocean. Transfer Factors (TF) have been calculated from the observed radionuclide concentrations and from the known release inventory as defined by Dahlgaard (1995): $TF = Bq \cdot m^{-3}/PBq \cdot yr^{-1} = 10^{-15}\ yr \cdot m^{-3}$. For example, a TF value of 1 indicates that for each tonne discharged to the coastal current, $1\ ng \cdot m^{-3}$ is observed at a given location t years later, where t is the number of years required for the water mass to reach the site. TFs, estimated for European sources of radionuclides to the Arctic Ocean to be about 1–10 (Dahlgaard 1995), can in principle also be used for other conservative contaminants discharged from the coast of Europe.

In the Pacific Ocean, the main current system is dominated by a large clockwise gyre occupying most of the ocean between 5 to 10°N and 45 to 50°N (Figure 2.4 and see Tabata 1975). This system moves water from the eastern side of the North Pacific (Philippines, Taiwan, Japan) across the Pacific Ocean to the west coast of North America at about 45° to 50°N latitude. Because the net flow through the Indonesian Archipelago is from the Pacific Ocean to the Indian Ocean (Verschell *et al.* 1995), it is not likely that contaminants from the Indian Ocean will be efficiently transferred to this Pacific Ocean gyre. Water is estimated to take one to two years to transit the Pacific Ocean and perhaps three to five years to go around the gyre. Flow splits at the North American coast with some of the water flowing southward and the remainder northward in surface currents to enter the Gulf of Alaska. This northward flow is augmented near the coast by large quantities of freshwater runoff from the coasts of Alaska and British Columbia (Royer 1982). Runoff from the Fraser and Columbia rivers, which drain populated, agricultural and industrial regions, supplies at least some of the fresh water entering this coastal current (Macdonald and Pedersen 1991). From the latitude of Vancouver Island, it takes approximately one to two years for a water parcel to reach the Aleutian Islands (Alaska).

The Aleutian Archipelago forms a boundary that steers currents toward the west (Figure 2.4). However, some of the northward flowing water enters the Bering Sea through the many passages. The Bering Sea contains a enormous, shallow (100m) shelf where water is subject to vigorous physical and biological processing (Walsh *et al.* 1989, Coachman and Shigaev 1992). Within the Bering Sea, currents following the

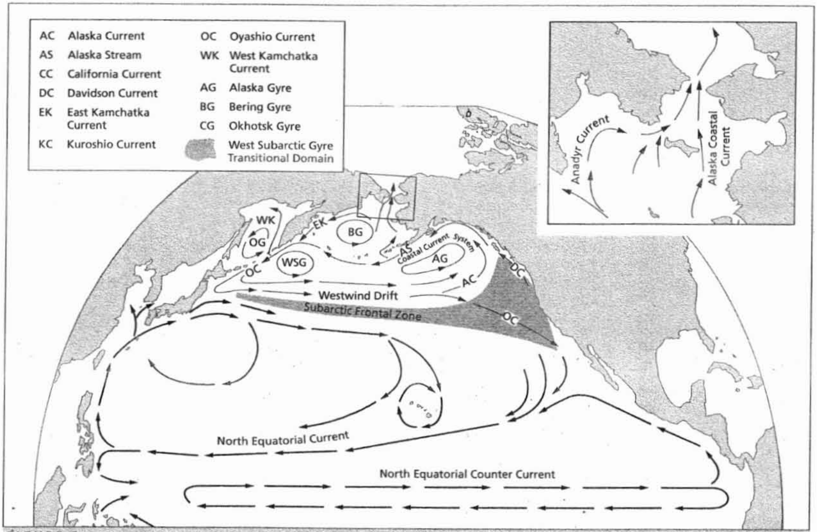


FIGURE 2.4

The main surface currents for the North Pacific Ocean transporting water toward the Arctic Ocean (after Tabata 1975, Tchernia 1980).

Bering slope supply the Anadyr Current found along the western side of the Northern Bering Sea while the Alaskan Coastal Current transports water along the North American side (Coachman 1993, and Nihoul *et al.* 1993). The Alaskan Coastal Current collects runoff from the land, including input from the Yukon River. These currents together supply the water that flows into the Arctic Ocean across the 50 m deep sill in Bering Strait. Because the north Pacific Ocean is about 0.5 m higher than the Arctic Ocean, the net flow is into the Arctic Ocean.

The estimation of oceanic material transport from the Atlantic or Pacific Oceans into the Arctic Ocean via these passages rests upon a reliable estimate of the rate and direction of exchange of the water. During the past decade, our knowledge of flows into and out of the Arctic Ocean has improved considerably. Thus, an improved estimate of the water mass budget for the Arctic Ocean was made (Table 2.1). The criteria used in constructing the mass budget were: i) the inputs and outputs of water reasonably reflect the information given in the source references, and ii) that the water budget balanced. There remain many uncertainties in both the volume flows and in their temporal variation, especially for the major passage in Fram Strait.

Of all the flows into the Arctic Ocean, the estimate of flow through the Bering Strait is most confidently known. The mean volume flow is northward (0.83 Sv (10^6 $m^3 \cdot s^{-1}$)) with a maximum error of 30% (Roach *et al.* 1995). Interannual variability is in the order of 0.1 Sv, but it can at times reach 50% of the mean. Since the sill depth of Bering Strait is about 50 m, only shallow water is exchanged. Ice transport can be safely ignored since it is much smaller than the errors in other terms.

The Canadian Archipelago consists of a maze of channels through which water from the Canada Basin flows predominantly out of the Arctic Ocean. Fissel *et al.* (1984) gave an overview of the bathymetry. The deepest channel through Nares Strait is 250 m deep while the next deepest channel is 105 m deep. Annual volume flows are not confidently known, but have been estimated at 1.7 Sv (Fissel *et al.* 1988). Consisting mostly of surface water, they are important to the Arctic Ocean freshwater and contaminant budgets. Contaminants exit the Arctic Ocean through the Archipelago.

Water flows into the Arctic Ocean from the Atlantic Ocean are difficult to estimate because there are at least two main branches into the Arctic Ocean (W. Spitzbergen Current and through the Barents Sea) and the flows through Fram Strait undergo considerable recirculation obscuring the calculation of net import of water (or contaminant). Fram Strait is also an important deep channel (sill depth 2500m) by which both water and ice leave the Arctic Ocean. It is through this passage that deep-water exchange occurs.

TABLE 2.1

Mean annual waterbudget for the Arctic Surface Ocean (0–200 m). Units of flow are in Sverdrups = 1 x 10^6 $m^3 \cdot s^{-1}$ = 31 500 $km^3 \cdot a^{-1}$. Uncertainties have been taken from the literature, where available, and by considering the ranges available from the various literature sources. The uncertainties, therefore, are for the most part not statistical calulations but rather estimates. Uncertainty for the totals has been estimated as square root of sum of squares of uncertainties.

Import/Export	In	Out	References
Ice through Fram Strait (81°N)		0.15 ± 0.04	Vinje and Finnekasa 1986, Aagaard and Carmack 1989, Bauch *et al.* 1995
Ice through Archipelago		<0.005± 0.005	Aagaard and Carmack 1989
Ice through Bering Strait	0.001		Aagaard and Carmack 1989
Runoff	0.11 ± 0.02		Treshnikov 1985, Rudels 1987, Aagaard and Carmack (1989)
Precipitation-Evaporation	0.05 ± 0.025		Walsh *et al.* 1994, Rudels 1987
Water through Bering Strait	0.80 ± 0.24		Roach *et al.* 1995, Coachman and Aagaard 1988, Rudels 1987, Aagaard and Carmack 1989
Water through Archipelago		1.7 ± 0.4	Fissel *et al.* 1988, Muench 1971, Rudels 1987
East Greenland current (<0° polar water)		1.0 ± 0.5	Foldvik *et al.* 1988
Arctic Intermediate Water (0<T<3°C)		2.0 ± 1.0	Foldvik *et al.* 1988
Norwegian coastal current	0.70 ± 0.35		Loeng 1991, Blindheim 1989, Rudels 1987
Barents Sea from Greenland Sea (net)	1.20 ± 0.6		Aagaard and Carmack 1989, Rudels 1987, Loeng 1993
West Spitzbergen Current (net)	2.00 ± 1.0		Aagaard and Carmack (1989)
Total	**4.86 ± 1.3**	**4.86 ± 1.3**	

Arctic Ocean Total Surface Area = 9.58 x 10^6 $(km)^2$
Total budget volume (top 200 m) = 1.92 x 10^6 $(km)^3$

Presently, and for the foreseeable future, budget calculations will be limited by the uncertainties in water exchange at this entrance to the Arctic Ocean.

The mean residence time of water in the surface ocean domain corresponding to the water volume and mean steady state inflow of 4.86 ±1.3 Sv, is 12.5 ±3.3 years. This is in reasonable agreement with previous estimates (Ostlund and Hut 1984, Wallace *et al.* 1987, Barrie *et al.* 1992).

- *Marine currents deliver 97% of the water to the surface Arctic Ocean (0 to 200 m depth) from the Pacific Ocean through Bering Strait (17% of total input) and from the Atlantic Ocean through eastern Fram Strait and the Barents Sea (80%). Precipitation and rivers contribute the remaining 3%.*
- *Marine currents carry 96% of the water flowing out of the Arctic annually. This occurs through the Canadian Archipelago (34%) and western Fram Strait (62%). The rest (4%) flows out in ice. The mean residence time of water in the Arctic Ocean surface layer is 12.5 years.*

2.2.1.3 Rivers

Northward flowing rivers that drain 10^7 km^2 of northern Asia, northern Europe and North America may be major conduits to the Arctic Ocean of contaminants originating from point sources and/or atmospheric deposition to the terrestrial ecosystem. Estimates of the total annual river discharge into the central Arctic Basin range from 2700 to 5000 x 10^9 m^3 with a mean annual discharge of 3640 x 10^9 m^3 (Cattle 1985, Aagaard and Carmack 1989). About 78% of the total annual riverine input to the Arctic Ocean is via 10 large rivers (draining 1.1 x 10^7 km^2): nine Russian and one Canadian — the Mackenzie River (Table 2.2). The remaining drainage is from a larger number of smaller rivers. For Canada, small-river water inputs are listed and compared to that of the Mackenzie River in Table 2.3.

There is an important difference between the Mackenzie River and the small Canadian arctic rivers. The Mackenzie River flows directly into the Arctic Ocean while most of the smaller rivers discharge into the Arctic Archipelago. As the Archipelago is known to be a major outflow region of the Arctic Ocean (see Section 2.3.3) these waters do not have a direct impact on the Arctic Ocean. Nonetheless, these rivers can play an important role in the generally shallow, more productive waters of the Archipelago with respect to the delivery of sediments and nutrients, mixing due to density currents and ice formation and breakup. Our understanding of these nearshore interactions is very limited at this time.

TABLE 2.2

Drainage area and annual flow of arctic rivers (adapted from Mackay and Loken 1974).

River	Drainage (10^6 km^2)	Annual Flow (10^9 m^3)	Fraction of total river input (%)
Yenisey	2.6	603	18.3
Ob	2.5	530	16.0
Lena	2.5	520	15.7
Mackenzie	1.8	340	10.3
Pechora	0.25	130	3.9
N. Dvina	0.35	110	3.3
Kotuy	<0.06	110	3.2
Kolyma	0.64	102	3.1
Pyasina	0.18	86	2.6
Indigirka	0.36	57	1.7
Other small	—	720	21.9
Total		**3308**	**100**

TABLE 2.3

Drainage areas and mean annual flow of major northern Canadian rivers.

River	Drainage (10^4 km^2)	Annual Flow (10^9 m^3)	Fraction of total river input (%)
Mackenzie	184.3	340	75
Coppermine	4.7	10.5	2.3
Burnside	16.8	5.4	1.2
Ellice	16.9	3.0	0.6
Dubawnt	67.3	13.8	3.0
Thelon	152.0	33.5	7.4
Back	93.9	15.8	3.5
Kazan	70.0	16.9	3.7
Hayes	18.1	4.1	0.9
Quoich	30.1	7.6	1.7
Lorillard	11.0	2.5	0.6

There is considerable variation between the suspended particulate matter abundance and natural water chemistry of Canadian arctic rivers. Drainage basins in the east are predominantly comprised of shield rocks and tundra with a shallow active layer while those in the western Arctic have steeper slopes, more erodible surficial material and a deeper active layer (Gregor 1990). The Mackenzie basin rivers, are characterized by higher total dissolved solids, pH, total alkalinity and suspended sediments than rivers in the eastern Arctic (Brunskill 1986, Telang *et al.* 1991). Rivers draining the tundra including those underlain immediately by the Pre-Cambrian Shield are characterized by low total dissolved solids, pH, total alkalinity and suspended sediments. Of course, transition type rivers occur between these two extremes. Northern rivers are also typified by low nutrient concentrations and generally low organic carbon.

The dominant hydrologic feature of arctic rivers is extreme seasonality in water flow marked by the snowmelt freshet in May and June. The hydrograph for the Mackenzie River (Figure 2.5a) is typical of large arctic rivers. River systems conduct not only water but also large quantities of suspended particles and dissolved organic matter to the receiving waters. Suspended solids, originate from soil and bank erosion in the watershed and by the resuspension of particles deposited in the river bed. The abundance and timing of input of these natural substances depends on the nature of the river, but for most northern rivers the bulk of the suspended solid

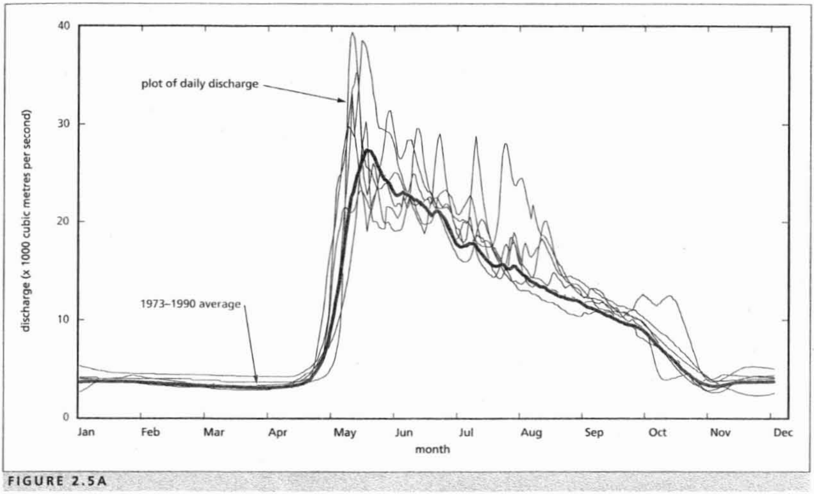


FIGURE 2.5A

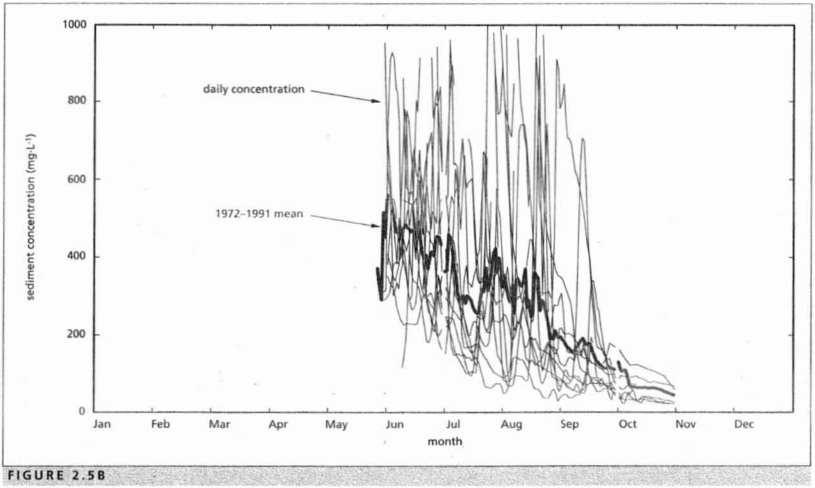


FIGURE 2.5B

Mackenzie River data provided by Environment Canada Inland Waters Directorate. Samples taken above Arctic Red River, NWT.

(A) Annual hydrographs of daily discharge (thin line) and 1973–1990 mean daily discharge (thick line). The plot shows the dominance of the spring freshet in May.

(B) Plot of daily (thin line) and 1972–1991 mean daily (thick line) suspended sediment concentrations for the period of high flow and high sediment concentration. Sediment transport loads are largely determined by discharge and consequently, the load peaks during May and June, with these two months accounting for approximately 60% of the total annual sediment transport (Lewis 1988).

transport (Figure 2.5b) occurs in association with the spring freshet. River flow rates and the concentration of suspended solids are often correlated (Meybeck *et al.* 1992) because of the observed phenomenon of resuspension of fine-grained bottom sediment with increasing discharge (Thomas and Meybeck 1992). Nevertheless, very large systems such as the Mackenzie River have complex transport regimes because different tributaries deliver suspended particulate matter at different times and locations (relative to the mouth). Arctic lakes can act as sinks for particle-associated contaminants. However, this is strongly dependent on their size, basin morphology and location.

- *78% of the total annual riverine input of water to the Arctic Ocean is via ten large rivers: nine Russian and one Canadian — the Mackenzie River. The Mackenzie River carries 75% of the total Canadian riverine water input to the Arctic Ocean. The rest is through a number of smaller rivers that discharge into the Canadian Archipelago and play an important role in the generally shallow, more productive waters of the Archipelago. River systems conduct not only water but also large quantities of suspended particles to the receiving waters. An important feature of arctic rivers is extreme seasonality in waterflow and suspended solid content marked by a maximum in May and June.*

2.2.2 Emissions/Usage

To model the pathways of contaminant transport, it is necessary to know the temporal and spatial distribution of contaminant release globally or at least hemispherically. Research on atmospheric emissions in the NCP has focused on OC pesticides and PCBs. A database was created of historical, present, and predicted global usage or sales of these compounds. Data used were from United Nations and government reports, scientific publications, Batelle Europe and the International Registry of Potentially Toxic Contaminants (IRPTC) of the United Nations Environment Programme (UNEP). An estimate of global cumulative usage (Table 2.4) has been made for selected agricultural pesticides and industrial compounds (PCBs). The inventory of historic use of many of these compounds is still incomplete.

TABLE 2.4

Best estimates of global usage of selected major organochlorine contaminants.

Organochlorine	Function	Period of time	Estimated Total Global Usage (megatonnes)
PCBs	dielectric/ hydraulic fluid	1930–1992	1.2[1]
DDT	insecticide	1950–1992	2.6[3]
		1970–1992	0.99[3]
Toxaphene	insecticide	1950–1992	1.33[2]
		1970–1992	0.67[2]
Lindane	insecticide	1950–1992	0.72[3]
Technical HCH	insecticide	1950–1992	5.6[4]
Chlordane	insecticide	1945–1988	0.078[1]
Aldrin	insecticide	1950–1992	0.50[1]
Dieldrin	insecticide	1950–1992	0.034[1]
Endosulfan	insecticide	1956–1992	0.057[1]

[1] *Barrie* et al. *1992*

[2] *Voldner and Li 1993*

[3] *Voldner and Li 1995*

[4] *Li* et al. *1997*

HCHs are the most abundant pesticides in Arctic air and water. Constructing a mass budget for them in the global environment helps us to understand the processes controlling the input and fate of other OC pesticides. From usage estimates of technical HCH (a mixture of mostly α-HCH and some γ-HCH) and γ-HCH (lindane), the total usage of α-HCH and γ-HCH globally and by country was calculated (Tables 2.5 and 2.6). This was done by assuming that technical HCH contains 70% α-HCH and 15% γ-HCH, and technical lindane contains 100% γ-HCH.

Usage of α-HCH and γ-HCH in 1980 and 1990 are listed in Tables 2.5 and 2.6, respectively, for the major consuming countries. In 1980, the annual consumption of α-HCH in China, India and the former Soviet Union, accounted for more than 95% of the total α-HCH consumption in the world. In 1990, although increased in India, the former Soviet Union and Mexico, the usage of α-HCH had

TABLE 2.5

Estimated annual α-HCH usage ($t \cdot a^{-1}$) in 1980 and 1990 for the highest consuming countries (Li 1996, Li *et al.* 1997)

Country	Usage in 1980	Usage in 1990
China	200 521	0
India	15 100	32 900
Former Soviet Union	11 718	17 528
Former East Germany	2 626	84
Argentina	1 470	0
Sri Lanka	770	n/a
Turkey	628	n/a
Niger	358	n/a
Mexico	105	1 218
South Korea	84	0

n/a = not available

TABLE 2.6

Estimated annual γ-HCH usage ($t \cdot a^{-1}$) in 1980 and 1990 for the highest consuming countries (Li 1996, Li *et al.* 1997)

Country	Usage in 1980	Usage in 1990
China	42 969	100
India	3 376	7 050
Former Soviet Union	2 511	3 756
Italy	1 580	600
France	1 172	1 863
Former East Germany	563	18
Argentina	315	n/a
United States	268	114
Canada	200	284
Sri Lanka	165	n/a
Turkey	135	n/a.
Niger	77	397
Mexico	23	261
South Korea	18	n/a
Former Yugoslavia	n/a	151
Spain	n/a	96
Pakistan	n/a	3

n/a = not available

decreased dramatically in other countries. In 1980, the annual consumption of γ-HCH in China, India and the former Soviet Union accounted for about 90% of the total γ-HCH consumption in the world. By 1990, the global γ-HCH consumption had decreased greatly due to the banning of technical HCH usage in China in 1983.

Figure 2.6 presents trends in usage of α-HCH and γ-HCH from 1948 to 1995 for India, China, and the former Soviet Union. Due to heavy usage of technical HCH, the usage of both α-HCH and γ-HCH in China was the highest in the world in the 1960s and 1970s, and reached 200 kt·a^{-1} for α-HCH and 43 kt·a^{-1} for γ-HCH in 1980. In 1983 China banned use of technical HCH, and started to use lindane in 1991. Since 1995, usage of γ-HCH has been about 500 t·a^{-1}. The usage of α-HCH in India increased constantly before 1970 and from 1980 to 1990, peaking at 33 000 t·a^{-1} in 1990, and has remained around 17 000 t·a^{-1} since 1991. The usage of γ-HCH in India reached 7000 t·a^{-1} in 1990, and kept around 3600 t·a^{-1} since 1991. The highest usage of α-HCH in the former Soviet Union was 20 000 t·a^{-1} in 1970, dropping to zero around 1992. The data for γ-HCH usage in the former Soviet Union after 1991 are not available.

Three-dimensional models of the global movement of pesticides (e.g. AES 1997, next section) require atmospheric emissions inventories on a regular grid. For HCH compounds most of what is applied is released to the atmosphere within a year of application. Thus annual usage and emissions to the atmosphere are roughly equivalent. Gridded usage of α-HCH and γ-HCH are shown in Figures 2.7 and 2.8, respectively, for the years 1980 and 1990 (Li 1996). In 1980, areas with the highest application of α-HCH were in India and eastern China. The highest usage densities of α-HCH in China reached more than 1000 t·a^{-1} per grid cell. Between 1980 and 1990, as many countries were phasing out the use of α-HCH, the magnitude and pattern of usage in India changed little. The usage patterns of γ-HCH in 1980 and 1990 were quite similar in Europe, North America, and Southeast Asia. Since its introduction in 1943, total global technical HCH usage has been estimated to be as high as 5.6 million tonnes (Li *et al.* 1997).

Emissions of PAHs to the atmosphere are not well quantified. The primary anthropogenic sources are the burning of fossil fuels and biomass which releases PAHs to the atmosphere. In the absence of a quantitative emissions inventory for PAHs, a qualitative picture of the spatial distribution of fossil fuel combustion PAHs can be obtained from the 1° x 1° gridded emissions inventory of black carbon (soot) emissions available for the Global Emissions Inventory Activity (GEIA) (Figure 2.9). Black carbon is particulate material consisting mainly of elemental carbon and black organic compounds produced as a result of incomplete fossil fuel combustion.

Secondary anthropogenic sources of PAHs to the environment are oil exploration, development and transport losses within the Arctic. This releases oil containing PAHs to marine waters. Oil exploration in the 1970s and 1980s resulted in the discharge of some hydrocarbons to the ocean around drill sites in the Beaufort Sea (Thomas *et al.* 1986), however, PAH inputs were small compared to natural budgets and could be detected only within relatively short distances of the drill sites. Because PAHs are scavenged by particles, sediments and soils are often the first place to find signs of anthropogenic inputs and their effects. For example, coastal zones near industrial regions often have elevated PAH concentrations in surface sediments (Hites *et al.* 1980, Bouloubassi and Saliot 1991) which can, in some cases, harm biota living close to the sediments (Malins *et al.* 1984).

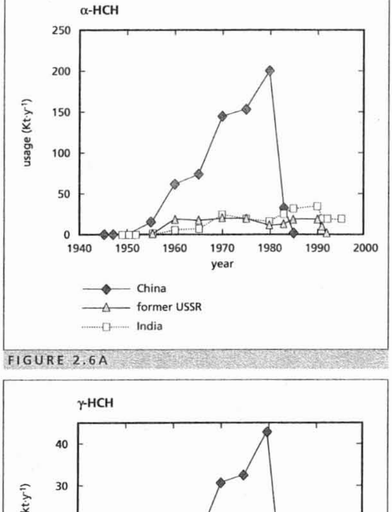


FIGURE 2.6A

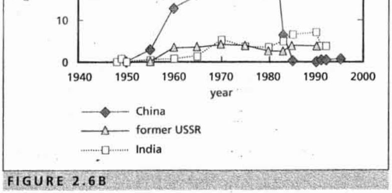


FIGURE 2.6B

Trends in usage of (A) α-HCH, and (B) γ-HCH from 1948 to 1995 for India, China and the former Soviet Union. (Li 1996).

- *Ranked in order of total global usage between 1950 and 1992, the most used persistent organochlorines were: HCHs (6.3 Megatonnes), DDT (2.6 Mt), toxaphene (1.33 Mt) and PCBs (1.2 Mt). HCHs are the most abundant OC in arctic air and water.*

- *Most usage of HCH has occurred in China, India and the former USSR. China and India switched to using only the pesticide-active isomer of HCH (γ-HCH, lindane) in 1983 and 1990, respectively. The annual usage of HCH globally shows a step-jump decrease in these years.*

FIGURE 2.7A

FIGURE 2.7B

Globally gridded annual usage of α-HCH usage (t·a^{-1} per 1° x 1° grid cell) for (A) 1980 and (B) 1990 after Li (1996).

- *A prime product of NCP research has been the production of a globally gridded (1 lat x 1 long) inventory of annual usage of the HCH isomers for 1980 and 1990. This is essential for 3-D environmental models of pathways.*

FIGURE 2.8A

FIGURE 2.8B

Globally gridded annual usage of γ-HCH usage (t·a⁻¹ per 1° x 1° grid cell) for (A) 1980 and (B) 1990 after Li (1996).

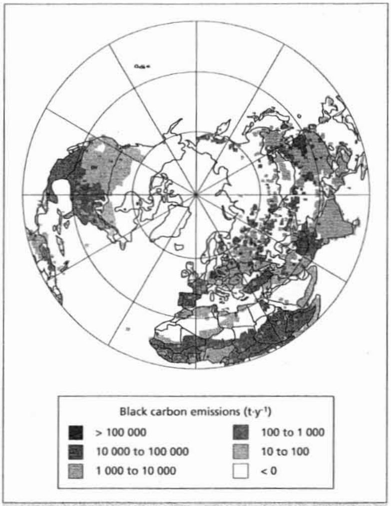


FIGURE 2.9

Global annual black carbon emissions in 1985 (1° x 1° grid) from the Global Emissions Inventory Activity (GEIA) data base ($t \cdot a^{-1}$) (Dignon et al. *1994).*

2.2.3 Physical Chemical Properties of Contaminants and the Important Role of Phase-Partitioning in Contaminant Pathways

The properties of OCs and PAHs that are key to understanding their movement through the environment include vapour pressure (VP), Henry's Law constant (H), and partition coefficients between octanol and environmental phases (e.g. octanol/air (K_{oa}), octanol/water (K_{ow}), and between organic carbon/water (K_{oc})). All of these are strongly dependent on temperature. Water temperatures range from about 30°C at the equator to -1.7°C in the polar regions. Air temperatures vary seasonally, latitudinally and with altitude from approximately -90°C to 50°C. Thus, the complexities of environmental transport and air-surface exchange cannot be understood without global models which include the temperature dependence of these physicochemical properties. Later in the section on global models, these interactions will be described in more detail.

The vapour pressure of a substance together with the surface area of atmospheric aerosols determines particle/gas partitioning. Nearly all of the contaminants of interest in the NCP are solids in pure form at arctic temperatures, yet it is the VP of the supercooled liquid, not the solid, that governs environmental partitioning (Bidleman 1988). The supercooled liquid VP of a substance below its melting point cannot be determined readily. One approach is to extrapolate the VP measured above the melting point (for example by gas chromatography Hinckley *et al.* 1990) to lower temperatures. A second way is to determine the solid-phase VP and use a thermodynamic relationship to estimate the liquid-phase VP at temperatures below the melting point (Hinckley *et al.* 1990). Solid-phase VP values of a few substances have been measured at arctic temperatures (Wania and Mackay 1995). The uncertainty in each of these approaches increases as the difference between ambient temperature and the melting point becomes greater.

In Figure 2.10, liquid-phase VP for various OCs and PAHs at 20°C are shown ranked in decreasing order. The relationship between VP and absolute temperature (T) is generally expressed by the Clausius-Clapeyron equation.

$$\log_{10} VP = m/T + b \qquad (2.1)$$

The parameters m and b have been determined for several organochlorine pesticides, PAHs and PCB congeners (Hinckley *et al.* 1990).

Henry's Law constant is defined as the ratio of the concentration or partial pressure of a substance in air (P) to its concentration in water (C_{liq}) at equilibrium.

$$H = P/C_{liq} \qquad (2.2)$$

As defined in equation 2.2, the gas partitioning is proportionally less to water as H increases. Henry's Law constant is temperature and compound dependent. It increases with increasing temperature. The critical role played by H in estimating air-water gas exchange fluxes is discussed in Section 2.4.1. Henry's Law constants are usually reported at 20°C to 25°C, and have been measured as functions of temperature for only a few of the NCP chemicals (Alaee *et al.* 1995, Kucklick *et al.* 1991, tenHulscher *et al.* 1992). The temperature dependence of H has a similar form to equation 2.1, but with values of m different than the corresponding temperature slopes for VP. Figure 2.11 shows Henry's Law constants for some compounds at -2°C, the freezing point of seawater. Values of H for naphthalene, phenanthrene, HCB and the HCHs were calculated from reported temperature-property relationships (Alaee *et al.* 1996, Kucklick *et al.* 1991, tenHulscher *et al.* 1992). Others were extrapolated from literature values at 20°C–25°C by assuming a temperature slope (Cotham and Bidleman 1991).

Figure 2.11 shows the percentage of total chemical in the gas phase in a closed container with air and water in a 20 to 1 ratio — roughly in the same proportion as the tropospheric air (10 km deep) and the surface ocean mixed layer (0.5 km deep). According to these calculations, only 0.03% of γ-HCH (low H) remains airborne after equilibration, and >1.3% of CB-153 (high H) would still be in the atmosphere. These calculations neglect particle partitioning in air

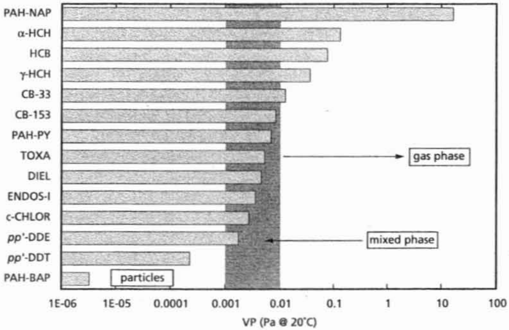


FIGURE 2.10

The vapour pressure (VP) of selected organochlorine and PAH compounds in air at 20°C. The vertical bar indicates the approximate dividing line between those compounds that are predominantly in the gas phase and those that partition significantly between the gas and particulate phase in the atmosphere.

or water. This would tend to increase the proportion of released material that ends up in the biosphere or marine reservoirs.

The octanol/water partition coefficient (K_{ow}) is used to describe the equilibrium distribution of organic contaminants between lipid phases and water, and is widely applied as a correlation parameter for bioaccumulation and sediment sorption. Compilations of K_{ow} values have been made by Hawker and Connell (1988) and Mackay *et al.* (1991). Several relationships have been proposed to estimate partition coefficients between the organic carbon fraction of soil or sediment and water (K_{oc}) from K_{ow} (Eisenreich *et al.* 1987, Schwartzenbach *et al.* 1992).

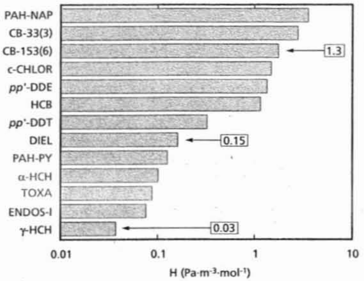


FIGURE 2.11

A comparison of Henry's Law constant at -2°C for selected organochlorine and PAH compounds. Numbers in boxes represent the percent of total mass remaining in air at equilibrium in a closed container at -2°C containing a mixture of air and water equivalent to 10 km of atmosphere and 0.5 km of ocean surface water.

Particle size and organic carbon content are acknowledged as playing key roles in the adsorption of chemical compounds to particulate matter (Knezovich *et al.* 1987). It is the surface area of the particles and the amount of organic carbon present that determine adsorption capacity. This capacity is inversely proportional to the grain size of suspended solids in water. Also important in the transfer of contaminants is that finer grained particles settle less quickly and thus are subject to riverine transport over long distances.

The octanol/air partition coefficient (K_{oa}) has been suggested as a parameter to describe the distribution of organic contaminants between air and lipid-like phases such as plant waxes (Harner and Mackay 1995) and organic films on aerosols (Finizio *et al.* 1997). Values of K_{oa} as a function of temperature have been measured for several PCB congeners, chlorobenzenes and *p,p'*-DDT (Harner and Mackay 1995, Harner and Bidleman 1996). The relationship between K_{oa} and temperature is similar to that expressed by equation 2.1, with values of m lying between those for VP and H.

2.2.3.1 Particle/Gas Partitioning and Atmospheric Deposition Processes

Exchange of organic compounds between the atmosphere and the Earth's surface takes place by rain and snow scavenging of gaseous and particulate species and transfer of gaseous compounds across air-sea, air-lake and air-land surfaces. Flux equations used to describe these processes necessarily take into account the partitioning of the compound between the particle and gas phases in air (Cotham and Bidleman

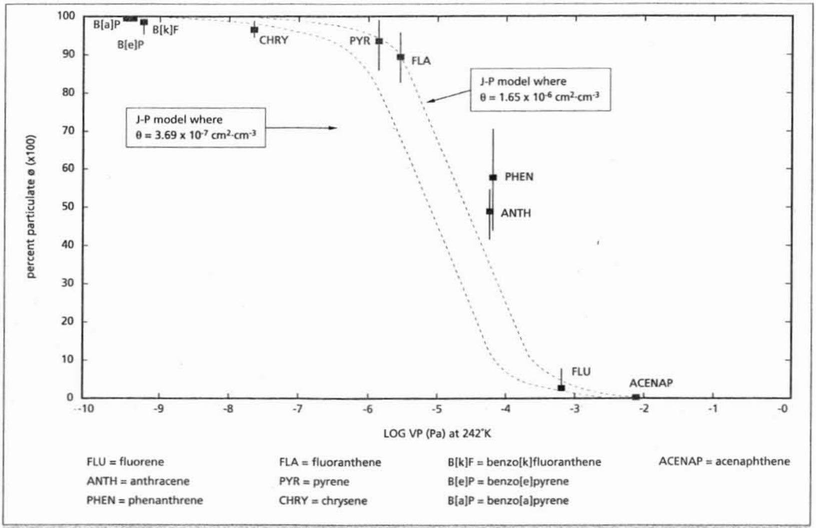


FIGURE 2.12

Modelled (Junge-Pankow) (dotted lines) and observed (solid squares) particle fractions (%) of PAHs at Alert (haze season 1993). Aerosol surface areas were generated from measured sulphate particulate measurements, including a water vapour growth factor of 2.84 for 90% RH. Mean temperature was –31°C from December to March, the bars indicating the particulate fraction range encountered during the period.

1991, Table 6.3). Estimates of the atmospheric phase distribution are frequently made with the Junge-Pankow (J-P) adsorption model (Pankow 1987):

$$\phi = c\,\theta \;/\; (VP + c\,\theta) \tag{2.3}$$

where ϕ is the fraction of the total airborne compound sorbed to aerosols, VP is the liquid-phase vapour pressure (Pa) at the ambient temperature and θ is the atmospheric aerosol surface area (cm^2 per cm^3 air). The parameter c, which depends on the thermodynamics of adsorption and the surface area occupied by a sorbate molecule, is not a constant and may change with compound and aerosol type and is often taken as 17.2 Pa-cm based on empirical evidence. Values of θ that are representative of urban, rural and clean air regimes are given by Bidleman (1988).

The adsorption of vapour-phase substances to arctic aerosols is most important during the winter haze season, when the air temperature is low (approximately -30°C to -40°C) and the aerosol concentration is relatively high. As a general rule of thumb, substances that have VP $\leq 10^{-3}$ Pa at arctic winter temperatures (-30°C to -40°C) will be 0.5% or more in aerosol form under typical haze conditions (Figure 2.10). During winter, the expected percentages of α-HCH, *p,p'*-DDE and benz[a]anthracene on haze aerosols are 2%, 50% and 90% respectively, reflecting the variation in VP for each compound from high to low. In summer when the arctic air temperature warms to above 0°C and the aerosol surface area density is less than one-tenth of winter values, these percentages fall to <0.001%, 0.5% and 4% respectively, for the three compounds. Thus, many of the organic contaminants of interest in the Arctic are transported in the gas phase during summer. In the winter, however, they are less likely to be transported in the gas phase because they become somewhat sorbed to haze aerosols, due to colder temperatures.

Air-monitoring of OCs and PAHs is often carried out with a high-volume (hi-vol) air sampler, which uses a glass- or quartz-fiber filter followed by a polyurethane foam trap to make an operational split between the particle and gas phases. Experimental investigation of sorption of PAHs and OCs to aerosols was carried out at Alert, NWT from February to April 1988 by Patton *et al.* (1991) and more recently for year-round data at several Arctic locations by Halsall *et al.* (1997). These studies show that the fraction of organic contaminants in particulate form increases for compounds of lower vapour pressure, in agreement with the J-P model, and that PAHs are sorbed to aerosols more strongly than OCs of the same volatility (Patton *et al.* 1991). Using PAH data at the Alert site for the haze period of 1993, Halsall *et al.* (1997) found that eight of 10 PAH particle fractions measuring at -31°C (242K) show a good fit to the theoretical J-P lines (Figure 2.13). The exceptions are phenanthrene and anthracene. The theoretical lines were derived using equation 2.3. The atmospheric aerosol surface area (θ) was calculated from the highest and lowest particulate-sulphate concentrations measured during the 1993 haze period. Particulate-sulphate concentrations were well correlated to independently measured aerosol surface area, obtained in the haze period of 1992 ($r^2 = 0.96$) by Staebler *et al.* (1994).

An important consideration in using sulphate particulate as a surrogate for total suspended particulate (TSP), is the growth of sulphate particulate by the sorption of water molecules. Although not measured, this has the effect of significantly increasing the aerosol surface area (Pitchford and McMurry 1994). Using a relative humidity of 90% for the haze period, the sulphate-particulate measurements were adjusted by a growth factor of 2.84. The corresponding aerosol surface areas, for the highest and lowest sulphate particulate concentrations respectively, are displayed in Figure 2.12 and have been used to generate the predicted particulate fractions in the J-P model.

The J-P model, using these generated aerosol surface areas, provides a good fit to the observed particulate fractions in the atmosphere of Alert. The two notable exceptions were phenanthrene (PHEN) and anthracene (ANTH), which lie to the right of the theoretical lines and displayed higher particulate fractions than the model predicted. In the study of Patton *et al.* (1991), where aerosol surface area was assumed, the additional compounds of fluoranthene and pyrene were also positioned to the right of these lines. This indicates that either a) the aerosol surface area has been underestimated, b) there is a significant artifact in the sampling protocol, c) the partitioning deviates from simple physical adsorption, or d) a combination of all of these. An adsorption artifact of the vapour component to the sampling filter can be considered unlikely, as use of a second in-line filter revealed negligible adsorption. Furthermore, under-

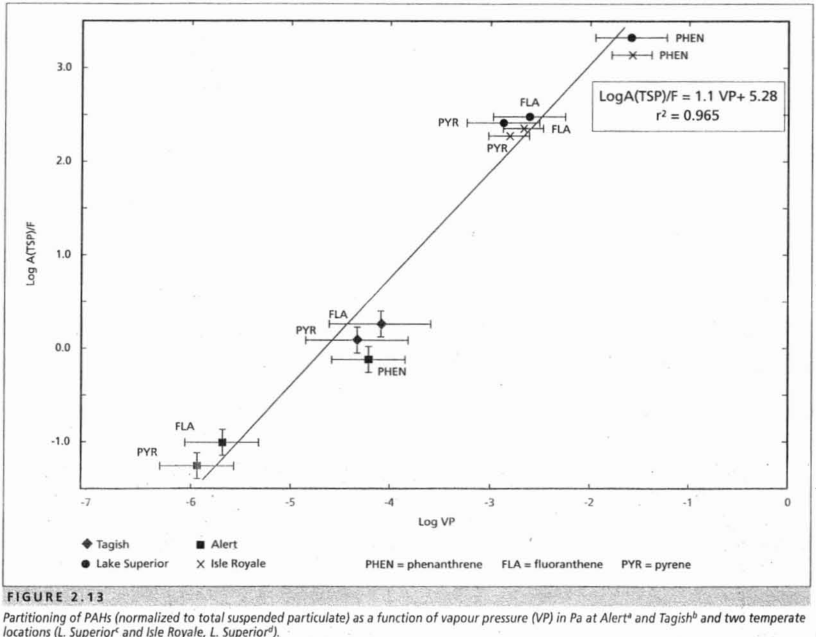


FIGURE 2.13

Partitioning of PAHs (normalized to total suspended particulate) as a function of vapour pressure (VP) in Pa at Alert[a] and Tagish[b] and two temperate locations (L. Superior[c] and Isle Royale, L. Superior[d]).

[a] Dec. 1992-March 1993 (Temperature range: -26°C to -36°C).

[b] Dec. 1992-March 1993 (Temperature range: -4°C to -21°C).

[c] Baker and Eisenreich (1990) (Temperature range: 10°C to 24°C).

[d] McVeety and Hites (1988) (Temperature range: 11°C to 20°C).

predictions of percent particulate by theory have also been found in temperate studies (McVeety and Hites 1988, Baker and Eisenreich 1990). Bidleman *et al.* (1997) suggested that this would indicate that some portion of the PAH associated with the aerosol is slow to exchange, or is in a non-exchangeable form and not in equilibrium with the vapour phase. Certainly in the extremely cold haze period the lighter PAH showed greater fractionation on particles than in warmer temperate regions.

A common way of expressing the vapour/particle partitioning of PAHs and other semi-volatile OCs is by use of a partitioning ratio A/F, where A (adsorbant) and F (filter) are nominally taken to represent the vapour and particle concentrations at equilibrium in the atmosphere respectively. The partitioning ratio, normalized to ambient total suspended particulate (TSP), has been shown to be directly related to equation 2.3 by:

$$\log A(TSP)/F = \log VP + \log C \qquad (2.4)$$

Where $C = TSP/\theta c$ and is a temperature-dependent constant (Ligocki and Pankow 1989, Cotham and Bidleman 1992). A plot of logA(TSP)/F vs log VP will have a slope approaching unity and an intercept of $\log TSP/c\theta$ and can be simply expressed as:

$$\log A(TSP)/F = m\log VP + b \qquad (2.5)$$

The two constants m and b refer to the slope of the line and the y-intercept respectively. Using equation 2.5 a comparison of Arctic to rural/remote temperate studies can be made for those PAHs that display significant concentrations in both phases over a wide temperature range. Figure 2.13 shows the resulting plot for three PAHs for the Arctic sites of Alert and Tagish and two temperate sites, Lake Superior and Isle Royale (McVeety and Hites 1988, Baker and Eisenreich 1990). For the two temperate studies TSP was actually measured or estimated (~10 $\mu g \cdot m^3$). Summer data for the Lake Superior sites was selected to provide a wide range in temperatures. For Alert and Tagish, TSP was assumed to be equal to the SO_4^{2-} concentration since this component with adsorbed water comprises most of the mass of Arctic haze. The mean temperature at Alert was -31°C while the mean temperature over Lake Superior was 17°C (Baker and Eisenreich 1990). Taking pyrene as an example, the vapour pressure decreased by approximately 3 orders of magnitude from Lake Superior in the summer to Alert in the haze period. With this large temperature fluctuation, the partitioning ratio also changes by ~3 orders of magnitude. Interestingly, the partitioning between the summer temporal studies and the haze period of the Arctic is strongly correlated ($r^2 = 0.965$), with the slope of the line being approximately 1. This would suggest that the partitioning behaviour for these compounds is similar for the various studies. Vapour pressure as a function of temperature adequately describes the partitioning behaviour of these PAHs, even at extremely cold temperatures. Furthermore, normalizing the partitioning to sulphate-particulate concentrations (used as a surrogate for TSP) proves successful in allowing direct comparisons to temperate studies, where TSP concentrations were significantly higher.

There is a need for additional studies of particle/gas partitioning in the haze season, especially to compare the hi-vol method of speciation with another technique, such as denuder measurements. Until more experimental data become available, the Junge-Pankow model is suggested as the best way of estimating particle/gas distributions in the NCP.

- *The partitioning of semi-volatile OCs and PAHs between the atmosphere and solid or liquid media (e.g. aerosol particles, snow crystals, snowpack ice, rain drops, ocean or lake water) is key to understanding atmosphere-surface exchange. Atmospheric gas and aerosol phase PAH observations in the Canadian Arctic show that partitioning of most PAHs are consistent with existing theory. Arctic haze sulphate aerosol mass can be used as a "surrogate" for aerosol surface area, a key parameter in this theory.*

2.2.4 Global Scale Models

Global scale models have the potential to:

(i) contribute significantly to our understanding of the extent of transport of contaminants from sources in tropical and temperate zones to the Arctic and Antarctic,
(ii) assist in the formulation and testing of hypotheses that there is a "cold condensation" effect tending to concentrate substances in regions of cold climate (Wania and Mackay 1993a),
(iii) quantify fluxes of contaminants to the Arctic,
(iv) estimate the fraction of the total global use of each contaminant that reaches the Arctic, and
(v) predict the time response to contamination and decontamination.

Since the models include several media including air, soils and ocean water, as well as a variety of biota, they synthesize the components of the global and Arctic system (Figure 1.2). The foundation for developing international contaminant control strategies with the aid of scientifically defensive models is obvious.

An attempt to model the global DDT cycle with a four latitude-zone box model by Ostromogil'skii *et al.* (1985) led to the conclusion that DDT contamination will persist for many decades and impact the Arctic much more than the Antarctic. After checking the model with observations in various environmental media, it was estimated that after a global ban of DDT the rate of decline of the contaminant in the atmosphere, soil and ocean will be a factor of 10% to 20%, 30%, and 1% to 2% per decade, respectively.

Although more comprehensive models are still in their infancy, considerable progress in model development has been made in the NCP. Existing global models for persistent organics deal with the multi-compartmental system (Figure 2.1.4) with varying degrees of complexity. For example, the atmospheric compartment in various global models has been treated in one (Wania and Mackay 1995), two (Strand and Hov 1993), and three dimensions.

Modelling studies are conducted in one of two ways dictated by the type of question to be answered and the type of model:

1. Long term-low spatial resolution mode
   The model starts with a "clean" global environment and simulates the entire time period that a chemical has been used on a global scale, or for as long as it is persistent in the environment. This is the approach taken in the 2-D and 1-D models described below. At this time, a 3-D model is impractical to describe this scenario due to its immense demand in computing time and input parameters.
2. Short term high spatial resolution mode
   The model defines the global contaminant loading in individual system compartments (Figure 1.2) at a single point in time. The model stimulates a shorter time period (e.g. one year). This is the approach taken by the 3-D models.

2.2.4.1 Global Zonally Averaged Multi-Compartmental Models

There have been two attempts to simulate dispersion of OCs on a zonally averaged, global scale. Strand and Hov (1996) modelled the distribution of HCHs using a 2-D, atmospheric model (Bergen-model), while Wania and Mackay developed a multimedia compartment model for OCs (Toronto-model, Wania and Mackay 1993b, 1995, Mackay and Wania 1995, Wania 1994, Wania and Mackay 1996).

The Bergen model is a global multi-compartmental model for HCHs that includes atmosphere-soil and atmosphere-ocean exchange. It is a two-dimensional zonally-averaged atmospheric model. The Bergen model simplifies an existing, detailed atmospheric model (Strand and Hov 1993). This was achieved by reducing the meridional and vertical resolution to six equally spaced latitude zones and four vertical layers. The model was then combined with a modified oceanic transport model (Siegenthaler and Joos 1992) and a soil-atmosphere exchange model for trace organics developed by Jury *et al.* (1983 1984a, 1984b). In summary, the model includes the compartments atmosphere, ocean water, cultivated and uncultivated soil and the processes of atmospheric advection and convection, diffusive gas exchange between atmosphere and soil or water, wet deposition and chemical degradation. Seasonality is taken into account by defining specific temperatures, precipitation rates and atmospheric transport parameters for four seasons.

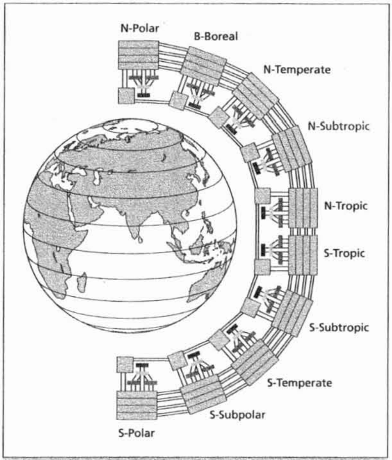


FIGURE 2.14

Schematic diagram of the Toronto model. Nine climatic zones, each containing a set of connected well-mixed compartments, are linked by meridional atmospheric and oceanic exchange (Wania 1994).

Consideration is given neither to particle-associated chemicals in the atmosphere and in surface water nor to particle-mediated transport processes such as dry particle deposition from the atmosphere or contaminant scavenging by the settling of suspended matter in the surface ocean. This may be an acceptable simplification in the case of HCHs, which due to relatively high volatilities and water solubilities, occur mostly in the gaseous or in the truly dissolved phase, but would require adjustments for less volatile or less soluble chemicals. Also, no land-to-ocean exchange is taken into account, which may be very important for river transported chemicals. Furthermore, ocean-atmosphere exchange is assumed to occur at ocean water temperatures, which are considered to be constant throughout the year and which often differ considerably from the temperature of the overlying atmospheric boundary layer. Since a sensitivity analysis revealed the Henry's Law constant (H, Figure 2.11) as one of the most influential input parameters, this is potentially a serious source of error.

The "Toronto" model (Figure 2.14) is essentially an expansion of a regional multimedia model of Mackay *et al.* (1992) designed to describe the fate of chemicals in smaller regions of area approximately 10^5 km^2. It consists of nine linked "unit worlds" containing air, fresh and marine water, cultivated and uncultivated soil, and fresh water sediments, each representing a zonally averaged climatic band similar to the zones defined in the Bergen model (Figure 2.15). Transport and exchange processes between these compartments as well as degradation and export to the deep sea are parameterized. The model includes phase partitioning of chemicals between air and aerosols as well as between water and suspended matter. However, there is no vertical subdivision of the atmospheric compartments, which makes the description of atmospheric transport processes very simplistic. Seasonally varying parameters such as temperature and atmospheric exchange rates are defined as sinusoidal functions. Recently the Toronto model has been applied to track the pathways of OCs through the global environment (Wania and Mackay 1996).

The Bergen model is superior in its treatment of advective and diffusive transport processes in the global atmosphere, while the Toronto model better succeeds in describing the reversible climate-dependent exchange processes between the atmosphere and the Earth's surface. Recently, Wania and Strand (1996) have combined the best parts of both approaches by incorporating into the Toronto-model a 2-D description of the atmosphere which adopts the vertical layering and the deduction of transport parameters from the Bergen-model. This "combined" model is the first fugacity-based model that includes stacked atmospheric compartments of variable density, and also gives special consideration to the treatment of vertical atmospheric transport and wet deposition processes. Temperatures in ocean water and the four atmospheric layers, as well as the vertical and horizontal atmospheric transport parameters are input as monthly averages. Figure 2.15 and Table 2.7 compare various characteristics of the three zonally averaged models.

2.2.4.2 Global Model Calculations for HCHs

Some insight into environmental pathways has already been obtained using these models. All three models have been used to describe the global fate of α- and γ-HCH in the past few decades. Strand and Hov (1996) used considerably lower emission estimates (1960–1989: 404 kt α-HCH and 147 kt γ-HCH) than the other two model simulations (1945–1993: 1964 kt α-HCH and 480 kt γ-HCH). More recent estimates (Table 2.4, Section 2.2.2) suggest a total global consumption between 1950–1992 of 5600

TABLE 2.7

A comparison of the characteristics of global zonally-averaged multi-compartmental toxics models.

	Bergen Model	Toronto Model	Combined Model
zonal subdivision	6 zones, equally spaced	9 zones, based on climatic criteria	10 zones, based on climatic criteria
compartments in each zonal "building block"	4 atmospheric layers (stratosphere, middle/upper and lower/middle troposphere, atmospheric boundary layer), surface ocean, deep ocean (subdivided into several layers), cultivated and uncultivated soil	1 atmospheric layer (entire troposphere), surface ocean, cultivated and uncultivated soil, fresh water, fresh water sediment	4 atmospheric layers (stratosphere, middle/upper and lower/middle troposphere, atmospheric boundary layer), surface ocean, cultivated and uncultivated soil, fresh water, fresh water sediment
treatment of atmospheric transport	meridional and vertical exchange described with horizontal and vertical advection velocities and eddy diffusion coefficients	meridional exchange described with latitude-dependent horizontal eddy diffusivities, lumping advective and diffusive processes	meridional and vertical exchange described with horizontal and vertical advection velocities and eddy diffusion coefficients
treatment of oceanic transport	modified High-Latitude-Exchange/Interior Diffusion-Advection Model (HILDA) by Siegenthaler & Joos 1992.	meridional exchange described with estimated horizontal eddy diffusivities, lumping advective and diffusive processes	
treatment of atmosphere-ocean exchange	diffusive gas exchange at ocean water temperature, dissolution in rain	diffusive gas exchange (at different air and ocean water temperatures), dissolution in rain, dry and wet particle deposition	
treatment of atmosphere-soil exchange	description of soil-atmosphere exchange based on Jury *et al.* 1983, 1984 a and b.	description of soil-atmosphere exchange based on Stiver and Mackay 1989	
treatment of seasonality	4 seasonal values for air temperature, precipitation, evaporation and atmospheric transport parameters	sinusoidal functions for air and ocean water temperature, sea ice coverage, and atmospheric exchange parameters	12 monthly values for air and ocean water temperature, sea ice coverage, and atmospheric exchange parameters
technicalities	zonally averaged continuity equation, step wise solution using finite difference approximations	54 linear mass balance equations in fugacity notation solved stepwise using finite difference approximation	90 linear mass balance equations in fugacity notation solved stepwise using finite difference approximation
test chemicals	α-HCH and γ-HCH	α-HCH and γ-HCH, DDT	α-HCH and γ-HCH
reference	Strand and Hov 1996	Wania 1994, Wania and Mackay 1995	Wania and Strand 1996

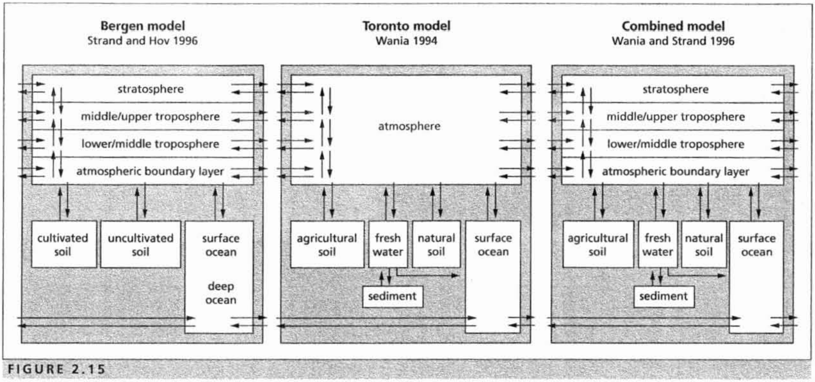


FIGURE 2.15

A comparison of the structure of the Bergen, Toronto and Combined zonally averaged global models.

kt technical HCH and 720 kt γ-HCH, corresponding approximately to 3920 kt α-HCH and 2400 kt γ-HCH (Section 2.2.2).

Figure 2.16 gives typical results of the "Toronto" model for lindane (γ-HCH) showing the seasonal variation (peaks) as well as the longer term trends in concentration in air and other media. Model results also show the latitudinal dependence of HCH concentrations in air and in sea water (Figure 2.17). Agreement with concentration data compiled by Bidleman (Atmospheric Environment Service, Canada, pers. comm.) is satisfactory, at least in an order of magnitude sense, but it must be appreciated that the reported data extend over a wide range. The fugacity models reproduce the pattern of relatively uniform atmospheric HCH concentrations within both hemispheres and the sharp concentration drop across the equator. They also capture the conspicuously high HCH concentration levels in high latitude sea water of the Northern Hemisphere. The Bergen model was less successful in reproducing the observed latitudinal concentration profiles, especially in sea water, but better predicts the atmospheric concentrations. The concentrations calculated using the fugacity models are too high, which quite likely is a result of overestimated HCH usage (approximately twice the estimate of Li 1996 and four times that used by Strand and Hov 1995). Even this degree of agreement is encouraging, especially considering the large uncertainty of the emission estimates used in these calculations.

One of the interesting results of this model comparison is that the inclusion of vertical atmospheric stratification in the model structure resulted in relatively minor changes in the calculated global fate of HCHs. The model without atmospheric stratification reproduced the latitudinal concentrations profiles in both atmosphere and surface ocean better than the Strand and Hov model, and is only slightly worse than the combined model. The conclusion to be drawn is that the temperature-dependent exchange between the atmosphere and the Earth's surface is more decisive for the global fate and distribution of HCHs than the overall atmospheric circulation.

2.2.4.3 The Three-dimensional AES model

A modelling study was undertaken by the Atmospheric Environment Service (AES 1997). It is a three dimensional multi-layer atmospheric model that uses objectively analysed winds (i.e. meteorological observations interpolated to a grid using fluid dynamic methods) and the Earth's surface physical properties (temperature, snow and ice cover) to describe the movement of OCs through the global atmosphere. The concentrations of an OC in surface reservoirs such as land, lakes and oceans are taken from observations, and emissions to the atmosphere are estimated from gridded pesticide use (Figs. 2.7 and 2.8) and soil-atmosphere exchange studies.

This model is currently being tested for HCHs according to the following scenario: The ocean is assumed to contain a prescribed constant concentration of α- and γ-HCH (see globally-gridded, surface-ocean HCH concentrations, Figure 2.18) and the initial atmosphere to contain no HCH. Then using 1993 meteorological observations, the model is run allowing exchange between the ocean and the atmosphere, and precipitation scavenging. Concentrations in the atmosphere are determined by the temperature dependence of Henry's Law constant as it affects both of these processes, as well as precipitation generated by the model. The seasonal variation of

HCH in air is predicted and compared to observations at Alert, Tagish and Dunay (Section 2.3.1). The purpose is to determine how much of the atmospheric concentration of HCH is controlled by ocean-atmosphere exchange. Then, the model is re-run as before but with the globally gridded HCH emissions of Li *et al.* 1996 (Figures 2.7 and 2.8) as a source to the atmosphere in addition to the ocean source. The purpose is to quantify the contribution of current annual usage to atmospheric HCH concentrations.

The detail in which atmospheric transport processes are handled in this model can be used to validate simpler parameterizations used in the Bergen and Toronto models. As an example of preliminary results, the global spatial distribution of α-HCH concentrations at ground level in the atmosphere are shown for December 1993 (Figure 2.18b). In the Arctic, observed concentrations are somewhat lower than the model predicted. However, this is likely due to higher ocean surface concentrations in the

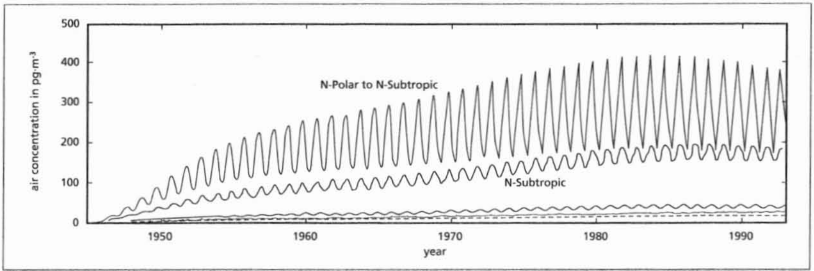


FIGURE 2.16A

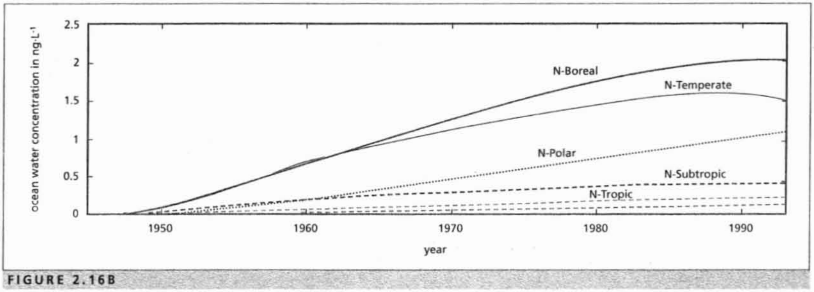


FIGURE 2.16B

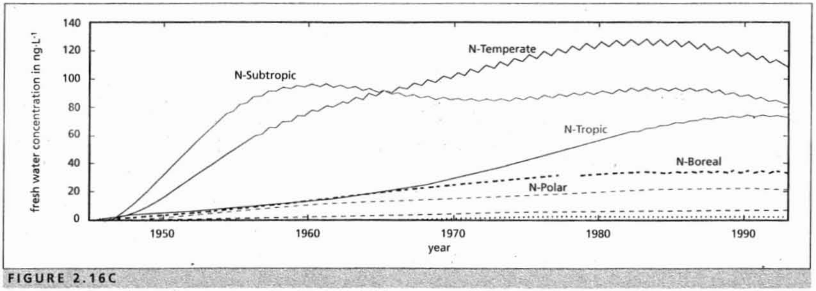


FIGURE 2.16C

Temporal variation of the zonal mean concentration of γ-HCH in air (A), ocean water (B), fresh water (C) as calculated by the Toronto global distribution model (Wania 1994).

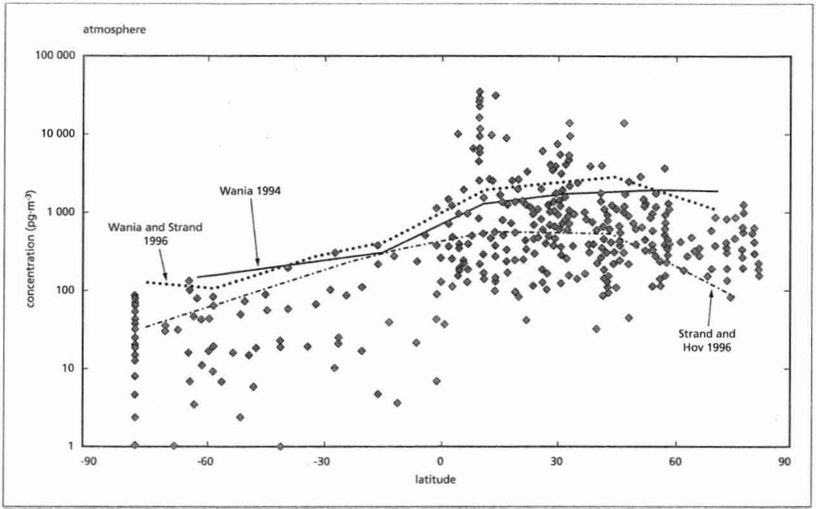


FIGURE 2.17A

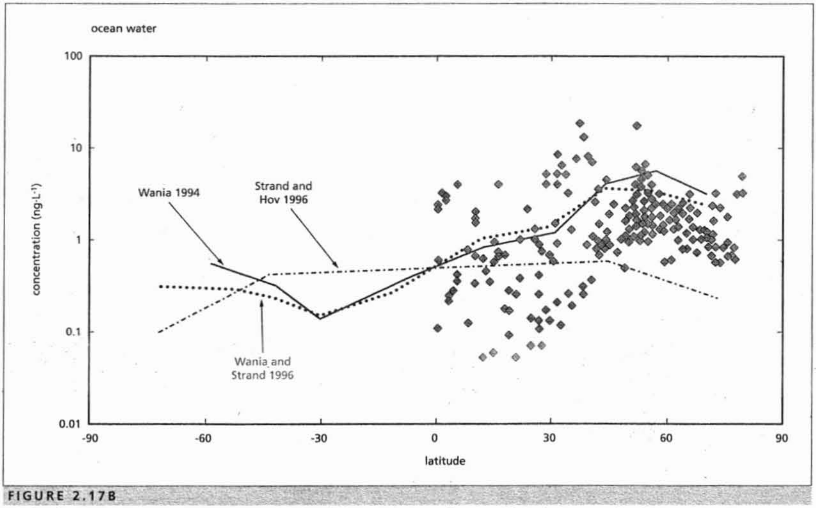


FIGURE 2.17B

Calculated and measured ΣHCH concentrations in (A) air and (B) ocean water as a function of latitude. Squares reflect measured data from different time periods compiled from various publications (Bidleman, T.F., pers. comm.). Lines represent the annual means for 1985 predicted by the Toronto model (solid line, Wania 1994), the Bergen model (dot-dash line, Strand and Hov 1996), and the combined model (dotted line, Wania and Strand 1996).

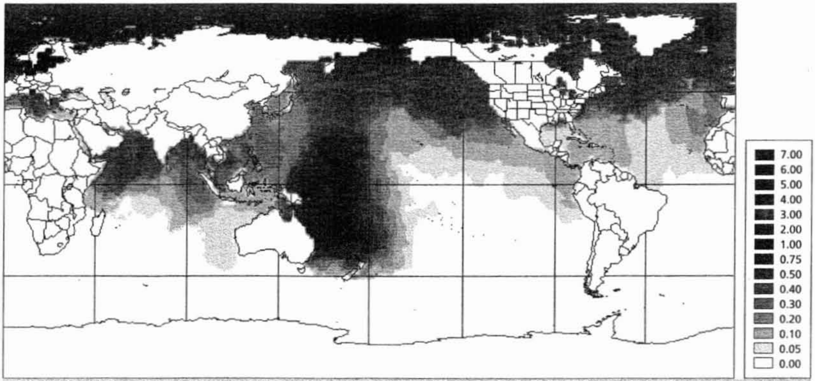


FIGURE 2.18A

Globally gridded distribution of observed ocean surface α-HCH concentration in ng·L^{-1}. The distribution shown is based on data compilation of Bidleman. The set of measurements from the sparse network was interpolated to the latitude-longitude grid (Pudykiewicz, pers. comm.). The distribution shown in this figure is representative of the state of the ocean surface around 1990.

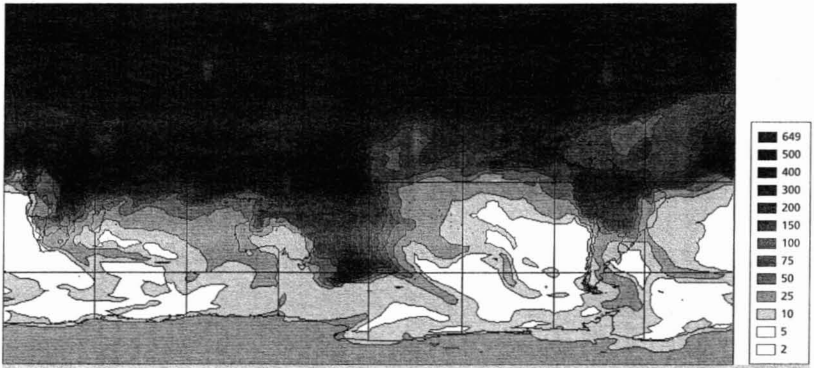


FIGURE 2.18B

Model predicted global distribution of α-HCH in picograms/cubic meter on December 30, 1993. The result shown was obtained after one year of simulation with a three-dimensional (3-D) atmospheric transport model using observed meteorological data (AES 1997; Pudykiewicz, pers. comm.) The model-predicted concentration in air is higher than observed because atmospheric destruction of HCH by OH has been ignored. When it is included concentrations are lowered substantially.

mid-1980s (Figure 2.18b) than in 1993 and also due to the neglect of atmospheric chemical destruction of HC by OH radicals. This last effect will be included in future model runs. This type of model will also be useful in interpreting differences in enantiomer ratios of α-HCH observed across the Arctic (see Section 2.6.1).

2.2.4.4 Summary

To date models confirm the existence and extent of significant long-range transport of contaminants to the Arctic as well as the global fractionation effect, in which chemicals differ in their propensity to reach the Arctic. If the global atmosphere is well mixed over the long term — as appears to be approximately the case for HCB and HCHs — then there will be higher concentrations of these contaminants in condensed media in the Arctic such as aerosols, soils, water and biota. This is because of the higher condensed media-to-air partition coefficients at low temperatures induced by lower vapour pressures and thus the increased tendency for condensation as is theoretically predicted by Clapeyron-Clausius relationships and Le Chatelier's Principle.

Although limited, the results obtained with the available models are useful in that, by forcing quantitative thinking on these global processes, they contribute to a fuller understanding of the factors influencing chemical fate and to the identification of those chemicals that may be subject to long-range transport and accumulation in cold climates.

- *Existing global models for persistent organics deal with the multi-compartmental Arctic ecosystem with varying degrees of complexity. In the NCP, two types of models are being used: (i) long-term 2-D low-spatial-resolution models and (ii) short-term, 3-D, high-spatial-resolution models. The 2-D models are excellent heuristic tools as well as able to address the policy question: "What is the fate of a persistent organic released over a period of decades?". They are highly parameterized and therefore can run for a 30-year simulation period very rapidly on available computers. 3-D models answer questions requiring resolution within a latitude zone that the 2-D latitude-zone averages cannot. They enable us to compare atmospheric contaminant observations for a particular date with model simulations for the same date. Furthermore, they can be used to test some parameterizations of processes that 2-D models are using. However due to their complexity, computing time prohibits their operation for periods of 30 years. The bottom line is that each model type plays an important role in answering questions about pathways and fate and we in Canada have access to them.*

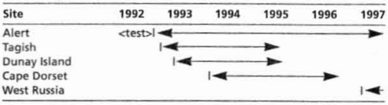
2.3 The Reservoirs: Occurrence and Internal Pathways

2.3.1 Atmosphere

Since January 1992, measurements of contaminants in air, including herbicides, pesticides, synthetic industrial compounds and PAHs, have been made on a weekly basis in the Canadian and Russian Arctic (Table 3.1). This research was supported by the AES-NCP and the Department of External Affairs. High-volume air samplers were placed at: Alert, NWT; Tagish, Yukon; Cape Dorset, Baffin Island; and at the mouth of the Lena River on Dunay Island in Russia (see Figure 1.1 for locations). A second site in western Russia is planned for spring 1997. Particulate and gaseous fractions of airborne contaminants were collected on filters and foam plugs. Samples were subsequently extracted in organic solvents and analysed at the Fresh Water Institute in Winnipeg for 29 OCs, many PCBs and 20 PAHs. The sampling schedule is shown in Table 3.1.

TABLE 3.1

The sampling schedule for the Canadian northern contaminants monitoring network.



2.3.1.1 Organochlorines

Table 3.2 presents a summary of the OC concentrations for 1993 and 1994 at Alert, Tagish and Dunay. Those compound classes comprised of several isomers and/or metabolites are reported as the sum of the individual components. The mean annual concentrations for 1993 are displayed graphically in Figure 3.1 and show the extent of spatial variation in air concentrations. Large spatial differences in the OC mean concentrations are not evident, indicating relatively widespread atmospheric contamination of the polar region on an annual basis. However, elevated mean concentrations of chlordanes, DDTs and endosulfan at Tagish may indicate the relative proximity of this site to North American and Asian source regions. Oehme *et al.* (1996) reported OC concentrations in the Norwegian Arctic throughout 1993, and found that elevated levels of *trans*-chlordane coincided with air mass movement primarily from North America. Similarly, veratroles (metabolites of chlorinated guaiacols and catechols released from bleached kraft mill effluents) have mean concentrations that decrease in the order of Tagish > Dunay > Alert, again probably indicating the proximity of these sites to this type of industry. OC concentrations range from almost 100 pg·m^{-3} for chlorobenzenes and HCHs, to less than 1 pg·m^{-3} notably for mirex, endrin and trifluralin (a fluorinated herbicide). Maximum concentrations at any of the sites do not exceed 1 pg·m^{-3}.

A more detailed analysis of the data from the Tagish site in the Yukon (Bailey *et al.* 1997) has shown a clear seasonality in air concentrations with several compounds showing a reasonable correlation with temperature. For example, heptachlor epoxide, oxychlordane and α-HCH displayed significant correlations with temperature. In other words, concentrations were elevated during the warmer months of the year and reduced during the cold periods. High concentrations of OCs that are associated with long-range atmospheric transport dampen this seasonal affect. For instance, elevated concentrations of *cis*- and *trans*-chlordane, oxychlordane, heptachlor epoxide, dieldrin, α-HCH, γ-HCH and *p,p*'-DDE were observed in the winter during the sample week of the 7–14th January 1993. Air mass back trajectories over this period revealed the source of the air from as far away as India, where DDT and HCH are still used for mosquito control in both agricultural and urban areas. Indeed, higher ratios of *p,p*'-DDE/(*p,p*'-DDT + *p,p*'-DDE), than those predicted by temperature for this period, are probably due to the conversion of applied *p,p*'-DDT to *p,p*'-DDE. This photodegradation process occurs after application in agricultural areas and/or during long-range transport.

Interestingly, source regions can be identified by air mass back trajectories and often accompany a change in OC profile. For instance, during the sample weeks of December 16th 1993 and January 27th 1994, concentrations of endosulfan were elevated, concentrations of hexachlorobenzene were reduced. Air mass back trajectories showed China and Siberia as the source regions. Bailey *et al.* (1997) summarized that reduced concentrations of HCB were probably due to there being no reported use of HCB in China. However, it must be noted that HCB is also released both as a by-product in the synthesis of organochlorine products and from the incomplete combustion of chlorinated wastes from the chemical industry (Muir, D.C.G., pers. comm.). Elevated levels of *p,p*'-DDE along with *o,p*-DDT were probably from the volatilization of past-use DDT. Furthermore, due to the distance of Tagish from source regions, more stable metabolites of many of the pesticides often prevail. For instance, the ratio of *trans*-chlordane

TABLE 3.2

Summary of OC concentrations (pg·m^{-3}) in the Arctic atmosphere — Alert, Dunay and Tagish (1993 and 1994).

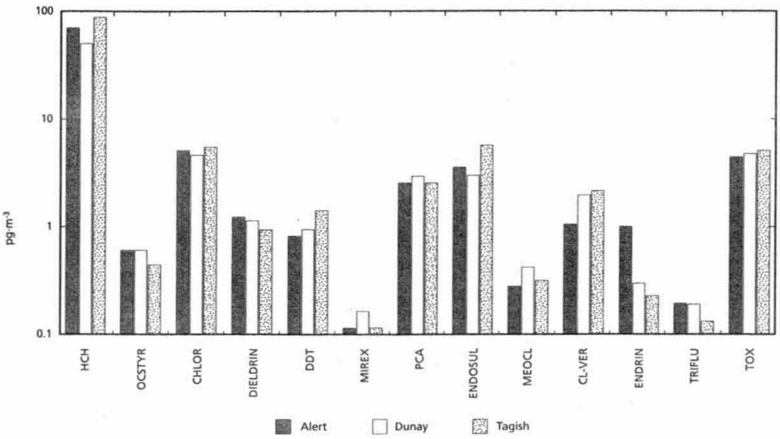
Alert														
1993	ΣCBz	ΣHCH	OCSTYR	ΣCHLOR	DIELD	ΣDDT	MIREX[1]	PCA	ENDOSUL	MEOCL	ΣCL-VER	ENDRIN	TRIFLU	ΣTOX
Mean	97.1	71.7	0.60	5.19	1.18	0.82	0.10	2.53	3.61	0.26	0.97	0.18	0.12	4.43
Min	13.5	15.2	0.10	0.65	0.06	0.12	0.03	0.01*	0.02*	0.07*	0.10	0.05*	0.03*	0.34
Max	205	145	1.47	17.2	3.18	2.24	0.26	8.19	9.42	0.72	1.95	0.50	0.32	16.4
1994[2]														
Mean	94.3	76.9	0.98	6.65	1.42	1.40	0.10	3.12	4.89	0.28	1.44	0.20	0.13	5.33
Min	5.08	7.22	0.20	1.24	0.03*	0.25	0.07	0.17*	0.07*	0.18	0.24	0.13*	0.08	1.09
Max	526	369	2.96	23.1	4.16	6.79	0.52	20.5	16.2	1.43	10.2	0.99	0.64	12.8
Dunay														
1993	ΣCBz	ΣHCH	OCSTYR	ΣCHLOR	DIELD	ΣDDT	MIREX	PCA	ENDOSUL	MEOCL	ΣCL-VER	ENDRIN	TRIFLU	ΣTOX
Mean	85.7	50.9	0.60	4.55	1.11	0.93	0.16	2.92	2.99	0.41	1.93	0.28	0.18	4.70
Min	21.2	0.71	0.11	0.40	0.02*	0.24	0.08	0.95	0.05*	0.22	0.30	0.15	0.10	1.05
Max	228	93.2	1.74	10.3	2.04	2.02	0.28	6.92	7.18	0.74	5.85	0.51	0.33	12.4
Tagish														
1993	ΣCBz	ΣHCH	OCSTYR	ΣCHLOR	DIELD	ΣDDT	MIREX	PCA	ENDOSUL	MEOCL	ΣCL-VER	ENDRIN	TRIFLU	ΣTOX
Mean	52.7	91.3	0.44	5.63	0.91	1.39	0.11	2.56	5.76	0.31	2.17	0.22	0.13	5.12
Min	12.6	27.6	0.08*	1.98	0.04*	0.26	0.05*	0.09*	0.08*	0.13*	0.34*	0.09*	0.06*	1.42
Max	196	178	1.31	10.4	2.15	3.21	0.17	5.11	15.3	1.12	6.97	0.44	0.21	14.6
1994[3]														
Mean	44.0	80.4	0.69	5.19	0.81	1.51	0.10	2.25	6.58	0.28	2.05	0.2	0.12	5.35
Min	13.9	0.52*	0.10	0.71	0.04*	0.25	0.06	0.04*	0.08*	0.17	0.23	0.11*	0.07	1.13
Max	88.3	166	1.65	7.61	2.83	3.23	0.14	5.27	13.9	0.38	6.94	0.47	0.17	10.5

[1] *including photo-Mirex*

[2] *up to week ending the 26th September*

[3] *up to week ending the 25th August*

* *below MDL*



HCH = Σhexachlorocyclohexanes
OCSTYR = octachlorostyrene
CHLOR = Σchlordanes
DDT = ΣDDTs
PCA = pentachloroanisole
ENDOSUL = endosulfan
MEOCL = methoxychlor
CL-VER = chloro-veratroles
TRIFLU = trifluralin
TOX = Σtoxaphene

FIGURE 3.1

Mean annual organohalogen pesticide concentrations (pg·m^{-3}) for 1993 (measurements taken on a weekly basis).

(TC)/*cis*-chlordane (CC) is ~0.8 during the colder months and drops further during the warmer months to ~0.2. As a comparison, technical chlordane has a ratio of TC/CC of 1.26, the lower ratios in Tagish air representing the loss of the less stable TC. Application of chlordane for agricultural uses in Asia would likely occur during the winter months and reflect the fact that ratios of TC/CC at Tagish for this time of year are closer to the technical mixture. The lower ratio of 0.2 during the warmer months may indicate a more aged source, possibly the volatilization of previously deposited chlordane. This is supported by the fact that the ratio of oxychlordane/TC actually increases at Tagish during the warmer months. Suprock *et al.* (1980) have shown that *trans*-chlordane is primarily removed in soils by transformation to oxychlordane.

A separate Norwegian study at Ny Ålesund on Spitzbergen Island has allowed for a direct comparison of OC observations from the Northern Contaminants Programme. Oehme *et al.* (1996) carried out air sampling for a 48h period every week for the greater part of 1993, where sampling commenced in April (week number 13) and finished in December. Figures 3.2a and b compare the weekly atmospheric concentrations of α-HCH and γ-HCH, respectively, at both Alert and Ny Ålesund. Mean concentrations (pg·m^{-3}) of both isomers were higher at Ny Ålesund (mean $\alpha = 78.4 \pm 5.3$, $\gamma = 14.2 \pm 1.1$) than at Alert (mean $\alpha = 60.4 \pm 3.5$, $\gamma = 10.6 \pm 1.0$). Furthermore, summarizing the whole period, Alert displayed a higher α/γ-HCH ratio (6.3 ± 1.7) than Ny Ålesund (5.8 ± 1.9), indicating (along with the lower air concentrations) that Alert is less influenced by regional sources on an annual basis than Ny Ålesund. Elevated inputs of γ-HCH at Ny Ålesund can be observed in Figure 3.2b for weeks 19 and 30, however the α/γ-HCH ratio does not change significantly. Oehme *et al.* (1996) suggested that this was due to air moving from a source region such as Russia or Asia, where technical HCH (85% α-HCH, 10–15% γ-HCH) is still in use, rather than Europe where application of pure γ-HCH (Lindane) still occurs. At Alert, the lowest ratios occur in mid May for weeks 19 and 20 (α/γ-HCH = 2.9), indicating fresh inputs of γ-HCH into the Canadian high Arctic at this time of year.

2.3.1.2 Polychlorinated biphenyls

The weekly ΣPCB (90 congeners) concentrations are displayed in Figure 3.3 for the Alert, Tagish and Dunay sample sites throughout 1993 and 1994. Mean concentrations for 1993, the year when all three sites were running concurrently, were 27.9, 17.2 and 34.2 pg·m^{-3} at Alert, Tagish and Dunay respectively. Oehme *et al.* (1996), monitoring PCBs at Ny Ålesund, Spitzbergen, reported a mean annual (1993) ΣPCB concentration of 13.1 pg·m^{-3}. In this study only a subset of 10 congeners were reported. The equivalent annual mean concentrations for Alert, Tagish and Dunay, using the same 10 congeners, were 6.04, 3.70, 8.15 pg·m^{-3} respectively. From Figure 3.3, seasonal variations in the total concentrations were not evident. This is in contrast to studies carried out in more southerly latitudes, where a summer maximum has been observed (Hoff *et al.* 1992, Halsall *et al.* 1995). Reasons for the lack in seasonality in the north are not clear, but may be related to the fact that these arctic sites are well removed from temperate source areas. Oehme *et al.* (1996) argued that temperatures at Ny Ålesund may not exceed a threshold whereby significant volatilization off surfaces occurs. This scenario may also be applicable to the high Arctic sites of Alert and Dunay of the Canadian Programme,

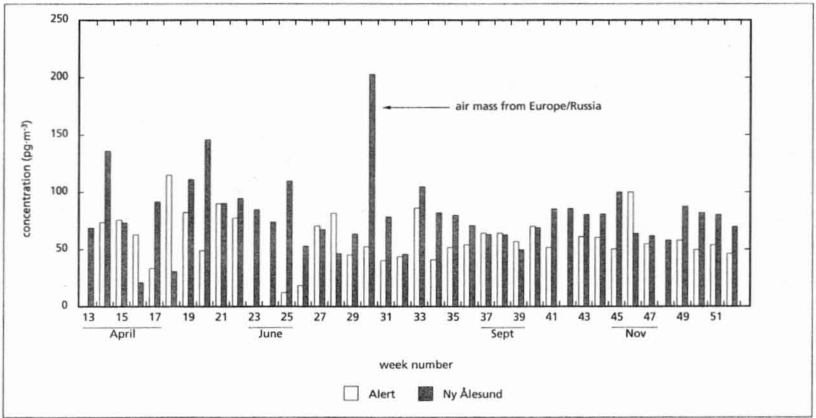


FIGURE 3.2A

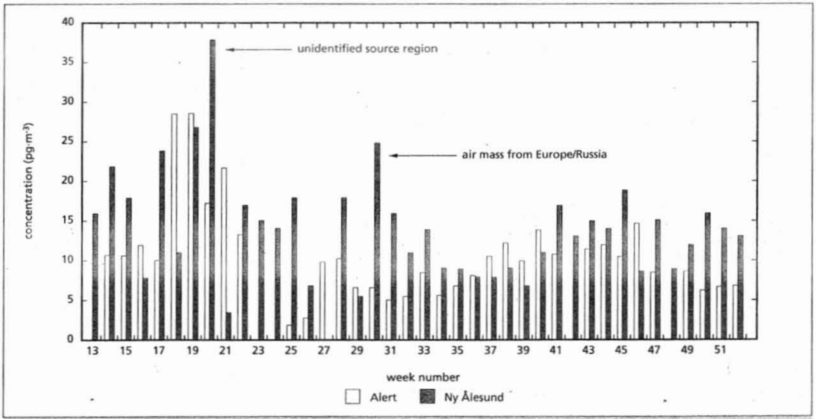


FIGURE 3.2B

Weekly (A) α-HCH and (B) γ-HCH concentrations (pg·m^{-3}) at Alert and Ny Ålesund, Spitzbergen. April–December 1993.

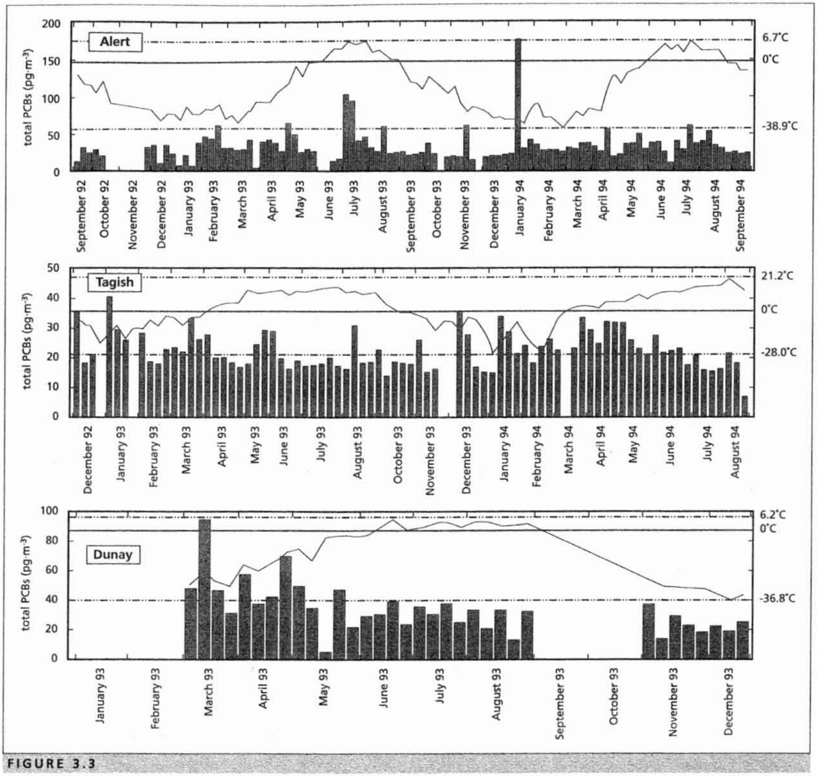


FIGURE 3.3

Weekly total PCB concentrations (pg·m⁻³) at Alert, Tagish and Dunay.

but does not explain the lack of seasonality at Tagish, where temperatures often exceed 20°C during the warmer months (Figure 3.3). Characteristics of the Tagish site, given its location in the Pacific North-West and its higher elevation (~1500 m) relative to the other sites, may be important factors in explaining this anomaly. Nevertheless, the lower temperatures in the Arctic region as a whole, compared with those in mid-latitudes, coupled with the fact that contamination of surface soil, snow and ice is considerably less than in temperate regions, may help to explain the lack of seasonality in concentrations of contaminants in the air.

Figure 3.4 presents a summary of PCB homologue concentrations on a monthly basis. The lighter tri-chlorinated homologue is dominant at each of the sites. Notable exceptions to this occur at Dunay from May to July 1993, where the profile is dominated by the heavier pentachlorinated homologue. Air mass transport is believed to play an important role in this phenomenon. Indeed, at Ny Ålesund, the incursion of air masses from the southerly regions of Europe/Russia accounted for the majority of sample weeks when PCB concentrations were elevated (Oehme *et al.* 1996). One notable contamination episode occurred at Alert during the 10th to the 17th of January 1994 (Figure 3.3). During this period the profile was dominated by the lighter di-, tri-, and tetra-homologues. The air during this period came directly from central Siberia and the island of Novaya Zemlya.

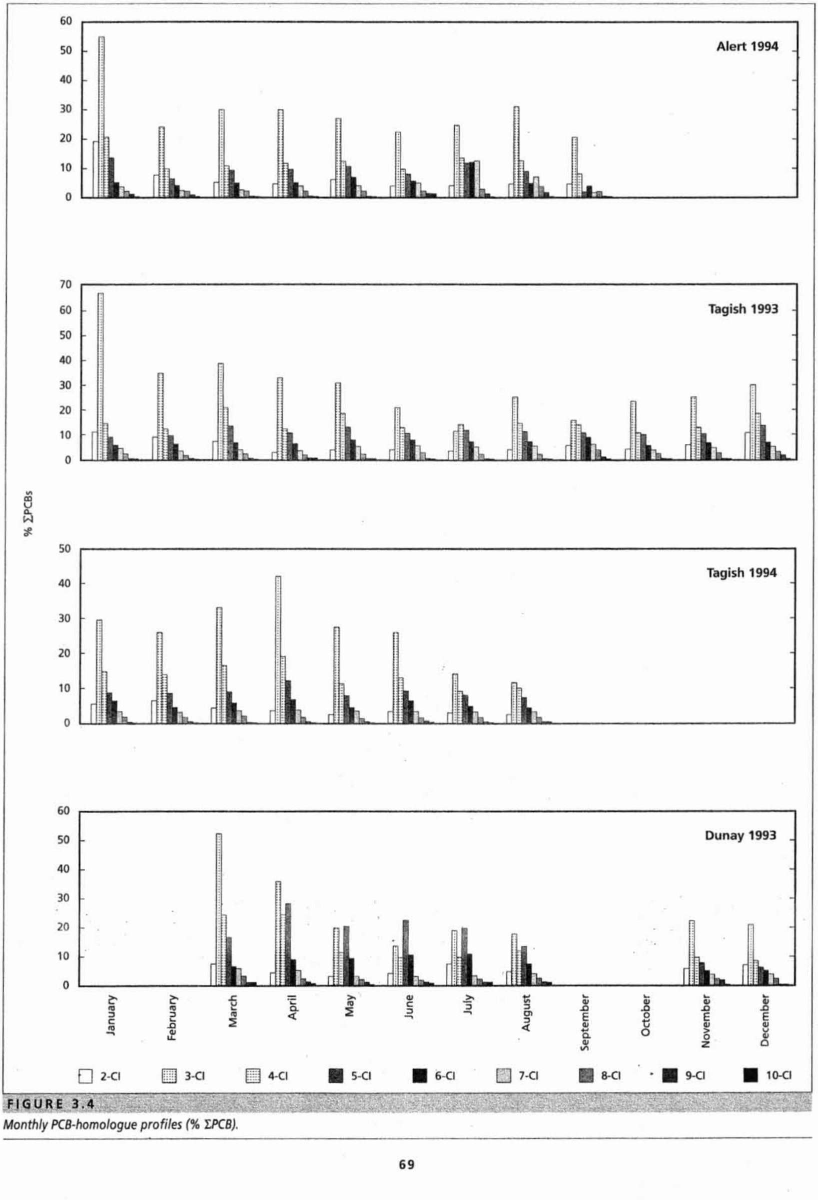


FIGURE 3.4

Monthly PCB-homologue profiles (% ΣPCB).

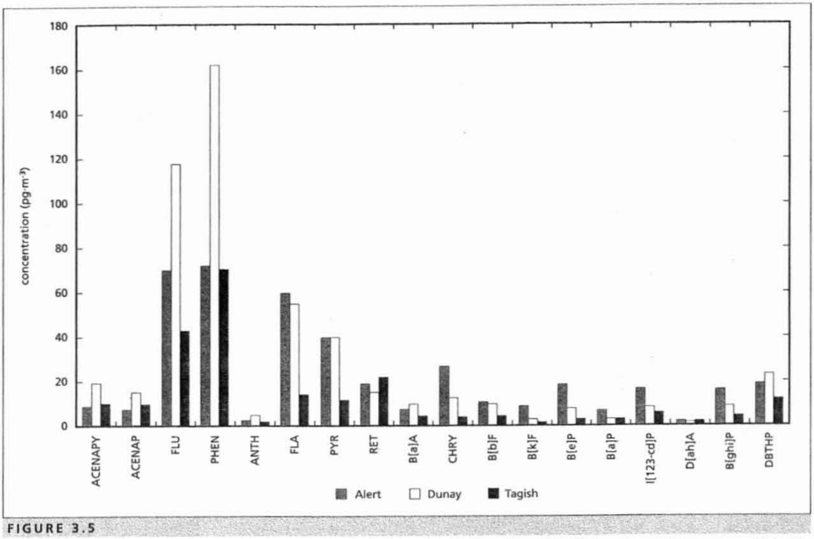


FIGURE 3.5

Geometric mean annual PAH air concentrations (vapour and particulate) for Alert, Tagish and Dunay in 1993 (samples were taken on a weekly basis) (See Figure 1.3 for explanation of PAH structure).

2.3.1.3 PAHs

Following several studies in the Norwegian Arctic input (Pacyna and Oehme 1988) and over the Greenland ice cap (Masclet *et al.* 1993), PAH contamination in the Arctic environment is now believed to be dominated by atmospheric. Ice cores from the Greenland ice cap have shown a dramatic increase in PAH concentrations over the last 100 years, which is in good agreement with the historical record of world petroleum production (Kawamura *et al.* 1994). Similarly, Jaffrezo *et al.* (1994) examining surface snow samples, also on the Greenland ice cap, reported that PAH contamination was essentially due to fossil fuel combustion with some input from biomass burning.

The annual geometric mean ΣPAH air concentrations measured at the Alert site were 465 (1992), 444 (1993), and 330 pg·m^{-3} (1994), while the Tagish site had concentrations of 194 and 249 pg·m^{-3} for 1993 and 1994 respectively. Dunay, in eastern Siberia, displayed the highest annual mean concentration of 508 pg·m^{-3} for 1993. (ΣPAH = 16 compounds: acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno [1,2,3-c,d]perylene, dibenzo[a,h]anthracene, benzo[g,h,i]perylene). The more southerly Tagish site experienced the lowest mean annual concentrations out of the three sites. For both 1993 and 1994 the mean ΣPAH concentrations were lower than Alert by factors of 2.2 and 1.3 respectively. Although naphthalene was found to be the most abundant PAH in the Arctic atmosphere it was excluded from these mean values due to its probable underestimation resulting from a sampling artifact.

Compared with earlier Arctic studies, Patton *et al.* (1991) reported a mean concentration of 542 pg ΣPAH·m^{-3} (excluding dibenzofuran and biphenyl) for 10 samples taken between February and April 1988 in the Canadian high Arctic at Alert. Pacyna and Oehme (1988) reported mean concentrations of fluoranthene at Ny Ålesund, Spitzbergen in the Norwegian Arctic of 57 and 48 pg·m^{-3} for samples taken in the winter/spring of 1983 and 1984 respectively. Mean fluoranthene concentrations at Alert ranged from 22.1 to 106 pg·m^{-3} throughout the sampling period from 1992 to 1994.

Figure 3.5 presents a summary of the individual PAH air concentrations (vapour plus particulate) at the Alert, Dunay and Tagish sites for 1993. Similar to the observations of PAHs in remote temperate regions (McVeety and Hites 1988, Baker and Eisenreich 1990), the annual mean PAH mixture at the three sites was dominated by the lighter compounds of fluorene and phenanthrene, with significant contri-

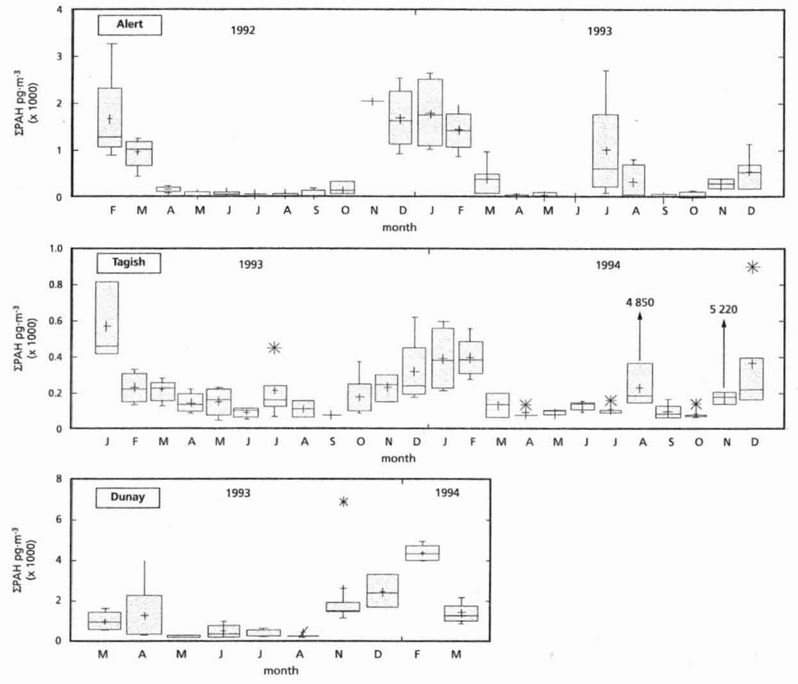


FIGURE 3.6

Box-and-whisker plots of ΣPAH concentrations in air (pg·m⁻³) (ΣPAH = 16 compounds) on a monthly basis for Alert, Tagish and Dunay. In each box the horizontal line represents the median and the cross represents the mean. Outlier weeks are represented by an asterisk. Each month comprises four weekly samples.

butions by fluoranthene and pyrene. An interesting point to note is that the geometric annual means (atmospheric PAH concentrations have a log-normal distribution) have the effect of smoothing elevated concentrations associated with events or episodes. For example, retene (an alkylated three ringed PAH, 1-methyl-7-isopropylphenanthrene) found to be predominantly produced from soft wood combustion (Ramdahl 1983), comprised >70% of the ΣPAH concentration in the Tagish atmosphere during August and November 1994. During the sample weeks starting August 11th, 18th, and November 24th, retene concentrations exceeded 1 ng·m⁻³. Elevated concentrations of PAHs in the Tagish atmosphere during this period, marked in particular by retene, are believed to be the result of forest fires.

Figure 3.6 displays a box-and-whisker plot of monthly ΣPAH concentrations in air (ΣPAH = 16

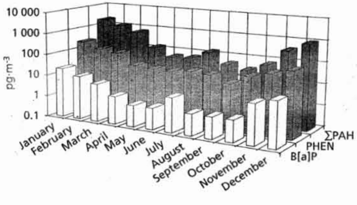


FIGURE 3.7

Monthly geometric mean concentrations of ΣPAH, phenanthrene (PHEN) and benzo[a]pyrene (B[a]P) at Alert (n = 3; 1992/93/94). (Note that July is elevated due to high concentrations occurring during July 1993).

compounds) for Alert, Tagish and Dunay. Seasonal fluctuations in air concentrations were clearly evident, with the colder months of the year (October–April) displaying higher concentrations than the warmer months (May–September). At Alert, the mean ΣPAH concentration during the cold period was an order of magnitude higher than the warmer season. Indeed, the colder months account for the majority of the annual PAH loading in the Arctic atmosphere. These elevated concentrations coincide approximately with the haze period, typified by an input of anthropogenic-derived aerosol into the Arctic troposphere (Barrie and Bottenheim 1991). Seasonal variability is illustrated in the comparison of the geometric mean concentrations of phenanthrene, B[a]P and ΣPAH at Alert (1992 to 1994) (Fig 3.7). All three compounds have a similar seasonal variation with elevated concentrations during the colder months. Although the winter haze period consistently displays the highest concentrations for all three sites, episodes can occur at other times of the year. As discussed previously, elevated PAH concentrations were observed in the Tagish atmosphere, not only in the winter haze period, but also during August and November 1994 (marked on Figure 3.6). Elevated PAH concentrations were also noted for the sample week of the 15th of July, 1995 at Tagish. The PAH profile was dominated by retene (~90%) (an indicator of wood combustion), indicating forest fires as the possible source.

Elevated PAH concentrations were also detected at Alert during July and early August 1993 (Figure 3.6), in particular the sample week beginning the 19th July, when a ΣPAH concentration of 2.73 ng·m^{-3} was observed. Elevated concentrations during this period were due to local contamination from the combustion of waste fuels and materials at a nearby military base.

In order to examine PAH contamination during the haze period, air mass back trajectories were constructed for February 1994. Figure 3.8 presents the mean air flow direction to each of the sample sites for February 1994. On each map, the five circles represent the consecutive days of the 5-day back trajectories carried out every six hours throughout the month. Interestingly, PAH concentrations are highest in the order of Dunay > Alert > Tagish. Both Dunay and Alert are clearly influenced by air moving from

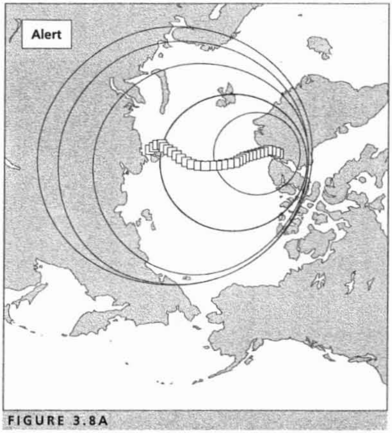


FIGURE 3.8A

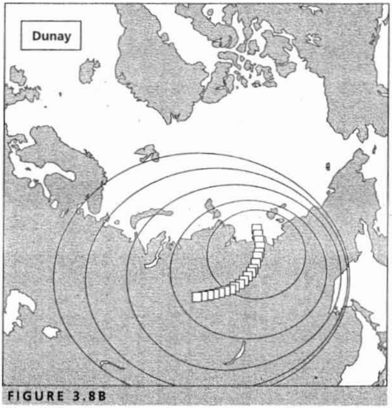


FIGURE 3.8B

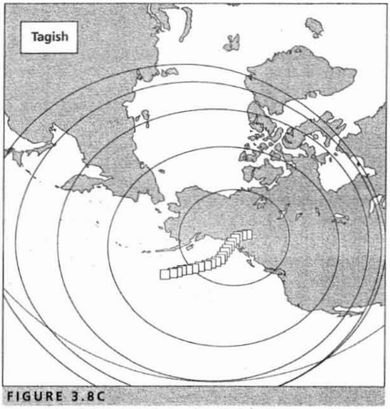


FIGURE 3.8C

Mean air flows for the three sites during February 1994. Five-day back trajectories carried out at the 925 hPa level for Alert and Dunay and 700 hPa for Tagish (an elevated site ~1500 m).

source regions in central and western Eurasia, Tagish on the other hand is influenced by air from the North Pacific region. Figure 3.9 presents the February mean concentrations of benzo[a]pyrene and phenanthrene for the three sites. Both benzo[a]pyrene (~100% particulate bound) and phenanthrene, (~50% particulate bound) show a marked decrease moving from Dunay to Tagish by factors of ~15 and ~10, respectively.

2.3.1.4 Mercury

It has been suggested that anthropogenic releases of Hg to the atmosphere have caused a three-fold increase in its concentration in air and marine surface waters since the pre-industrial era (Mason *et al.* 1994). This increase has been observed in arctic lake sediments (Section 2.5.5). Prior to the NCP there were no atmospheric measurements of Hg in the Arctic. In 1992/3, weekly-integrated sampling of total gaseous Hg° in air was undertaken at Alert, NWT (Schroeder 1996, Schroeder *et al.* 1995). Results from this preliminary study provided the first such data set for the Canadian Arctic (Figure 3.10). Observed air concentrations ranged from 0.67 to 2.8 ng·m^{-3} with an arithmetic mean of 1.5 ng·m^{-3}. These concentrations are less than those observed in "background" situations in southern Ontario (Schroeder and Markes 1994) and are comparable with those measured in the winter of 1993 for the northern Greenland Sea and Fram Strait (i.e., >90% samples <1 ng·m^{-3} Hg, Schroeder *et al.* 1995). In Antarctic air during the period 1987 to 1989 Hg concentrations were on average three times lower than in the Arctic (range: 0.02–1.9 ng·m^{-3}; mean: 0.55 ng·m^{-3}, De Mora *et al.* 1993). New technology for measuring atmospheric Hg has enabled sampling at a high level of time resolution (0.5 h). Continuous observations over the period January–April 1995 indicate that the mean concentration of total gaseous Hg in air is approximately 1.5 ng·m^{-3}, in excellent agreement with the 1992/93 results.

2.3.1.5 Radionuclides

Radionuclides in the environment may be of natural or artificial origin. Natural radionuclides include potassium-40 (^{40}K) and members of the uranium and thorium decay series. They form a significant part of the natural radiation background that humankind has always been exposed to. Environmental levels of these radionuclides may become enhanced as a result of human activities such as uranium mining.

Artificial radionuclides in the environment are mainly products of nuclear fission. During the 1950s and 1960s a great deal of this material entered the Arctic as fallout from the atmospheric testing of nuclear weapons (Barrie *et al.* 1992). The signing of the Limited Test Ban Treaty in 1963 did much to reduce this source of contamination; however, France and China were not parties to the treaty and continued atmospheric testing until 1980. Underground testing continued until 1996 and this process may occasionally release small amounts of radioactivity to the surface. The signing of the Comprehensive Test Ban Treaty on September 24, 1996, effectively put an end to the testing of nuclear weapons.

Fission products may also enter the Arctic environment as a result of large-scale releases from nuclear facilities. The Chernobyl reactor accident of 1986 injected fresh fallout into the Canadian Arctic;

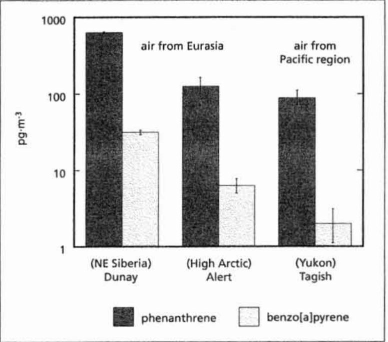


FIGURE 3.9

Mean concentrations (pg·m^{-3}) of phenanthrene (PHEN) and benzo[a]pyrene (B[a]P) for February 1994 at the three Arctic sites (samples take on a weekly basis).

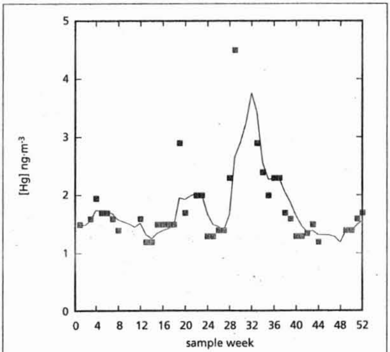


FIGURE 3.10

The mean seasonal variation of total gas phase Hg concentration in air at Alert based on weekly measurements from August 1992 to July 1993 (adapted from Schroeder et al. 1995). The fitted line is a 5-point running mean of weekly mean observations.

the effects were much less, however, than they were in northern Europe (Barrie *et al.* 1992). An accident at the Tomsk re-processing facility in Siberia in 1993 released traces of fission products, some of which were detected at Resolute, NWT, and Barrow, Alaska. Trends in fission product concentrations across Canada have been reported by Létourneau *et al.* (1994).

Health Canada routinely analyses air filters from Arctic monitoring stations to characterize radionuclide concentrations. Fission products are not normally seen on these samples, however, strong seasonal trends are observed in concentrations of natural beryllium-7 (^{7}Be) and lead-210 (^{210}Pb). As well as contributing to the natural radiation background, these two radionuclides serve as tracers of atmospheric movement. Although the seasonal variations in ^{210}Pb concentrations are better defined than for ^{7}Be, concentrations of both peak in winter, and decline during the summer months. Observed concentrations range from about 0-2 mBq·m^{-3} for ^{210}Pb, and 0.5-7 mBq·m^{-3} for ^{7}Be.

- *For the first time, a database of at least 2 years of routine observations of the important OCs, PAHs and Hg in the atmosphere exist in the Arctic at Alert, NWT.; Tagish, Yukon; Cape Dorset, NWT; and Dunay Island, Russia. Ranking OCs according to their mean annual concentrations in air yields the following abundance order: ΣHCH> ΣPCB> toxaphene≈Σchlordane≈endosulfan > pentachloranisole> chloroveratroles> ΣDDT≈dieldrin> octachlorostyrene >methoxychlor≈endrin> trifluralin ≈mirex. Many compounds (e.g. toxaphene) have a strong seasonality in their atmospheric concentrations that is likely related to the temperature dependence of phase partitioning between the atmosphere and Earth's surface.*

TABLE 3.3

The location and schedule of snowpack and snowfall chemistry studies conducted in the Canadian Arctic. (see Figure 3.11 for site locations). X = snowpack core measurement near end of snow season; C = routine weekly snowfall chemistry.

	1990	1991	1992	1993
Western and Central Arctic				
Peel River above Ft. Mcpherson (67°13'N 134°57'W)	X		X	X
Nahanni River near Nahanni Butte	X		X	X
Great Bear River (65°8'N 123°31'W)	X			
Great Bear Lake at Hornby Bay (66°36'N 117°37'W)		X	X	X
Ellice River near mouth (67°42.5'N 104°8.5'W)	X	X	X	X
Akasta River	X	X	X	X
Thonokied River (WRB site discontinued)			X	
Cameron River below Reid Lake (62°29'N 113°31'W)	X	X	X	X
Kakisa River	X		X	X
Mackenzie River near Ft. Good Hope		X	X	X
Snare Rapids			XC	
Keewatin Arctic				
Baker Lake (VOR Lake) (64°18'N 96°5'W)	X	X	X	XC
Yathkyed Lake (62°42'N 98°18'W)	X		X	X
Hayes River (67°32'N 94°5'W)	X	X	X	X
Back River below Deep Rose Lake (66°5'N 96°30'W)		X		X
Brown River (66°2'N 91°50'W)	X		X	X
Thelon River above Thelon Bluffs (64°32'N 101°24'W)	X	X	X	X
Lorillard River (64°15'N 90°26'W)	X	X	X	
Seal Hole		X	X	
High Arctic				
Stanwell Fletcher Lake, Somerset Island	X	X	X	X
Lake Hazen, Ellesmere Island		X		
Agassiz Ice Cap, Ellesmere Island (80°40'N 73°30'W)	X	X	X	X
Unnamed Lake near Resolute Bay (74°42'N 94°54'W)		X	X[1]	
Mould Bay, Prince Patrick Island (76°15'N 119°16'W)	XC	XC	XC	XC
Eureka, Ellesmere Island (80°00'N 86°36'W)	XC	XC	X[2]	X
Alert, Ellesmere Island	X		C	XC
Lady Melville Lake (68°38'N 92°30'W)	X	X	X	X
Chartrand Lake (near Spence Bay)	X	X	X	X
Penny Ice Cap, Baffin Island		X	X	
Iqaluit			X	
Yukon				
White Pass			XC	X
Tagish			XC	X
Beaver Cr.			X	X
Buchanan Cr.			X	X
Dawson City			XC	X
Watson Lake (60°4'N 128°46'W)				X
Teslin (60°9'N 132°40'W)				
Faro (62°12'N 133°32'W)				
Ross River (61°57'N 132°29'W)				X

[1] *This site has been replaced by the Amituk Lake site (about 50 km to the east) which is reported as part of that study.*

[2] *The snow collector at Eureka was moved to Alert to be co-located with the high volume air sampler.*

Note: A number of sites were missed in 1991 due to lack of funds at the end of FY 1990/1991 (sampling had to be done in March in the west) as well as to weather problems in the Keewatin. Also, stations such as Lake Hazen and Alert are collected only as opportunities permit due to the high cost.

- *ΣPCB concentrations in the arctic atmosphere do not vary much seasonally. This contrasts with a summer maximum observed at a mid-latitude site. The composition of the complex PCB mixture of compounds differs between the North American Arctic and the Russian Arctic. Heavier more chlorinated compounds are found in Russia.*
- *ΣPAHs in the Arctic peak in the winter months of December to February overlapping the Arctic haze season. In the high Arctic, the most abundant PAH compounds in air are fluorene, phenanthrene, fluoranthene and pyrene.*
- *Mercury concentrations at Alert average 1.5 nanograms per cubic meter of air with little seasonal variation. This concentration is three times higher than concentrations observed in Antarctic air. Natural and anthropogenic sources of atmospheric Hg are greater in the northern hemisphere than the southern hemisphere.*

2.3.2 Snowpack and Snowfall

2.3.2.1 Snowpack

Because a significant proportion of annual precipitation in the Arctic occurs as snow, it can play an important role in contaminant deposition to ecosystems. The efficiency of scavenging semi-volatile OCs and PAHs from the atmosphere differs for snow and rain (Section 2.4.1). Through the course of the winter, the snowpack receives contaminants from the atmosphere from both wet and dry deposition and loses them by volatilization, relocation due to wind, and meltwater percolation. Consequently, the contaminant burden of a snowpack is likely different from that in accumulated fresh snows. Snowpack melt is the major hydrologic event in northern regions and a knowledge of its contaminant burden (B) is essential in understanding the atmospheric input of contaminants to surface waters of the Arctic.

The history of snowpack observations in the Arctic was reviewed in Barrie *et al.* (1992). From 1990/91 through to the end of the winter of 1992/93, the NCP continued to support an expanded snowpack chemistry measurement program.

Sampling sites in the NWT and Yukon included in the NCP are listed in Table 3.3 and shown on the map in Figure 3.11. Average concentrations in snowpack samples collected in late winter of 1991/92, are reported in Table 3.4. The implications to exchange of contaminants between compartments (Figure 3.12) are discussed in Section 2.4.1. In general, ΣHCH (sum of α-HCH and γ-HCH concentrations) is considerably higher in the eastern part of the mainland region (on or near the Boothia-Somerset Island peninsula) than in other areas. Part of the variability in the results and the attendant difficulty in interpreting patterns is due to intra-site variability. Detailed studies at Amituk Lake (Section 2.3.4) show that deeper snow retains its burden of the more volatile OCs (e.g. HCH) better than shallow snow. The choice of a gully versus a hummock in sampling can make a significant difference to the result.

TABLE 3.4

Concentrations of contaminants in snow ($ng \cdot L^{-1}$) from the 1991/1992 snowpack survey.

Location	ΣHCH	ΣChlord	ΣDDT	HCB	ΣPCB
Peel R.	0.39	<0.01	<0.01	0.07	3.6
Ft. Good Hope	0.03	0.05	0.07	0.06	1.9
Great Bear L.	0.13	0.11	0.03	0.23	3.6
Nahanni Butte	0.41	<0.01	0.03	0.24	2.1
Slave R.	0.09	<0.01	0.03	0.06	1.9
Kakisa R.	0.11	<0.01	0.02	0.26	4.2
Reid L.	0.19	0.02	0.02	0.13	3.4
Akasta R.	0.21	<0.01	0.03	0.16	1.9
Back R.	0.02	0.03	<0.01	0.20	0.9
Baker L.	0.90	0.01	0.10	0.11	3.3
Yathkyed L.	0.23	0.02	0.02	0.38	1.6
Thelon R.	0.50	<0.01	0.05	0.22	3.3
Brown R.	0.80	0.03	0.10	0.25	3.1
Lorillard R.	0.21	<0.01	0.02	0.20	3.3
Seal Hole L.	0.70	0.11	0.04	0.13	2.2
Ellice R.	0.43	<0.01	0.06	0.31	4.8
Hayes R.	3.7	0.09	0.14	0.18	3.6
Stanley Fletcher L.	1.87	0.14	0.39	0.04	6.7
Chartrand L.	2.1	0.17	0.42	<0.01	9.0
Lady Melville L.	0.77	0.01	0.20	0.04	13.
Penny Ice Cap	2.3	<0.01	0.12	0.04	9.0
Averages	0.76	0.04	0.09	0.16	4.1
Standard Deviation	0.93	0.05	0.11	0.10	2.9

ΣHCH (α- plus γ-HCH); ΣDDT (total of DDT related residues: *p,p'*- and *o,p'*- isomers of DDT, DDE and DDD); ΣCHLOR (total of α- and γ-chlordane and *trans*-nonachlor). ΣPCB- the number analysed changed from 69 (the above data) to 116 congeners for data in 1994. The change increased ΣPCB by approximately 70% (see Section 2.3.4, Table 3.11).

TABLE 3.5

Snowfall-amount weighted-mean concentrations ($ng \cdot L^{-1}$) of selected OCs for the snow season at all locations at which weekly snowfall measurements were made in the Canadian Arctic. (see Table 3.4 footnote for explanation of OC groups and Figure 3.11 for site locations).

Location/Winter	ΣHCH	ΣChlord	ΣDDT	HCB	ΣPCB
Alert 93/94	6.3	0.13	0.66	**1.4**	4.6
Eureka 91/92	**20.2**	0.16	0.34	0.14	2.6
Mould Bay 93/94	4.3	0.10	0.45	0.36	4.0
High Arctic Means	5.2	0.13	0.47	0.24	3.7
Standard Deviation	3.7	0.14	0.50	0.73	2.3
Cape Dorset 93/94	0.96	0.05	0.27	0.04	2.3
Snare Rapids 93/94	0.88	0.04	0.13	0.09	1.7
Dawson City 93/94	1.4	0.19	0.32	0.06	3.0
Whitehorse 93/94	0.73	0.10	0.14	0.03	1.2
Whitehorse 92/93	0.39	0.38	0.03	0.13	3.2
Tagish 93/94	1.1	0.17	0.25	0.07	2.5
Tagish 92/93	0.73	0.72	0.13	0.18	1.6
Fraser 93/94	1.3	0.14	0.17	0.02	1.4
Fraser 92/93	1.1	1.0	0.75	0.25	2.2
Other Arctic Means	0.95	0.31	0.24	0.10	2.1
Standard Deviation	0.30	0.32	0.20	0.07	0.66
Lake Superior Winter 1992	2.2	n/a	0.11	0.10	1.0

N.B.: bold values are outliers and are not used in the means or standard deviations

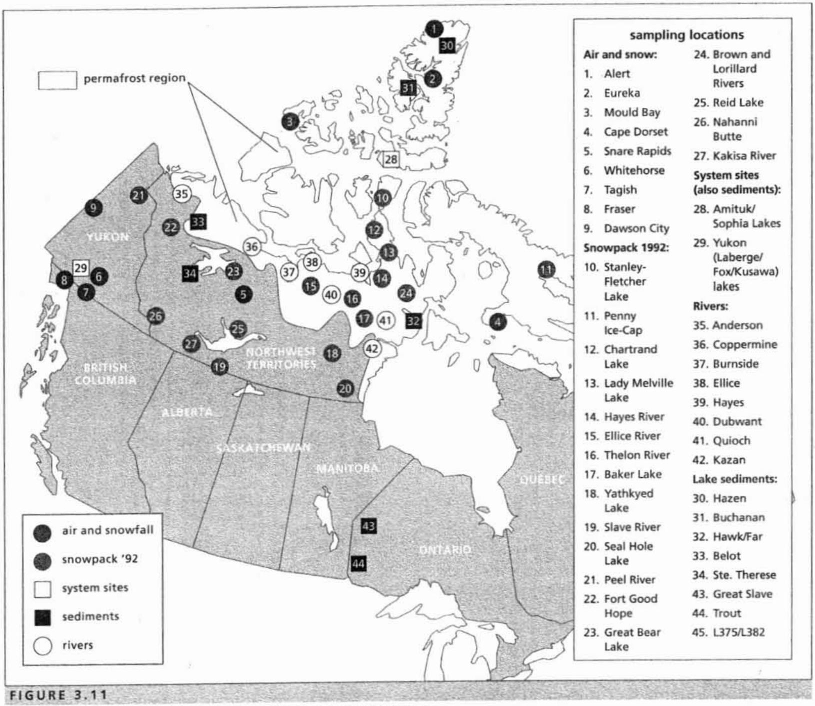


FIGURE 3.11

Map of Canada showing snowfall, snowpack and lake sediment sampling sites.

2.3.2.2 Snowfall

Snowpack measurements of OCs to estimate seasonal deposition rates have the following disadvantages: (i) there is post depositional loss of contaminants from the snowpack, and (ii) it is difficult to select a representative sampling site due to blowing snow and drifting. To better estimate the deposition of OCs by snow, a snowfall sampling program was undertaken. Large area samplers (4.5 m^2) were used to collect snow continuously for a period of 1 or 2 weeks. Immediately afterwards, samples were tightly enclosed in metal cases and kept frozen to prevent loss of contaminants. Site locations are shown in Figure 3.11.

A qualitative time-series comparison of observed OC concentrations in weekly snowfall (Figure 3.12) at Alert and Tagish, Yukon shows no consistent temporal deposition patterns. Observed variations in OC concentrations were not correlated with temperature or snowfall amount.

The snowfall-amount weighted-mean concentrations for the snow season at several sites are presented in Table 3.5. Concentrations of ΣHCH, HCB and ΣPCBs in snowfall are higher at high arctic locations (Alert, Eureka and Mould Bay) relative to mainland arctic locations. This may be due to enhanced scavenging of these compounds at the colder temperatures found in the high Arctic in spring or enhanced post depositional losses as one moves further south where temperatures increase earlier in spring. HCH, HCB and some of the lighter PCB congeners are relatively volatile.

Mean concentrations of OCs in snowfall at Lake Superior during the winter quarter of 1992 (December–February, inclusive) are also shown in Table 3.5 (Hoff *et al.* 1995). Mean PCB and DDT concentrations in snowfall are somewhat higher in the Arctic than in the Lake Superior region.

Precipitation-amount weighted mean concentrations of HCH, ΣChlor and ΣDDT were higher in

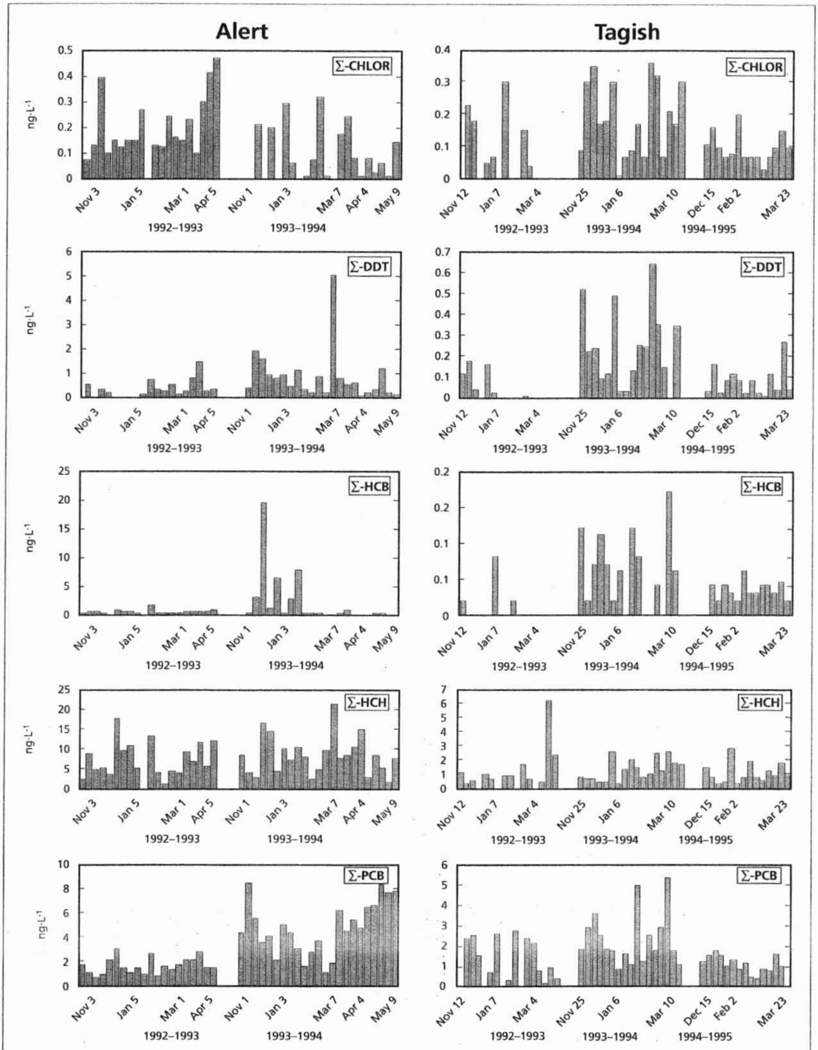


FIGURE 3.12

Time series of OC concentrations in weekly snowfall measurements at Alert and Tagish in the Canadian Arctic (see Figure 3.11).

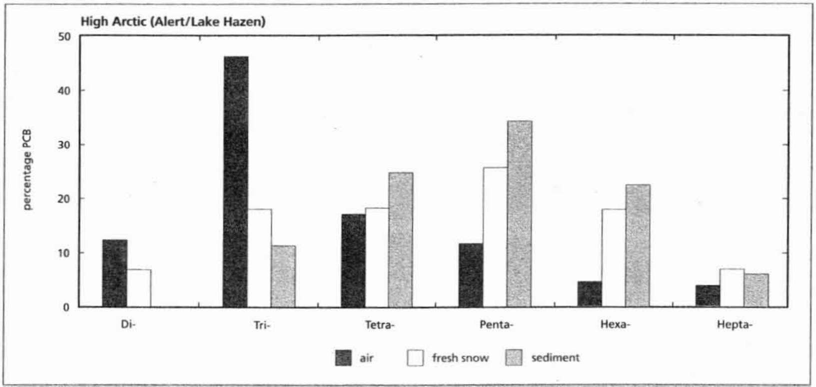


FIGURE 3.13A

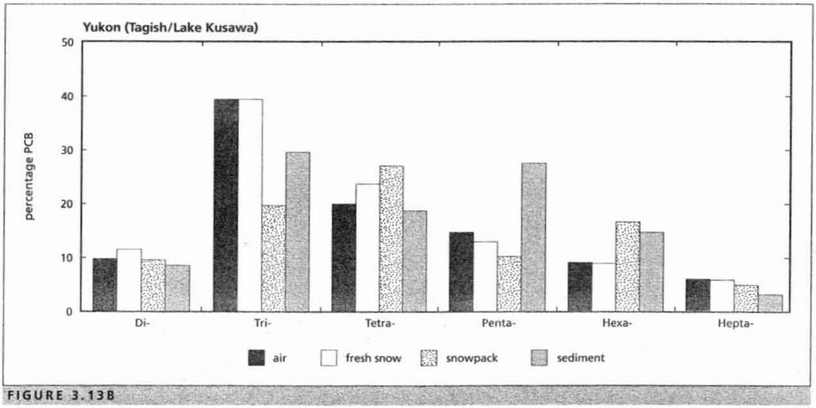


FIGURE 3.13B

A comparison of PCB homologue distributions in various environmental media in the high Arctic of Ellesmere Island and in the southern Yukon. Atmospheric data from Stern et al. *(1997). Winter of 1993/94.*

snowfall (Table 3.5) than in the snowpack (Table 3.4). Comparing the data for the Arctic mainland snowfall sampling stations with snowpack observations in Table 3.4, the following changes in contaminant concentration means in snow are observed over winter: ΣHCHs -20%, ΣChlor -87%, ΣDDT -63% and ΣPCBs +93%. However, due to high variance in each parameter and different sampling years, the differences between these multi-site data set means are not statistically significant (for a discussion of evidence of post-depositional losses of OCs from the snowpack and a site-by-site comparison of contaminant levels in cumulative snowfall and snowpack (see Section 2.4.1 and Figure 4.4). HCB was discounted in the above comparison since concentrations were generally low and approaching detection limits.

Mean PCB congener patterns in air, snowfall and lake sediments were compared for the region of northern Ellesmere Island (Alert/Lake Hazen) and the southern Yukon (Tagish/ Lake Kuzawa) (Figure 3.13). Scattered snowpack observations were also available for years other than 1991/92 in the southern Yukon. In the Yukon, congener distributions are very similar for atmosphere and fresh snowfall, peaking in the tri- and tetra-homologue groups. For snowpack and sediments, there is a slightly higher proportion of heavier congeners. In the high Arctic near Alert, congeners are relatively heavier in snowfall and sediment than in the atmosphere.

- *The most abundant OCs in arctic fresh snowfall at Alert NWT are:* $\Sigma\text{HCH} \approx \Sigma\text{PCB} > \Sigma\text{DDT} \approx \Sigma\text{HCB} \approx \Sigma\text{Chlordane}$*. Concentrations are generally much higher in the high Arctic than at the Tagish, Yukon site.*

- *A detailed snowpack chemistry survey in the Arctic in spring 1992 yielded the following ranking of OCs according to abundance:* $\Sigma\text{PCBs} > \Sigma\text{HCHs} > \Sigma\text{DDT}$ $\Sigma\text{HCB} \approx \Sigma\text{Chlordane}$*. Thus compared to snowfall, snowpack contains relatively less* ΣHCH *than* ΣPCB*. The former is more volatile than the latter.*

2.3.3 Marine/Delta

2.3.3.1 Processes that Control the Movement of Contaminants

Considerable progress has been made during the past decade in understanding the structure and circulation of the Arctic Ocean. Recently acquired insights into the functioning of the Arctic Ocean owe much to basin-scale geochemical transects carried out by surface ships since 1987. The most recent of these, the Arctic Ocean Section-94, had strong contaminant measurement support by the NCP. In addition to these icebreaker studies, large-scale synoptic surveys have been and continue to be carried out by submarines. It is therefore timely to update the previous review given in Barrie *et al.* (1992) wherein most of this recent work was not available. Canada's arctic and subarctic coastline stretches 166 000 kilometres, more than twice the length of Canada's Pacific and Atlantic coastlines combined (Figure 1.1). The oceanography of the marginal seas depends strongly on shelf area, water depth distribution (hypsometry), exchange with the interior ocean, seasonal ice cover, ice melt and freshwater runoff. These factors vary greatly from region to region.

In the west, Canada's largest arctic continental shelves, occupying about 100 000 km^2, are directly adjacent to the Arctic Ocean. The western shelves are seasonally ice-covered. Discharge from Canada's largest arctic river, the Mackenzie River, is a dominant influence on the regional oceanography, sedimentology and biogeochemistry. The Canadian Beaufort shelf has been the focus of many oceanographic and contaminant studies, due both to oil exploration in the 1970s and 1980s, and to ease of access by ships. (Macdonald and Thomas 1991, Thomas *et al.* 1990).

The "centrepiece" of Canada's Arctic is the Canadian Archipelago. The outer islands face directly onto the Arctic Ocean where there is only a relatively narrow continental shelf. There are no major rivers in this region and runoff enters seasonally from many small streams during the brief summer. This region contains some of the heaviest sea ice in the Arctic Ocean (Colony and Thorndike 1985). Because heavy ice has prohibited access by surface ships, the oceanography of this outer shelf is relatively poorly studied. Our knowledge of marine contaminants for this region depends almost entirely on data collected from an Ice Island in 1986–87 (Hargrave *et al.* 1988). The channels within the Archipelago provide an important conduit for water (top 200–250 m) flowing out of the Arctic Ocean and into Baffin Bay (Fissel *et al.* 1988). Although there is a better oceanographic data base within the Archipelago than for the outer shelf (Birch *et al.* 1987 1990, and Thomas *et al.* 1983a,b 1990), the channels of the Archipelago still cannot be considered well studied especially from the perspective of contaminants. Earlier studies carried out by mining interests focused on metals (Thomas *et al.* 1984). More recently under the NCP, OCs have been studied (Hargrave *et al.* 1997, Falconer *et al.* 1995a).

Hudson Bay, which is a broad, shallow, semi-enclosed sea, lies almost entirely south of the Arctic Circle. It does not interact directly with the Arctic Ocean but rather, it is externally forced by the Atlantic Ocean via Hudson Strait and by river runoff (Prinsenberg 1991).

Aside from water currents, three oceanographic factors control the transport of contaminants within the Arctic Ocean:

(i) Vertical stratification forms distinct layers that differ in origin and motion. The stratification inhibits mixing between the layers.
(ii) The seasonal cycle of ice melt/formation and runoff in the marginal seas affects water-mass formation and air-sea interactions, and
(iii) Phase partitioning (see Section 2.2.3) between water and inorganic or biogenic particles. The particle-associated contaminants tend to be sequestered into marine sediments and therefore do not travel as far with ocean currents as do the dissolved contaminants.

A simplified view of the vertical structure of the Arctic Ocean is shown in Figure 3.14. Detailed classification of water masses can be found in Carmack (1990), Anderson and Jones (1986) and Macdonald *et al.* (1989). The surface water has residence times on the shelves of from less than a year up to three years and, in the interior ocean, perhaps 10 years or more (Hanzlick and Aagaard 1980, Ostlund 1982, Macdonald *et al.* 1987, 1989, Anderson *et al.* 1989, Omstedt *et al.* 1994, Schlosser *et al.* 1994). Seasonal ice formation, ice melt and runoff affect density stratification of the water. In winter, a polar mixed-layer is produced by the freezing of ice and concomitant exclusion of brine (approximately the top 50 m). In summer a stratified surface layer is produced by the melting of sea ice and the addition of runoff at

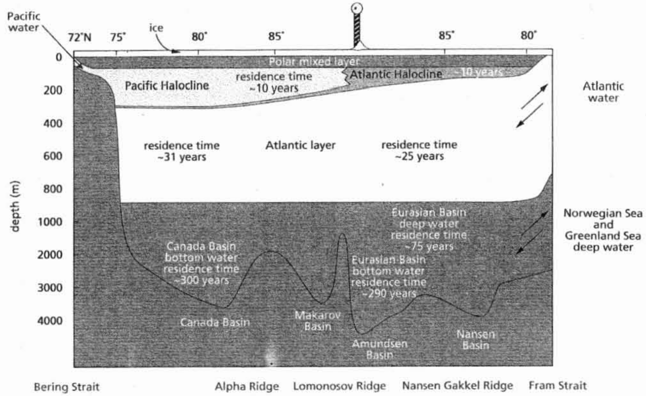


FIGURE 3.14

The vertical structure of the Arctic Ocean showing the polar mixed layer (approximately the top 50 m), the regions of density gradients between 50 m and 200 m (the haloclines and Pacific layer), the Atlantic Layer (approximately 200 m to 1000 m), and the basin waters (the figure has been modified from Chapter 3 of the Arctic Monitoring and Assessment Programme Report (AMAP 1997)). It should be noted that our understanding of the structure of the Arctic Ocean as shown in this figure is continuing to undergo significant revision following recent high-quality tracer transects of the Arctic Ocean.

the margins (5–10 m deep). The influence of direct exchange of dissolved gases with the atmosphere is therefore limited to these shallow ocean depths.

The near-surface water layer (30–50 m) is probably the most important layer in terms of the transport of contaminants and their entry into marine biota. This layer is coupled to the ice and moves approximately according to a large-scale characterized by the Beaufort Gyre and the Transpolar Drift currents (Figures 3.15 and 3.16). This latter feature is seen clearly in oceanographic sections of tracers of river water (e.g., tritium and $\delta^{18}O$ (Ostlund and Hut 1984, Bauch *et al.* 1995) and alkalinity and total carbonate (Anderson *et al.* 1989, 1994)). Much of the Russian river outflow appears to coalesce into the Transpolar Drift to be carried across the Eurasian Basin diluting only by about a factor of 10 by the time it exits through Fram Strait. Some of this water, together with associated contaminants, may enter the Beaufort Gyre, eventually exiting via the Canadian Archipelago. Currently, it is not known what fraction of the Russian river water enters into the Beaufort Gyre by this route. Despite the fact that the large Russian rivers discharge into surface waters of the Eurasian Basin, it is the Canadian Basin that contains the largest amounts of freshwater (Aagaard and Carmack 1989). This may be explained partly by the longer residence time of surface water in the Beaufort Gyre and partly by freshwater inputs coming directly from the Mackenzie River and indirectly from the Pacific inflow, which is relatively fresh and also carries the Yukon River's discharge.

Below the surface layer (Figure 3.14) is a structurally complex region of increasing density called the halocline (approximately 50 to 200 m depth). The halocline is crucial to the transport contaminants. On the one hand, it is maintained by lateral transport of water from the shelves (Aagaard *et al.* 1981, Melling and Lewis 1982, Moore and Smith 1986, Schlosser *et al.* 1994a) where it may acquire contaminants. On the other hand, it provides an effective barrier between the surface water and deeper water, thus blocking contaminant transport. Within the Canada Basin, the halocline is largely maintained by water that is imported from the Pacific Ocean and processed over the Chukchi Shelf (Figure 3.15). The motion of water in this layer (centered at about 200 m) is shown in Figure 3.16. The surface field of motion is dominated by the Beaufort Gyre which is clockwise, while at depth the direction of flow reverses moving water eastward along the basin margins (Aagaard 1984). Clearly, the path of any water property entering at the margin will depend on where it is inserted into the interior (i.e., in a buoyant surface plume or denser brine-enhanced water).

Within the Eurasian basin the halocline is maintained by Atlantic water processed on the large Barents, Kara and Laptev shelves (Rudels *et al.* 1996); the motion of this layer probably follows the Atlantic layer circulation which is shown in Figure 3.16. Water in the halocline has a residence time within the Arctic Ocean of the order of a decade or more (Wallace *et al.* 1987, Ostlund and Hut 1984).

Beneath the halocline is the warm Atlantic layer, which circulates throughout both arctic basins at a water depth of between approximately 200–800 m. Upon entering the Arctic Ocean through Fram Strait, the Atlantic Ocean water submerges below the surface layer and circulates on time scales of the order of 30 or more years (Rudels *et al.* 1994). Circulation is faster near the basin edges where currents are steered by topography while slower replacement occurs toward the centers of the basins. The deep basin waters (1500–4000 m) are oldest and have the longest residence times taking in the order of centuries to replace their water (Schlosser *et al.* 1990, Macdonald *et al.* 1993, Schlosser *et al.* 1994). The multi-compartmental model is a simplified yet useful description of a complex flow. The ocean surface layer compartment combines the surface and halocline layers discussed above (0 to 200 m depth) while the deep ocean compartment represents the Atlantic water layer (200 to 800 m).

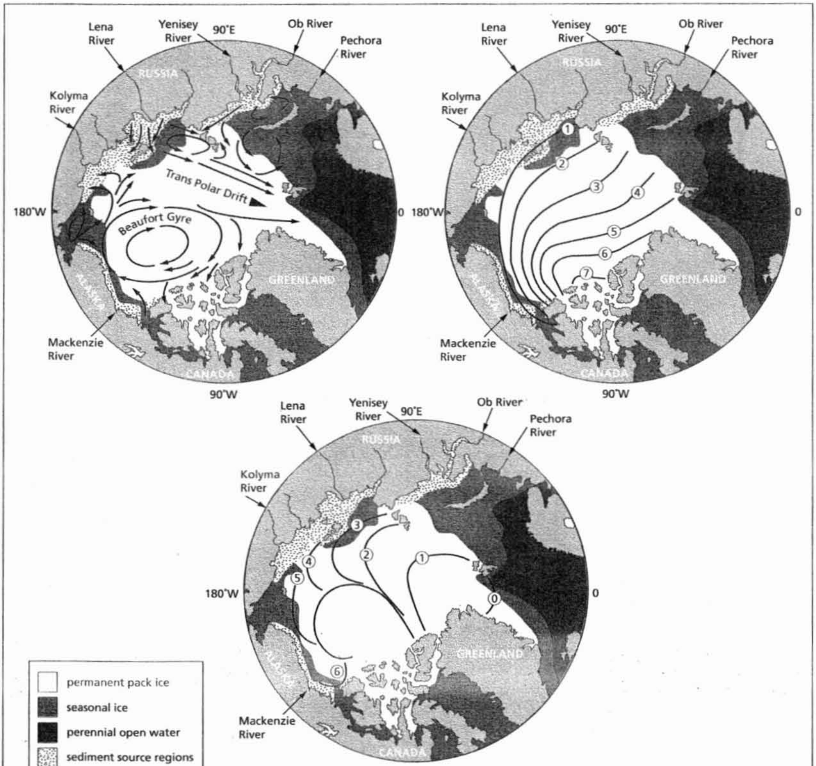


FIGURE 3.15

The circulation of ice and the surface layer (compiled from Gloerson et al. 1992, Colony and Thorndike 1985, Rigor 1992). The top left view shows the predominant motion of the ice; the top right view shows contours of mean present age of ice and the bottom view shows the approximate time (years) taken for ice at a given position to reach Fram Strait (the zero isopleth). These figures represent long-term averages and considerable variability can be expected in time and space.

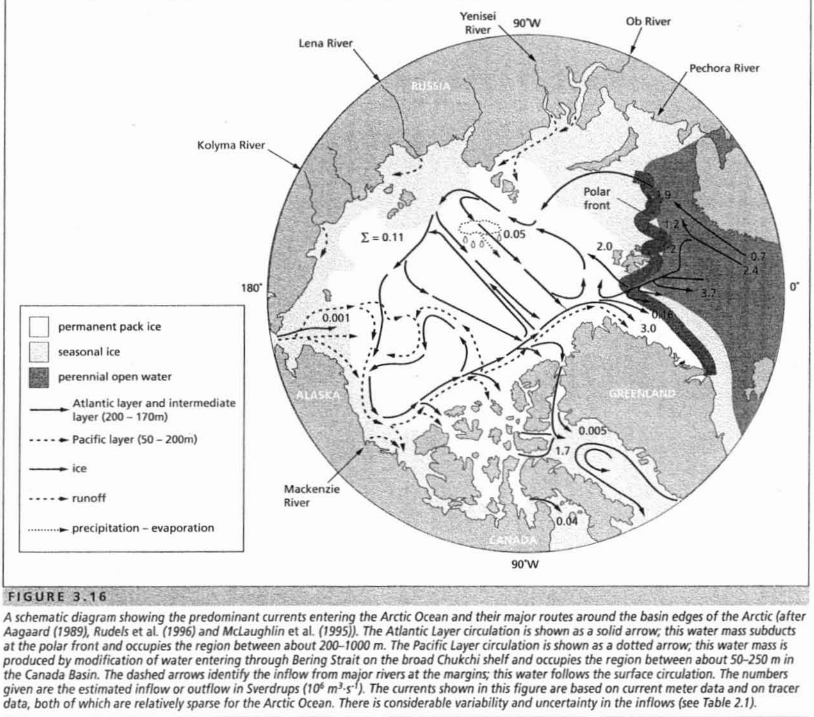


FIGURE 3.16

A schematic diagram showing the predominant currents entering the Arctic Ocean and their major routes around the basin edges of the Arctic (after Aagaard (1989), Rudels et al. *(1996) and McLaughlin* et al. *(1995)). The Atlantic Layer circulation is shown as a solid arrow; this water mass subducts at the polar front and occupies the region between about 200–1000 m. The Pacific Layer circulation is shown as a dotted arrow; this water mass is produced by modification of water entering through Bering Strait on the broad Chukchi shelf and occupies the region between about 50–250 m in the Canada Basin. The dashed arrows identify the inflow from major rivers at the margins; this water follows the surface circulation. The numbers given are the estimated inflow or outflow in Sverdrups (10^6 $m^3 \cdot s^{-1}$). The currents shown in this figure are based on current meter data and on tracer data, both of which are relatively sparse for the Arctic Ocean. There is considerable variability and uncertainty in the inflows (see Table 2.1).*

Ironically, much of what we know about transport rates within the Arctic Ocean is derived from recent measurements of a series of contaminant tracers. These tracers include, among others, carbon tetrachloride (dry cleaning fluid), freons (released to the atmosphere for the past 50 years), tritium (released by atmospheric weapons testing during the 1950s and early 1960s and entering the Arctic Ocean through runoff) and ^{137}Cs (released to the atmosphere by atmospheric weapons testing, and directly to the ocean by nuclear re-processing plants in Europe). These tracers currently exist in the upper Arctic Ocean, but have not penetrated the basin waters much. They have clearly demonstrated the importance of stratification and ocean currents in the movement of anthropogenic contaminants.

Figures 3.14 to 3.16 strongly imply that there are two major oceanographic domains within the Arctic Ocean. Within the Canada Basin, surface waters including the halocline are greatly influenced by water from the Pacific Ocean. Surface currents are organized in the clockwise drift of the Beaufort Gyre. In contrast, within the Eurasian Basin water from the Atlantic Ocean predominates and the dominant feature of the surface currents is the Transpolar Drift. A front, historically located approximately at the Lomonosov Ridge, provided the demarcation between the Atlantic and Pacific domains. However, recent basin transects from surface ships in 1993 and 1994 (supported in part by NCP) and submarines have changed our views on circulation within the Arctic Ocean (Macdonald 1996). Sections from these transects show that this front is not locked at the Lomonosov Ridge and that a large, rapid incursion of Atlantic water has recently displaced much of the Pacific layer up to the Mendeleev Ridge (Carmack *et al.* 1994, McLaughlin *et al.* 1996, Aagaard *et al.* 1996). This displacement of water along the basin margin has consequences both for climate and contaminant transport (Carmack *et al.* 1994).

Within the Arctic Ocean, contaminants may be transported in the water by ocean currents (Figure 3.16) or by ice (Figure 3.15) each of which has a different time scale and pattern of motion. The manner of contaminant entry (e.g., atmospheric particles, gas exchange, rivers, ocean currents), the contaminant's biogeochemical properties (e.g., phase association, volatility, solubility, bio-uptake) and the different transport modes interact to produce a bewildering number of potential pathways. Nevertheless, knowledge of the mode of entry and the biogeochemical properties of a contaminant allows prediction of behaviour. This is the basis for studying not only contaminant concentrations in the ecosystem, but also the physical and chemical properties and the modes of entry of contaminants into the Arctic.

Mesoscale eddies are potentially an important transport and redistribution mechanism for water properties in the Canadian Basin (D'Asaro 1988, Aagaard 1989). These features, which are characteristically 20 km in diameter and have maximum speeds of about 25 cm·s^{-1}, are long lived and may be one of the most important ways for water properties (including contaminants) at the basin boundaries to penetrate and mix into the interior ocean. The seasonal cycle of runoff and ice formation on the continental shelves is illustrated in Figure 3.17. In summer, the shelves tend to clear of ice which enhances atmospheric exchange through winds, waves and open water. However, the coastal ocean also stratifies, limiting the depth of exchange, and warms, enhancing outgassing. The summer freshet (Figure 2.6) brings with it a peak in river-borne particles and, possibly, contaminants. High Arctic lakes act as conduits of OCs (Section 2.3.4).

For the Mackenzie Shelf, the source of inorganic, terrestrial particles will be very important for the scavenging and transport of particle-associated contaminants (Macdonald and Thomas 1991). However for most other regions of the Canadian arctic shelves, the supply of inorganic particles will be much less, and biologically produced particles will be more important. Particle settling moves particle-associated contaminants vertically through the water column to the sediments. In turn shelf sediments can be resuspended and transported to the interior ocean, incorporated in ice and further transported, or taken up in bottom-dwelling organisms and thus re-enter the food chain.

During winter on the shelves (Figure 3.17), runoff is sharply reduced and sea ice forms (Macdonald *et al.* 1995). New ice covers the marginal seas and reduces, but does not eliminate, areas for gas exchange. Ice formation drives thermohaline circulation through the rejection of brine (Melling and Lewis 1982, Aagaard *et al.* 1985, Melling 1993), and therefore indirectly affects the movement of contaminants on the shelves. Dense water can travel along the shelf bottom to enter the Arctic Ocean interior at an appropriate density level, carrying with it both particulate and dissolved contaminants. Measurements of the contaminant loads have not been made and are presently speculative. Melling (1993) estimated the average contribution to halocline waters in the Arctic interior from the Mackenzie Shelf to be about 0.04 x 10^6 m^3·s^{-1} (1300 km^3·a^{-1}).

The Arctic Ocean's marginal seas undergo the greatest variations in fraction of open water (Figure 3.16, Gloersen *et al.* 1992). As noted above, inorganic particles enter the coastal seas from rivers, coastal erosion and resuspension of shelf sediments in shallow water. Perhaps 90% of these particles are trapped as sediment on the shelves and slopes (GESAMP 1993). The large shelves of the Arctic Ocean (about 30% of the total area) are, therefore, likely to be important sites for removing particle-associated contaminants like Pb, PAH, highly-chlorinated OCs and some radionuclides like the plutonium isotopes. In contrast to the shelves, biological productivity is lower under the permanent pack of the Arctic interior ocean (Subba Rao and Platt 1984), and there tends not to be as much inorganic or organic sedimenting material (Macdonald and Carmack 1991, Macdonald *et al.* 1993, Hargrave *et al.* 1994). Therefore the process of contaminant removal by particle scavenging and settling is expected to be more important around the margins of the Arctic Ocean than under the permanent pack.

The defining characteristic of the Arctic Ocean is sea ice. Ice thickness varies from first-year ice (2 m), to multi-year ice (3 m), to ice in pressure ridges (up to 20 m). The ice extent of the entire Polar Ocean (including seasonally ice covered regions in the Bering and Greenland Seas) varies from about 9.3 x 10^6 km^2 in early September, when the marginal seas are largely ice free, to 15.7 x 10^6 km^2 in late March (Figure 3.16). This variation modulates the exchange of both particulate and vapour phase atmospheric contaminants, whose behaviour depends critically on whether they interact with an ocean surface or an ice surface. Despite its small contribution to the total volume of water in the Arctic Ocean, sea ice has a potentially important role in the transport of contaminants and their entry into biota. First, sea ice provides a solid interface at the ocean's surface, and therefore, is an important habitat for a wide range of organisms from plankton to polar bears. Second, it collects atmospheric inputs throughout winter and spring to be released in summer into melt ponds and surface water at a time when biological productivity is at its highest. Third, much of the ice in the Arctic is produced in shallow marginal seas during winter and exported to the interior ocean. Ice forming on shelves can incorporate contaminated sediments during suspension freezing, frazil ice formation or from bottom-anchored ice (Reimnitz *et al.* 1990,

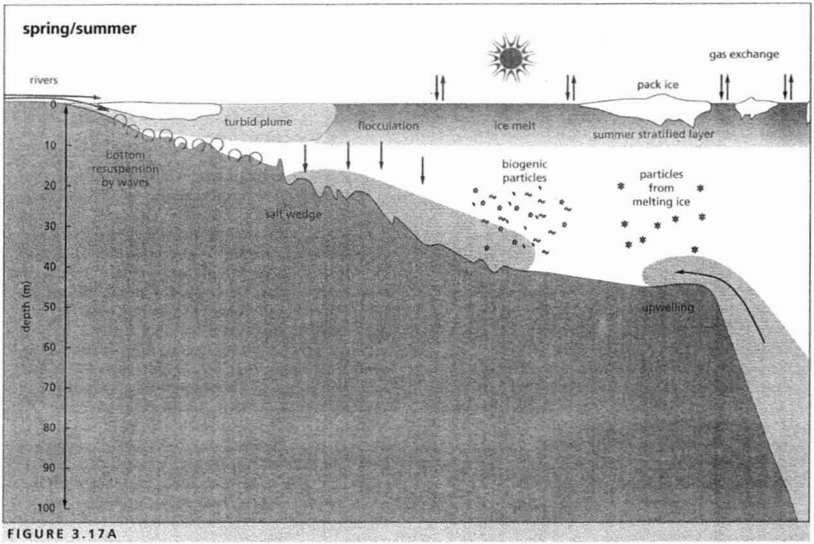


FIGURE 3.17A

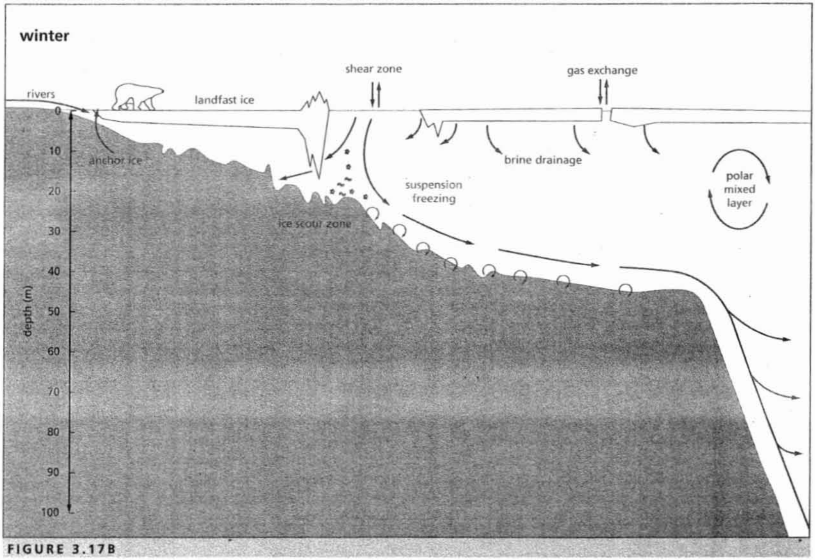


FIGURE 3.17B

Shelf processes affecting (A) the transport of dissolved and particulate matter in summer and (B) the transport of dissolved and particulate matter in winter.

1993a). This contaminated ice may then be transported thousands of kilometres undergoing little or no alteration (Pfirman *et al.* 1993).

The general motion of ice in the Arctic Ocean has long been known (Figure 3.15, Gordienko and Laktionov 1969). Recent measurements of the tracks of drift buoys (Rigor 1992) confirm the general picture, and from these data models have been constructed to predict the age and rate of movement of ice (e.g., Colony and Thorndike 1985). The Beaufort Gyre and the Transpolar Drift dominate the general motion of sea ice (Figure 3.15, top). The time taken for ice to transit the Arctic Ocean depends on where it is formed (Figure 3.15, bottom left). Ice produced in the Beaufort Sea takes six or more years before it exits the Arctic Ocean at Fram Strait while ice formed over the Russian shelves takes one to five years depending on location (Rigor 1992, Gloersen *et al.* 1992). The oldest multi-year pack ice in the Arctic tends to be found north of the Archipelago (Figure 3.15, bottom right) attesting to sluggish ice motion blocked by land (Gloersen *et al.* 1992). Most sea ice escapes the Arctic in the East Greenland Current through Fram Strait. However, a small amount also exits through the Archipelago (Table 2.1). Contaminants carried by the ice through Fram Strait will be released to the surface water in the Greenland Sea as the ice melts.

Of the major ice circulation features, the Beaufort Gyre clearly governs the motion of ice in the Canadian sector of the Arctic Ocean. In this gyre, ice circulates clockwise, potentially gathering atmospheric contaminants delivered during winter and spring. As can be seen from Figure 3.15, atmospheric contaminants deposited on the ice as far north as the pole may then be transported toward the Archipelago and the Canadian Beaufort Sea The multi-year pack ice in the center of the gyre will tend to grow toward an equilibrium thickness of 3 metres. It can travel around the gyre up to five times (e.g., the T3 ice island) before escaping at the northwest boundary and entering the transpolar drift (Thorndike 1986, Jeffries and Shaw 1993). Atmospheric contaminants, that are not readily lost in the summer melt, accumulate in this portion of the pack ice over the course of several seasons. New ice, produced in marginal seas during winter, is imported into the Beaufort Gyre primarily from the Beaufort Shelf and secondarily from the Chukchi and East Siberian shelves (Figure 3.15, Colony and Thorndike 1985). We have little information on the distribution and amounts of sediment carried by this ice, nor do we know whether such sediments may be contaminated. Reimnitz *et al.* (1993b) determined the particle loading for ice in the Beaufort Gyre to be about 40 $mg \cdot L^{-1}$ corresponding to 60 tonnes per square kilometre, which is about 50 times higher than in seawater. Nevertheless, sediments on the Beaufort shelf appear to have low contaminant burdens (Yunker *et al.* 1994, Macdonald, unpublished). The East Siberian Sea may have higher contaminant burdens originating from the Russian Rivers, but this region has not been the major direct recipient of contaminants, nor apparently is it a major exporter of ice to the central Arctic (Pavlov and Pfirman 1994). It is, therefore, unlikely that ice in the Beaufort Gyre delivers contaminated sediments from the Russian shelves to Canadian waters.

The Transpolar Drift, together with the Polar and Siberian branches (Figure 3.15), moves ice from source regions in the Laptev, Kara and Barents Seas across the ocean to exit principally by Fram Strait (Zacharov 1976, Pfirman *et al.* 1993, Pavlov and Pfirman 1994). Although the Laptev Sea is the major exporter of ice to the central Arctic Ocean (Reimnitz *et al.* 1994), it is primarily within the Kara Sea and secondarily within the Barents Sea that nuclear materials, which have caused such recent concern, have been dumped (Yablokov *et al.* 1993). In addition to radionuclides, the Kara Sea is most likely to receive contaminants from the extensive industrialized watersheds that border it, either directly from the atmosphere (e.g., see Melnikov 1991) or indirectly from the large rivers that discharge there, or from offshore oil exploration and production. Ice from the Kara and Laptev Seas takes about two to three years to reach Fram Strait (Figure 3.15, Rigor 1992) during which time modifications will occur. During summer, snow and ice melt off the surface and during winter growth, ice is added to the bottom. According to Zubov (1943), this process moves particles to the surface of the ice within two to three years. Thus, it may play a role in explaining the extraordinarily high OC concentrations observed in ice collected near Fram Strait (Gaul 1989). Upon exiting the Arctic this sea ice melts and releases particle-associated contaminants to the water in Fram Strait.

There is evidence that the Transpolar Drift can at times migrate toward the Canadian side of the Arctic Ocean (McLaren *et al.* 1987, Serreze *et al.* 1989) thereby delivering contaminated ice to the north of Greenland and into the Lincoln Sea. From there, the ice could travel south into Nares Strait, west and then south into the Archipelago or further west to join the Beaufort Gyre. Transit of ice through Nares Strait or the Archipelago would take a year or two (see Jeffries and Shaw 1993), during which the ice melts completely releasing its contaminant burden to the water. There are no data to support or refute such movements of contaminated ice, but this appears to be the most likely mechanism whereby contaminated sediments from the Kara Sea could eventually arrive in the Canadian Arctic.

2.3.3.2 Recent Findings Concerning Contaminants in Seawater, Sediments and Ice

Rather than adding to or repeating previous reviews summarizing contaminant burdens in oceanographic media (Muir *et al.* 1992, Lockhart *et al.* 1992, Barrie *et al.* 1992), we attempt to synthesize what has been learned recently about contaminants in marine abiotic media including sediment, sea water and ice. We emphasize work carried out under the NCP, but include other relevant studies. Knowledge of contaminant levels in sea ice is one of the weakest areas of Arctic contaminants research. Ice likely provides a critical path for contaminant transport over long distances within the Arctic Ocean (Pfirman *et al.* 1993, 1994) and therefore future measurements of the transport of ice and of contaminant burdens in ice within the Canadian Arctic are required.

2.3.3.3 Organochlorines

Measurements of OCs in the marine water column and ice up to 1991 have been reviewed by Muir *et al.* (1992). The most complete water-column data base is for α- and γ-HCH. Since 1985, vertical profiles have been measured for both α- and γ-HCH allowing us to assess the inventories of these compounds and their distributions in the ocean. Most of the data have been collected near the surface of the ocean with only relatively few points from water deeper than about 400 m. Concentrations range from "very low" in the deep ocean to as much as 7 $\mu g \cdot m^{-3}$ for α-HCH and 1.1 $\mu g \cdot m^{-3}$ γ-HCH at the surface. The concentrations of both forms of HCH are highest in the top 200 m of the water column. This is consistent with the choice of the ocean surface layer compartment of depth 200m (Figure 1.2). HCH is not uniformly distributed horizontally in surface waters; sampling stations in the southern part of the Canadian Basin contain relatively high concentrations — more than twice those observed in the Chukchi and East Siberian Seas and in the Greenland Sea. There is a large pool of surface water in the Canada Basin with a relatively long residence time. Due to permanent ice cover, it does not easily outgas. The surface concentrations in the Canada Basin may, therefore, reflect conditions of a decade or so ago when atmospheric HCH concentrations were much greater (Figure 5.1). The Canadian Archipelago draws its water from the surface of the Canada Basin. This explains the similarly high concentrations of HCH observed in Archipelago waters (Falconer *et al.* 1995a, Hargrave *et al.* 1997). Indeed, the greatest outflow of HCH from the Arctic Ocean is estimated presently to be through the Archipelago (Table 6.1).

TABLE 3.6

A summary of organochlorine concentrations ($ng \cdot m^{-3}$) in sea water (mean ± s.d.)

Location	Resolute Bay[1] n=8 (4 for DDTs) (1992)	Bering Sea[2] n=7 (1993)	Chukchi Sea[2] n=2 (1993)
CHBs	48 ± 8	22 ± 6	14 ± 0
trans-Chlordane	7.3 ± 4.2	1.2 ± 0.9	0.50 ± 0.08
cis-Chlordane	4.5 ± 2.5	0.90 ± 0.62	0.46 ± 0.05
trans-Nonachlor	1.5 ± 1.3	0.57 ± 0.35	0.26 ± 0.01
Heptachlor Epoxide		2.4 ± 0.5	2.8 ± 0.1
Dieldrin		3.7 ± 0.7	3.4 ± 0.2
Endosulphan I		2.1 ± 0.4	1.8 ± 0.1
p,p'-DDE	1.0 ± 0.3		
p,p'-DDT	< 4.0		

[1] *Bidleman* et al. *(1995b)*

[2] *Bidleman (1997)*

New observations confirm the relative abundance of OCs in arctic seawater previously described by Bidleman *et al.* (1990) (α-HCH > HCBs > γ-HCH ≈ CHBs (Toxaphene) > chlordanes ≈ PCBs > DDTs). A summary of recent surface ocean measurements for OCs is provided in Table 3.6. For OCs other than HCH, there are far too few data to assess the spatial or vertical distribution. At 200 m depth at the Ice Island (Hargrave *et al.* 1988a), HCB, chlordane and dieldrin concentrations were one-half or less than half of their surface concentrations.

Concentrations of OCs in snow melt water (see Tables 3.3 and 3.4) differ from those in Bering and Chukchi surface waters. ΣHCH in snow melt is a factor of approximately two lower than in surface sea water, while the PCBS and HCBs are a factor of two to five times higher. Other constituents (i.e., ΣDDT and ΣChlordane) are similar in both media. An explanation for these differences is not readily apparent, but could involve the kinetics of partitioning between the gas phase and either snow or water.

Canadian arctic marine sediments have generally not proven to be as fruitful a medium for OC measurements (Barrie *et al.* 1992) as they have in Canada's temperate coastal regions (Macdonald *et al.* 1992, Lebeuf *et al.* 1995) and elsewhere (Kjeller and Rappe 1995 and Evers *et al.* 1993). There are few marine sediment measurements for the Arctic. OC concentrations in sediment were often below detection limits. Surficial sediment samples collected from the Ice Island in 1986 and 1987 contained detectable quantities only for three types of compounds — PCBs, DDT, and *cis*-chlordane — but concentrations were less than 50 $pg \cdot g^{-1}$ (Hargrave *et al.* 1989a). In contrast, surficial sediments from a highly productive region in the Chukchi Sea have been found to contain more elevated concentrations (e.g., ΣHCH - 350 $pg \cdot g^{-1}$, Σ DDT - 200 $pg \cdot g^{-1}$, ΣPCB - 260 $pg \cdot g^{-1}$; cis-chlordane - 30 $pg \cdot g^{-1}$), possibly produced by rapid sedimentation of organic material (Macdonald *et al.* 1997). It seems likely that marine sediment concentrations of OCs will generally be very low except possibly at specific sites where accelerated fluxes could occur (e.g., at the ice margin where productivity and contaminant delivery to the water column may be enhanced).

2.3.3.4 PAHs

Historical concentration data for aromatic and aliphatic hydrocarbons in Arctic sediment and bulk (small volume) water samples have been comprehensively reviewed by Muir *et al.* (1992). Within the marine environment of the Canadian Arctic, the Mackenzie shelf of the Beaufort Sea provides the best opportunity for a thorough assessment of PAHs. A large, comprehensive hydrocarbon data set, including PAHs, was produced during the NOGAP program (Northern Oil and Gas Action Plan) and these data have since been thoroughly interpreted in a series of publications (Yunker *et al.* 1991, 1993, 1994, 1995, Yunker and Macdonald 1995).

Naturally occurring hydrocarbons, which predominate on the Beaufort Sea shelf, derive primarily from the Mackenzie River (Yunker *et al.* 1993). They include biogenic alkanes and triterpenes with a higher plant/peat origin, diagenetic PAHs from peat and plant detritus, petrogenic (i.e. geological source) alkanes, triterpenes and PAHs from oil seeps and/or bitumens and combustion PAHs that are likely relict in peat deposits. In the environment, parent PAHs (PAHs with unsubstituted rings) are usually accompanied by a series of alkyl-substituted homologues. While essentially the same pattern of alkanes, diagenetic hopanes and alkyl PAHs is observed in all river and most shelf sediment samples, alkane and triterpene concentration variations are strongly linked to the relative amount of higher plant/peat material. In the Beaufort Sea, parent PAH concentrations were found at relatively high values (in the range of 220 to 760 ng·g^{-1}) in sediments (Table 3.7). These concentrations exhibit little change between the Mackenzie River and the Beaufort shelf, decrease slightly at the shelf edge (approximately 200 m depth), and decrease further in the offshore (2700 m, Yunker *et al.* 1996, Table 3.7). The suspended particulate or dissolved PAH concentrations across the shelf were found to be very low (< 1 ng·L^{-1} to 6 ng·L^{-1}, Yunker *et al.* 1994).

In late winter, the total PAH concentration (that is, the sum of parent PAHs with molecular mass 178 to 278) on suspended solids was found to vary little across the shelf. However throughout spring and summer, progressively higher concentrations of suspended solids in the river led to a maximum in parent and alkyl PAH concentrations in the nearshore and a decrease with distance offshore. The alkyl PAH retene is representative of trends in PAHs from higher plant sources. The proportional contribution of retene at freshet to total PAH concentrations was the same in the river, in shelf sediments, and in most suspended particulate samples collected out to the mid-shelf (Yunker and Macdonald 1995). The proportion of retene decreased in suspended solids obtained further offshore and generally increased with depth; retene was not detected in most offshore spring samples.

In the water column of the shelf in winter and spring, dissolved alkanes and PAHs are an important component of the hydrocarbon budget. However, during the open water season, they are only a minor component of the hydrocarbon budget because concentrations of suspended solids are high. At this time, nearly all of the alkanes and parent PAHs are particle-associated. Finer particulate fractions are only a minor component of the PAHs in the nearshore, but, in the offshore marine regime, become a larger proportion of the PAH load (Whitehouse *et al.* 1989, Yunker *et al.* 1994).

A dissolved phase odd-even predominance (OEP) is found wherever significant higher plant alkanes are present (that is, there is a marked difference in concentration of alkanes with even and odd carbon numbers). The loss of this OEP, on going from the estuary toward the mid shelf, is most likely due to a reduction in the particle-associated supply of alkanes from a land-based source.

In the dissolved phase, the lower molecular mass (<178), more water-soluble PAHs predominate. This change in composition from the particulate phase is probably due to partitioning of these more water-soluble PAHs into the dissolved phase with coincident sedimentation of the less water-soluble, higher molecular weight PAHs (McVeety and Hites 1988). Total dissolved PAH concentrations are often lower in the nearshore than in the offshore. This is consistent with results of other studies that have reported both greater scavenging of PAHs by particles in areas with higher suspended sediment loads and stronger interaction of PAHs with terrestrial organic matter (Whitehouse *et al.* 1989, Bouloubassi and Saliot 1993).

The more water-soluble parent PAHs and the alkyl PAHs cadalene, pimanthrene, simonellite and retene are present in the same proportions in the dissolved and particulate phases (Yunker *et al.* 1994). These

TABLE 3.7

PAH concentrations in sediments (ng·g^{-1})[a] by sampling location for the Mackenzie River and Beaufort Sea of the Canada basin.

Location	n	PAH Total[b]		C0–C4 Alkyl Naphthalene[c]			C0–C4 Alkyl Phenanthrene[c]		
		Mean ± SD	Range	Mean ± SD	C_{max}	Range	Mean ± SD	C_{max}	Range
Mackenzie River delta	10	980 ± 120	820–1100	1500 ± 360	3	1000–2000	2100 ± 250	1	1800–2500
Mackenzie shelf	5	860 ± 230	590–1200	1400 ± 500	3	800–2000	1700 ± 680	1	650–2500
Mackenzie shelf edge	5	500 ± 240	290–890	660 ± 240	3	420–980	720 ± 570	1	260–1500
Beaufort Sea, 2700 m	8	400 ± 200	220–760	680 ± 350	2	360–1300	650 ± 440	2	300–1600

[a] *Mean ± one standard deviation; n is the total of the number of sample replicates. See Yunker* et al. *(1996) for station locations.*

[b] *Parent PAHs of molecular-mass 178 to 278, perylene excluded.*

[c] *Concentration sum for substituted naphthalenes and phenanthrene/anthracenes with 0 to 4 alkyl carbons. C_{max} is the series maximum.*

natural two- and three-ring PAH compounds originate in plant sesqui- and diterpenoid essential oils and apparently have solubilities, particle associations and phase distributions similar to the two to four ring parent PAH. Hence, solubility is a factor controlling dissolved-phase PAH composition.

PAH ratios indicate that anthropogenic, atmospheric PAHs are minor contributors to particle-associated PAHs in the Mackenzie River estuary but relatively more important for the surface and offshore waters of the shelf. Because higher PAHs are only occasionally detected in the dissolved phase, the relative abundance of fluoranthene and pyrene becomes the most reliable indicator of atmospheric PAHs. The fluoranthene-to-fluoranthene plus pyrene ratio is frequently larger in the dissolved phase than in the particulate phase, and larger in fine particulate than in coarse particulate, indicating greater combustion input to these components. The predominance of naphthalene and anthracene in the dissolved and colloidal phases suggests a petrogenic origin; this is consistent with the predominance of petrogenic aliphatic hydrocarbons in the dissolved phase (e.g., Ehrhardt and Petrick 1993).

2.3.3.5 Metals

Metal concentrations in sea water from the Arctic Ocean were reviewed by Muir *et al.* (1992). Few new data have been reported since that time. As noted in the earlier review, concentrations are low and great care is required to avoid sample contamination during processing. For the Arctic Ocean, there is only a relatively small number of trace metal profiles in sea water: Moore 1981 (Zn, Cd, Cu) and 1983 (Cd), Campbell and Yeats 1982 (Ni, Cu, Cd), Danielsson and Westerlund 1983 (Cd, Cu, Zn), Mart *et al.* 1983 (Cd, Pb, Cu, Ni), Mart *et al.* 1984, Schmidt and Freimann 1984 (Hg), Yeats 1988 (Ni, Zn, Cd),Yeats and Westerlund 1991 (Cd, Cu, Ni, Zn, Co), Seakem 1991 (Zn, Pb, Cd, Cu) and recently, work in the Lena and Yenesey estuaries by Martin *et al.* 1993 (Cd, Cu, Ni, Pb, Zn); Dai and Martin 1995 (Cd, Cu, Ni, Pb) and Coquery *et al.* (1995).

Because the oceans have been contaminated globally by industrial Pb, there now exists no baseline against which to assess Pb. Schaule and Patterson (1981) estimated that pre-historic oceans had Pb concentrations of about 0.6 $ng{\cdot}L^{-1}$. Measurements by Mart *et al.* (1983, 1984) for the eastern Arctic Ocean give surface water Pb concentrations of about 15 $ng{\cdot}L^{-1}$ decreasing to about 3–4 $ng{\cdot}L^{-1}$ at 3000 m. These authors concluded that the values were typical of "clean ocean areas." Nevertheless, considering the estimate for pre-industrial ocean Pb concentrations, and the fact that contaminant Pb is known to have been delivered in significant quantities to the Arctic (e.g., Rahn 1981, Sturges *et al.* 1993, Akeredolu *et al.* 1994, Pacyna 1995) it seems probable that much of the Pb in the Arctic Ocean derives from industrial rather than natural sources. Recent data from the Lena River estuary (Martin *et al.* 1993) and from Garrow Bay in the Archipelago (Seakem 1991) suggest that concentrations of Pb in the nearshore are similar to the Arctic offshore (i.e., in the low background range).

Numerous vertical profiles of cadmium concentrations in Arctic Ocean sea water have been measured. The most recent of these, by Yeats and Westerlund (1991), confirms earlier profiles finding a range of 30 to 70 $ng{\cdot}L^{-1}$. The most distinct feature of these Cd profiles is a pronounced maximum coincident with the nutrient maximum observed at about 100 metres in the Canadian Basin. The well-known correlation of Cd with phosphate (Bruland 1980, de Baar *et al.* 1994) is due to the bio-activity of Cd; it is incorporated in plankton and remineralizes along with the soft body parts in sinking particles. Yeats and Westerlund (1991) determined the cadmium-phosphate relationship for the Arctic Ocean to be: $Cd\ (nM) = 0.40\ P\ (\mu M) - 0.12$. The biological and geochemical activity of Cd generally makes oceanographic profiles unreliable for the assessment of contaminant inputs. Cd profiles for the Arctic Ocean appear entirely consistent with natural oceanic processes, and the concentrations fall into ranges observed in other oceans. Yeats and Westerlund (1991) concluded that most of the Cd associated with the nutrient maximum in the Arctic Ocean was actually imported from the Pacific in association with the nutrients.

There appear to be no recent Hg data for the Arctic Ocean with which to assess its natural or anthropogenic geochemistry or its trends. In earlier work, Schmidt and Freimann (1984) reported an average of 3.7 $ng{\cdot}L^{-1}$ for Arctic Ocean water, and Thomas (1983a,b) reported 1–15 $ng{\cdot}kg^{-1}$ in the Beaufort Sea. Total dissolved Hg concentration in unpolluted ocean water is currently thought to be about 0.2–1 $ng{\cdot}kg^{-1}$ (NATO-ARW 1996). In reviewing the available information on global Hg cycling, Mason *et al.* (1994) concluded that the atmosphere and surface ocean were in rapid equilibration. Because Hg fluxes to ocean sediments are small, the dominant sink for contaminant Hg is continental. Nevertheless, these authors concluded that anthropogenic emissions over the past century have probably resulted in a three-fold increase in global atmosphere and surface ocean Hg concentrations and, if all anthropogenic emissions were removed, elimination of this anthropogenic load from the ocean would take 15 to 20 years. It is probable that the rate of decrease of Hg concentrations in ocean sediments could be slower because the Arctic Ocean is both ice-covered and has low particle fluxes. In addition, several properties of mercury in the environment (i.e. water-soluble, atmospherically exchanged, weakly particle-associated) are similar to those of HCH summarized in

Section 2.6.1. Thus, Hg may be similar to HCH in its fate and distribution in the Canadian Arctic. Pacyna and Keeler (1995) estimate that large emissions of Hg occur from both Eurasia and North America and that presently about 60–80 tonnes of anthropogenically mobilized Hg is deposited in the Arctic annually. Since Hg is volatile, toxic and biomagnifies, it has the same characteristics that cause such concern for many of the OCs. Determining the distribution of Hg in the Arctic Ocean remains a priority for future research.

Data for metals in arctic marine sediments have been reviewed by Muir *et al.* (1992). The salient points made by these authors were that the observed concentrations of metals in sediments depends to a large degree on regional and local geology, particle size, organic content, depth in the sediment, and proximity to a contaminating source. Biomixing of surface sediments and geochemical processes that cause metals to move within the sediment can also play crucial roles. For these reasons, regional maps of metal concentrations in surficial sediment generally provide little information regarding contamination and no information on flux. The only clear case of contamination identified by Muir *et al.* (1992) was in Strathcona Sound where metals in sediments were clearly augmented by nearby mining activities (Nanisivik). In contrast, samples collected in Garrow Bay near the Polaris Mine operation show no such contaminant metal enrichment (Seakem 1991). The apparent absence of metal contamination in the marine environment near Polaris may be due to efforts to contain the tailings and metals by discharging them into Garrow Lake.

Cadmium profiles have been measured in sediment cores collected from the Mackenzie River and Banks Island shelves in 1990 (Gobeil *et al.* 1991) and in cores collected in 1993 during a cruise to the Chukchi and East Siberian seas (Gobeil, unpublished data). These core profiles, an example of which is shown in Figure 3.18, often exhibit Cd concentrations exceeding the Canadian marine sediment quality guideline of 0.7 $\mu g \cdot g^{-1}$. In some cores, maximum Cd concentrations have been found to attain 3 $\mu g \cdot g^{-1}$. The elevated concentrations are always found below the manganese-enriched surface layer, whose thickness ranges from 1 to 25 cm. Manganese, because it forms insoluble oxides or hydroxides in the presence of oxygen but is remobilized under reducing conditions by forming Mn^{2+}, is a good indicator of the redox zones in a sediment. Typically, low concentrations of Mn are found at depth in the sediments under reducing conditions whereas, near the sediment surface, solid-phase Mn is enriched by diffusion from below, followed by precipitation once the dissolved Mn comes into contact with oxygen. The consistent appearance of Cd below the Mn enriched layer indicates that the source of Cd enrichment is not anthropogenic but, rather, linked to the sediment redox conditions. The Cd distributions, therefore, are more likely to be a consequence of diffusion, which for Cd occurs under oxic conditions, downward into the sediment to precipitate in sulphide phases in the reducing zone where sulphide first begins to form. The formation of an authigenic Cd phase in suboxic sediment, which has also been observed in other coastal areas (Gobeil *et al.* 1987, Pedersen *et al.* 1989, Rosenthal *et al.* 1995, van Geen *et al.* 1995), implies that Cd profiles in sediments cannot be used in any straightforward fashion to calculate anthropogenic Cd fluxes or trends in the Arctic Ocean.

Contrary to the case for Cd, lead is a particle-associated element and its sedimentary record has often been used to document temporal trends of lead contamination in the aquatic environment. Sediment cores collected on the Beaufort Shelf (Gobeil *et al.* 1991) generally show low sediment Pb concentrations (80–150 $nmol \cdot g^{-1}$ or 16–31 $\mu g \cdot g^{-1}$) and no conclusive evidence of significant contaminant Pb input. In contrast, sediment cores collected from the basin margin in the Canadian Basin show such inputs (Gartside *et al.* 1995, Figure 3.19). The contaminant Pb in basin cores is evident in Pb enrichments near the surface (25 $\mu g \cdot g^{-1}$ compared with 20 $\mu g \cdot g^{-1}$ at depth) and also in a dramatic shift in the stable isotope composition (right panel in Figure 3.19). Work is continuing on these sediment cores to determine the source of the Pb (e.g., see Sturges *et al.* 1993 for

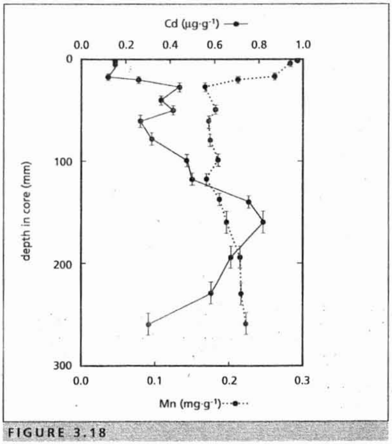


FIGURE 3.18

Cadmium (Cd) (solid line) and manganese (Mn) (dotted line) profiles (solid phase concentrations) for a core collected on the Banks Island shelf (73° 11.6' N 126° 28.9' W). High Cd concentrations at about 15 cm depth in the core are due to natural redox processes that cause Cd to diffuse to depth and become incorporated in sulphides (data are from Gobeil et al. *(1991).)*

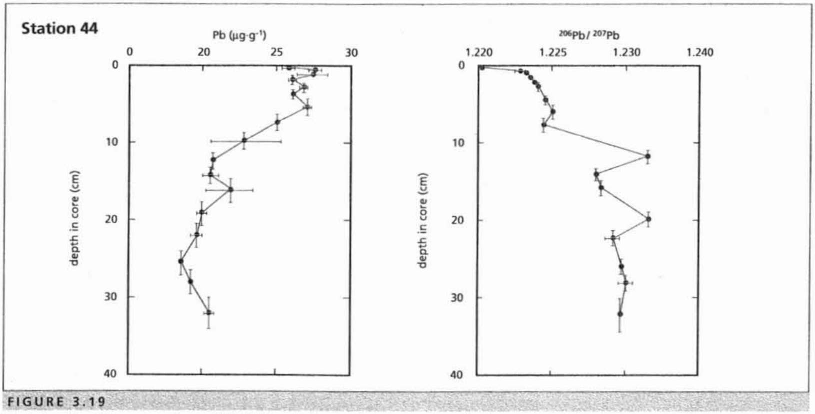


FIGURE 3.19

Vertical profile of Pb concentration and ^{206}Pb/^{207}Pb ratio for a sediment core collected in the Canada Basin (71° 19.0'N 141° 27.1'W, 2650m). Data are from Gartside et al. *1995.*

a discussion of Pb isotopes and sources within the Arctic Ocean).

There are no new Hg data for marine sediments from the Canadian Arctic Ocean. Two marine sediment cores collected in Hudson Bay (Lockhart *et al.* 1995) have sediment Hg concentration ranges of 16–34 µg·g^{-1}. Both cores have Hg enrichments (i.e., the ratio of post-industrial to pre-industrial deposition) near the surface of the sediments of 1.4 to 1.5 (see Table 5.2). However, Lockhart *et al.* (1995) conclude that such small enrichment factors at the sediment surface are not necessarily due to addition of anthropogenically mobilized Hg, but may derive from other factors such as diagenesis. Concentrations in Russian arctic rivers and estuaries (Lena, Ob, Yenesey) are lower than those observed in non-arctic rivers affected by human inputs (Coquery *et al.* 1995).

There are few measurements of metals in sea ice. The limited data suggest that concentrations are higher in ice than in surface sea water possibly due to deposition from the atmosphere. For example, Melnikov (1991) found that ice on the Russian shelves often contained high concentrations of contaminant metals, and that these metals may be released to the surface water when ice melts. Campbell and Yeats (1982) reported concentrations for Cd of 0.3 µg·L^{-1} for ice collected in Baffin Bay. Pfirman *et al.* (1995a) draw attention to the role of ice as a potentially effective means of transporting atmospheric and sediment-bound contaminants, and also for providing a critical pathway into biota. More work is needed to establish contaminant concentrations in ice from the Canadian Arctic Ocean.

2.3.3.6 Radionuclides

The principal inputs of anthropogenic radionuclides to the Canadian Arctic have been fallout from atmospheric nuclear weapons testing and from the nuclear power plant accident at Chernobyl in 1986. The Chernobyl input was distinguished by the presence of the short-lived radioisotope, ^{131}I (half-life of 8 days), which was detected on air filter samples collected within a week of the accident at stations (including those at Resolute and Alert) in the Canadian Arctic (Barrie *et al.* 1992) and samples collected one month after the accident at the Canadian Ice Island (Smith and Ellis 1995). Elevated ^{134}Cs/^{137}Cs ratios were measured in snow, precipitation, and lichen samples collected in the Canadian Arctic following the Chernobyl accident, which indicates that the quantity of Chernobyl-derived, radioactive Cs deposited between 60°N and 70°N in northern Canada was approximately 10% to 30% of the total fallout inventory. It is difficult to determine the actual inventory of Chernobyl-derived Cs deposited in the Arctic Ocean because there were very few atmospheric monitoring stations operating in the Arctic in 1986.

In addition to the direct atmospheric inputs of Chernobyl radioactivity to the Arctic Ocean, there will be contributions due to initial deposition in the North and Baltic Seas and subsequent advective transport into the Arctic Ocean with Atlantic water. The total Chernobyl contribution of ^{137}Cs to the Arctic Ocean is of the order of 5 x 10 ^{15}Bq (Aarkrog 1993). Most will eventually circulate through the Arctic Ocean with the water in the Atlantic layer (Figure 3.16). Finally, the one source of radioactivity unique to Canada is associated with the re-entry of the Soviet nuclear powered satellite COSMOS 954, which gave rise to elevated concentrations of short-lived fission products in the Great Slave Lake area of the NWT in early 1978 (Tracey *et al.* 1984). Little of the 2 x 10^{12} Bq each of ^{137}Cs and ^{90}Sr contained in the satellite was actually deposited on the Earth's surface.

Table 3.8 provides a summary of radionuclide concentration ranges for marine media measured in the Beaufort Sea in 1994. Surface seawater concentrations of ^{137}Cs are much lower in the Canada Basin (2 to 6 Bq·m^{-3}) than those presently found in the Makarov and Eurasian Basins (about 15 Bq·m^{-3}). This is because labelled water from the Sellafield nuclear fuel re-processing plant in England (see Section 2.2.1), which is making a significant contribution to surface waters in the Eurasian Basin, has not yet penetrated into the Canada Basin. Surface water input in the Canada Basin is derived mostly from global fallout. In view of the recent incursion of Atlantic water masses across the Lomonosov Ridge (see Section 2.6.5 and Figure 6.10) it is probable that ^{137}Cs concentrations will be observed to increase in Canada Basin surface water during the next decade as the Sellafield signal propagates around the basin. Ultimately, the Sellafield signal will be seen in the Canadian Archipelago water, which draws its water predominantly from the top 100–150 m of the Canada Basin.

Levels of radioactivity in nearshore marine sediments in the Canadian Arctic are typical of those measured at similar latitudes in other regions of the Arctic, with ^{137}Cs and ^{239,240}Pu measured in surface sediments at activities of the order of 1–10 and 0.1–1 Bq·kg^{-1}, respectively and derived almost exclusively from fallout (see Section 2.4.4. and Figure 4.9). The concentration of these radionuclides in the sediments is partly influenced by factors such as particle size and organic matter content. Biological activity often affects the distribution of radionuclides within the sediments by mixing surface sediments, to a greater or lesser degree, to depth within the sediments. There is evidence for enhanced inventories of radioactive contaminants measured in fine-grained material overlying the shelf/slope regions compared with the deep basins of the Arctic Ocean, probably owing to the greater productivity and water column particle supply in the marginal sea regions.

Radionuclide profiles in core SS-4 (Figure 4.10), collected from a water depth of 268 m on the shelf of the Beaufort Sea, are typical of those measured in Canadian Arctic marine sediments. ^{210}Pb is a naturally occurring radionuclide that is produced *in-situ* in the water column as part of the ^{226}Ra radioactive decay series. Similar concentrations have been reported for both ^{210}Pb and the human-made radionuclide ^{137}Cs, in sediments from the shelves of the Eastern Chukchi Sea (Baskaran and Naidu 1995). These authors concluded that the inventories of natural ^{210}Pb are low relative to those found on subarctic shelves. Further they suggested that the ^{137}Cs levels can be explained entirely by fallout — that is, there is not detectable excess ^{137}Cs from the former Soviet Union ocean dumping activities.

^{210}Pb is produced naturally at a relatively constant rate and is readily adsorbed onto particulates and deposited in sediments. In contrast, the vertical distribution of the human-made radionuclides, ^{137}Cs and ^{239,240}Pu, is governed by the historical record of atmospheric fallout (Figure 4.9).

^{137}Cs concentrations in sediment-laden ice in the Beaufort Sea exhibit a wide range, including some unusually high activities (>70 Bq·kg^{-1}) for which there is no clear explanation. The source of this ice appears to be the Canadian Archipelago yet, to date the only surface sediments identified having such elevated activities of ^{137}Cs are found in the Kara Sea. ^{137}Cs-laden ice has an interesting potential as a tracer of ice transport, but it does not appear that the measured inventories of ^{137}Cs represent a significant radiological risk.

- *In the west, Canada's largest arctic continental shelves, occupying about 100,000 km², are directly adjacent to the Arctic Ocean. The western shelves are seasonally ice-covered. Discharge from Canada's largest arctic river, the Mackenzie River, is a dominant influence on the regional oceanography, sedimentology and biogeochemistry.*
- *The centrepiece of Canada's Arctic is the Canadian Archipelago. The outer islands face directly onto the Arctic Ocean where there is only a relatively narrow continental shelf. There are no major rivers in this region and runoff enters seasonally from many small streams during the brief summer. Our knowledge of marine contaminants for this region depends almost entirely on data collected from an Ice Island in 1986–87.*
- *Aside from water currents, three oceanographic factors control the transport of contaminants within the Arctic*

TABLE 3.8

Radionuclide concentration ranges for marine phases in the Beaufort Sea in 1994.

Medium	Concentration ^{210}Pb	^{137}Cs	^{239,240}Pu	Reference
Surface Seawater (Bq·m^{-3})	0.5–1.5	2–6	0.005–0.015	Smith and Ellis (1995)
Surface Sediment (Bq·kg^{-1})	20–250	2–15	0.1–1	Gobeil *et al.* (1991), Baskaran and Naidu (1995), J.N. Smith (pers. comm.)
Ice (Bq·m^{-3})	—	< 1	—	Meese *et al.* (1995)
Ice-Sediment (Bq·kg^{-1})	—	5–80	—	Meese *et al.* (1995)

Ocean: (i) vertical stratification (ii) the seasonal cycle of ice melt/formation and runoff in the marginal seas and (iii) phase partitioning. In winter, a polar mixed-layer is produced by the freezing of ice and concomitant exclusion of brine (approximately the top 50 m). In summer a stratified surface layer is produced by the melting of sea ice and the addition of runoff at the margins (5-10 m deep). The influence of direct exchange of dissolved gases with the atmosphere is therefore limited to these shallow ocean depths. The near-surface water layer (30-50m) is probably the most important layer in terms of the transport of contaminants and their entry into marine biota.

- *Beneath the surface Arctic Ocean layer (0-200km) is the warm Atlantic layer which circulates throughout both arctic basins between about 200 - 800 m water depth. Upon entering the Arctic Ocean through Fram Strait, the Atlantic Ocean water submerges below the surface layer and circulates on time scales of the order of 30 or more years.*
- *Circulation is faster near the basin edges where currents are steered by topography while slower replacement occurs toward the centers of the basins. The deep basin waters (1500 - 4000 m) are oldest and have the longest residence times taking in the order of centuries to replace their water.*
- *The defining characteristic of the Arctic Ocean is sea ice. Ice thickness varies from first-year ice (2 m) to multi-year ice (3 m) to ice in pressure ridges (up to 20 m). Despite its small contribution to the total volume of water in the Arctic Ocean, sea ice has a potentially important role in the transport of contaminants and their entry into biota. First, sea ice provides a solid interface at the ocean's surface, and therefore, is an important habitat for a wide range of organisms from plankton to bears. Second, it collects atmospheric inputs throughout winter and spring to be released in summer into melt ponds and surface water at a time when biological productivity is at its highest. Third, much of the ice in the Arctic is produced in shallow marginal seas during winter and exported to the interior ocean. Ice forming on shelves can incorporate contaminated sediments during suspension freezing, frazil ice formation or from bottom-anchored ice. This contaminated ice may then be transported thousands of kilometres undergoing little or no alteration.*
- *New observations confirm that the relative abundance of OCs in arctic seawater is α-HCH > HCBs > γ-HCH≈CHBs (Toxaphene) > Σchlordane≈PCBs >DDTs and are consistent with earlier ranking of relative abundance.*
- *ΣPAH composition indicates that anthropogenic, atmospheric PAHs are minor contributors to particle-associated PAHs in the Mackenzie River estuary but relatively more important for the surface and offshore waters of the shelf.*
- *Because the oceans have been contaminated globally by industrial Pb, there now exists no baseline in the surface oceans against which to assess contaminant inputs. However, contaminant Pb is known to have been delivered in significant quantities to the Arctic. In surface marine sediment cores of the Canada basin, it is present at higher concentrations near the surface than in deeper, pre-industrial period sediments. Also, this was associated with a dramatic shift in the stable isotope composition from natural ratios at depth to anthropogenic ratios at the surface.*
- *There is no recent data for Hg concentrations in Arctic Ocean waters with which to assess its natural or contaminant geochemistry or its trends. Hg may be similar to HCH in its fate and distribution in the Canadian Arctic. Determining the distribution of Hg in the Arctic Ocean remains a priority for future research.*
- *The principal inputs of anthropogenic radionuclides to the Canadian Arctic have been fallout from atmospheric nuclear weapons testing and from the nuclear power plant accident at Chernobyl in 1986.*
- *Levels of radioactivity (^{137}Cs and ^{239,240}Pu) in nearshore marine sediments in the Canadian Arctic are typical of those measured at similar latitudes in other regions of the Arctic. ^{137}Cs inventories can be explained entirely by fallout — that is, there is no detectable excess ^{137}Cs from the former Soviet Union dumping activities. ^{137}Cs-laden ice has an interesting potential as a tracer of ice transport, but it does not appear to be a significant radiological risk.*

2.3.4 Terrestrial/Freshwater

2.3.4.1 Rivers

Organochlorines

Information on contaminant loadings to the Arctic Ocean from northward flowing rivers is limited. Prior to the NCP, most North American research focused on the Mackenzie River and its tributaries since it is the largest riverine system in the north. Contaminant observations were limited to PAHs (Yunker and Macdonald 1995, Yunker *et al.* 1991, Thomas *et al.* 1986). It was concluded that the suspended sediment load of the Mackenzie River is the major source of PAHs to the Beaufort basin of the Arctic Ocean. Although the major transport of these compounds was via suspended solids, an increasing proportion was carried in the dissolved phase during low flow periods. The PAHs were primarily natural in origin with only a small fraction from anthropogenic sources including long-range transport.

As part of NCP, Jeffries *et al.* (1996) analysed Canadian Arctic rivers for OCs as well as PAHs during the period 1992 to 1994. Samples were separated into dissolved and suspended solid fractions. Average concentrations of selected OCs measured in the Mackenzie River and 11 other north-flowing rivers of the Canadian Arctic in 1993 are presented in Table 3.9. The contribution of the suspended solid fraction to the total OC concentration was less than 10%. Except for the Mackenzie River, the concentration of suspended particulate matter (SPM) was generally low, ranging from 1 to 10 mg·L^{-1}. In the Mackenzie River, SPM was much higher (50 to 200 mg·L^{-1}). Many of the analytes were at or below their detection limits and hence the variability was high whenever replicates were done. This has obvious implications for any comparisons.

Volume-weighted-mean concentrations were most accurate for ΣHCHs (sum α- and γ- isomers). Values for the Mackenzie River (0.32 ng·L^{-1} at the Arctic Red River) were lower than the means of those in other northern rivers flowing to either the Arctic Ocean (1.2 ng·L^{-1}) or Hudson Bay (1.1 ng·L^{-1}).

The much longer residence time of water in the Mackenzie River system allows greater time for volatilization or degradation to take place than in smaller rivers. To a lesser extent, and with less certainty, lower PCB concentrations were also observed in the Mackenzie River (0.64 ng·L^{-1} at Arctic Red compared to 0.75 ng·L^{-1} and 0.81 ng·L^{-1} for rivers flowing to the Arctic Ocean and Hudson Bay, respectively).

On the other hand, ΣDDT concentrations were approximately the same in the Mackenzie River (0.10 ng·L^{-1} at the Arctic Red) as in the other rivers flowing to the Arctic Ocean (0.09 ng·L^{-1}). Rivers draining to the Arctic Ocean had higher ΣDDT concentrations than those draining to Hudson Bay (0.03 ng·L^{-1}). ΣChlor concentration, like that of ΣHCH, is less in the Mackenzie River (0.01 ng·L^{-1}) than in the other arctic rivers (0.10 ng·L^{-1}) and Hudson Bay rivers (0.07 ng·L^{-1}). HCB concentrations were too close to detection limit to be useful.

No assessment of contaminants in the Arctic would be complete without considering inputs from the Russian sub-continent whose rivers contribute more than 68% of total annual riverine input of water to the Arctic Ocean (Table 2.2). However, data on the concentrations of OCs in Russian rivers are sparse. Analyses of a limited number of river waters sampled from 1992 to 1994 have been reported in an internal Russian government publication (Roshydromet 1995). Samples were collected two to six times during each year, depending on the geochemistry of the basin and the likely use of the chemicals. Most of the data pertain to the HCHs. The reported ΣHCH concentrations (range 3 to 17 ng·L^{-1} in 1992 and 1993 for all Russian rivers except the Ob River) are higher than those in Canadian rivers (0.3 to 1.7 ng·L^{-1}); the Ob River results were exceptionally high (43 to 59 ng·L^{-1}). However, a confident comparison with North American data awaits an intercomparison of sample collection, handling and analytical methodologies.

PAHs

For PAHs, several noteworthy conclusions can be drawn from the study of Jeffries *et al.* (1996). PAH concentrations in replicate samples had a relative standard deviation of 50% to 100%. The order of prominence of unsubstituted PAHs in dissolved and suspended fractions of the Mackenzie River and delta (all season) was: perylene > phenanthrene > naphthalene > pyrene, chrysene, benzo[g,h,i]perylene. Others were present in lesser concentrations. Methyl naphthalenes dominated the methyl-substituted PAHs in both dissolved and suspended fractions. For unsubstituted naphthalene and the mono-methyl naphthalenes, dissolved concentrations exceeded those in suspended solids. This was reversed for the other PAHs.

In 11 other rivers from the region (6 flowing directly to the Arctic Ocean), concentrations of unsubstituted and substituted PAHs in suspended solids were more than 10 times lower than in the Mackenzie River and, in most cases, were below detection limits. Concentrations of dissolved low molecular weight PAHs in the smaller rivers were similar to those in the Mackenzie River, while dissolved higher molecular weight PAHs were 10 times lower. Dissolved concentrations of PAHs greatly exceeded those in the suspended solids. It is not clear at this time whether this is due to differences in the carbon content of the suspended solids or in the dissolved carbon content of these waters.

The Slave River is part of the Mackenzie River system. It is the major Canadian river entering the Arctic System (Figure 1.1), draining an area of approximately 600 000 km^2. In the period between

TABLE 3.9

1993 mean concentrations (sum of dissolved and suspended solid fractions) (ng·L^{-1}) of selected contaminants in Canadian rivers flowing to the Arctic and to Hudson Bay (Jeffries *et al.* 1996). See Figure 3.11 for river locations.

	ΣHCH	ΣDDT	ΣPCB	ΣChlord
Mackenzie at Arctic Red	0.32	0.10	0.64	0.01
Mackenzie East	0.45	0.25	3.04	0.05
Mackenzie West	0.35	0.00	4.83	0.07
Andrews	0.28	0.00	0.91	0.00
Coppermine above Copper Creek	0.80	0.11	1.02	0.00
Burnside at mouth	1.17	0.06	0.71	0.08
Ellice at mouth	1.70	0.04	0.79	0.06
Back above Hermann River	1.54	0.12	0.62	0.16
Hayes above Chantrey Inlet	1.66	0.11	0.54	0.20
Volume weighted mean in non-Mackenzie rivers flowing to the Arctic Ocean	**1.2**	**0.09**	**0.75**	**0.10**
HUDSON BAY RIVERS				
Thelon below Shultz Lake	0.90	0.04	0.71	0.08
Dubawnt at Marjorie Lake Outlet	1.14	0.03	1.20	0.07
Kazan above Kazan Falls	1.41	0.02	0.73	0.04
Quoich above St.Clair Falls	0.78	0.03	0.77	0.08
Lorillard (flow estimated)	1.23	0.04	0.82	0.07
Volume weighted mean in Hudson Bay Rivers	**1.1**	**0.03**	**0.81**	**0.07**

1980 and 1995, the Slave River Environmental Quality Monitoring Programme undertook to describe baseline water quality conditions at Fort Smith, NWT. In part, the impetus for the studies came from a need to determine possible contamination from the Alberta tar sands, and hence PAHs were a significant component of the studies. Sixteen priority PAHs were investigated — from two-ringed naphthalene to six-ringed structures such as benzo*[g,h,i]*perylene — during each of 1992, 1993, and 1994. Dissolved concentrations were generally less than detection limits. Suspended sediments sometimes exceeded the Canadian sediment quality guidelines (Environment Canada 1996) but their significance with regard to transport or effects of contaminants within the Slave system is unknown. The Slave River has near-median PAH concentrations in the suspended solids compared to other arctic rivers (Jeffries *et al.* 1994). The impact of seasonal differences with regard to the PAH concentrations in the suspended solids in the river water is unknown.

Metals

Metal concentrations observed in filtered water and suspended sediments from the Slave River are presented in Table 3.10 (McCarthy *et al.* 1995). While mercury and cadmium were infrequently found in water, lead concentrations in water often exceeded national water quality guidelines (CCREM 1987). Only arsenic did not exceed any water guidelines at any time during the sampling program. Also shown in Table 3.10 are concentrations of the metals in the suspended sediments from the Slave River. Some seasonality was observed, with elevated concentrations occurring during times of higher levels of clay particles. Concentrations of arsenic and copper found in suspended sediment were above the bed sediment guidelines in 70% and 50% of the samples, respectively. Zinc exceeded guideline levels only during October of 1990. There are no known local anthropogenic sources of these metals in the immediate region of Fort Smith.

TABLE 3.10

Trace metal concentrations ($\mu g \cdot L^{-1}$) in the Slave River (Fort Smith). (from McCarthy *et al.* 1995).

	Concentration			
	Median	Range	Guidelines	# of Samples
Dissolved				
Arsenic	2	0.6–9.	50[1]	15
Copper	6	1.0–23.	2[1]	15
Lead	4	1.0–10.	1 to 7[1]	15
Susp.Sediment				
Arsenic	7	4.2–17	5.9[2]	17
Copper	17.5	6.3–95	36[2]	17
Lead	8.2	3.6–21	35[2]	17
Cadmium	0.6	0.2–1	0.60[2]	17
Mercury	0.044	0.017–0.15	0.17[2]	17

[1] *Canadian Water Quality Guidelines (CCREM 1987)*

[2] *Canadian Bed Sediment Guidelines (Environment Canada 1996)*

No comprehensive measurements of metals in the Mackenzie River have been made that are suitable for pathways studies.

2.3.4.2 Lakes and Aquatic Systems

Lakes are a dominant feature of the arctic terrestrial landscape. They are of particular interest as hosts of aquatic food sources, for historical records of contaminant deposition recorded in their sediments, and as potential sinks of contaminants. A schematic of the major physical pathways of contaminants for a lake basin is shown in Figure 3.20. Prior to the NCP, contaminant dynamics in northern lakes had not been examined. The status of contaminants in lakes was addressed by three NCP research initiatives including: (i) detailed studies of a high Arctic lake and catchment area, (Amituk Lake), (ii) studies of a sub-arctic lake system (southern Yukon River basin), and (iii) measurement of the geographical distribution of contaminants in lake sediments. The majority of these initiatives focused on measuring OCs. Few measurements of the levels of heavy metals in northern lake surface waters were taken as part of the NCP. Trends in concentrations of heavy metals in lake sediments are discussed in Section 2.6.4. Measurements of radionuclides in northern lake surface waters are discussed toward the end of this section. Radionuclides in the Arctic are discussed in more detail in Section 2.6.5.

Arctic and subarctic lakes and their catchment areas are snow or ice covered from 32 to 52 weeks of the year. The lakes are characterized by their extremely low productivity and extended ice cover. Contaminant loadings to the lakes are controlled by atmospheric scavenging processes and by the behaviour of contaminants in the snowpack during winter, and as the snowpack warms in spring, by interactions between meltwater and catchment substrates, and finally, by the extent of mixing that occurs between meltwater and the lake itself. Contaminant concentrations in the lake and hence exposure of aquatic biota, are also controlled by limnological factors such as thermal stratification, basin morphology, and sediment load.

Amituk Lake Study

The Amituk Lake project investigated the behaviour of inorganic and organic contaminants, as well as major ions, in the spring snowpack and the ice-free lake, and included a detailed analysis of lake hydrology. Field studies were conducted during the summers of 1992 to 1994. Chemical analysis, data interpretation and modelling efforts are ongoing. Amituk Lake is a landlocked freshwater lake located on the eastern coast of Cornwallis Island at latitude 75° 02′ 57″ and longitude 93° 45′ 51″. The catchment area contains six small watersheds of combined area 26.5 km^2. The 0.38 km^2 lake has a single outlet that ultimately flows

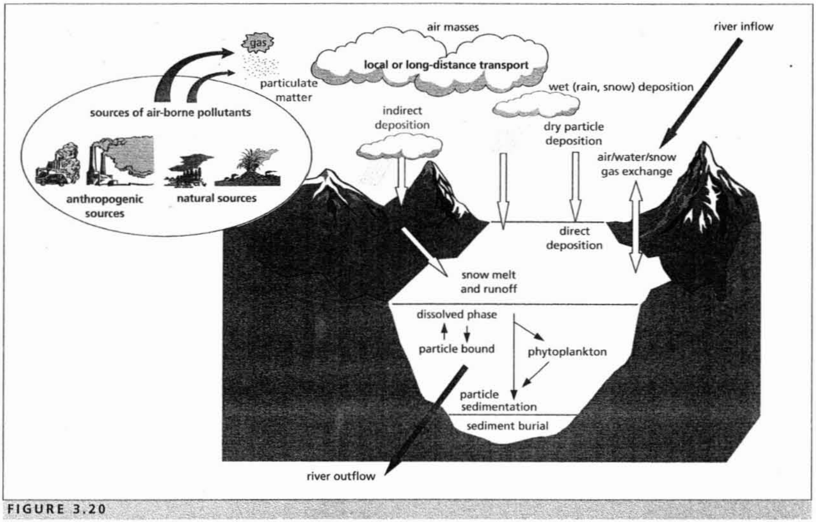


FIGURE 3.20

Schematic of the major pathways of contaminants in freshwater lake systems.

into Read Bay and the Arctic Ocean. The lake is bow shaped, with mean and maximum depths of 19.4 and 43 metres, respectively. It is monomictic, turning over once yearly during summer. The climate is typical of High Arctic regions with an annual mean temperature of -16.6°C. As a desert, precipitation rates are extremely low, with a climatological average of 131 mm (reported for nearby Resolute Bay, Environment Canada 1990). Annual precipitation during the study period was somewhat higher than this mean — 150 to 292 mm. About half the precipitation falls as snow in two peak periods, late spring and early fall. High winds during winter cause the snow to be unevenly distributed throughout the catchment (Woo 1976). These features have implications for contaminant storage in the snowpack and delivery to the lake upon melting.

Based on the characteristics of the Amituk snowpack, the snow cover has been differentiated into two snow types. In 1994, the shallow snowpack that collects on the upland plateaux and lake ice had an average depth, specific gravity and snow water equivalence (SWE) of 0.47 m, 0.433 g·cm^{-3} and 204 mm, respectively. Whereas the deeper snow that accumulates in stream valleys had values of 2.76 m, 0.642 g·cm^{-3} and 1803 mm, respectively. SWE is essentially the depth of liquid water formed if a snowpack were melted. A unit of 1 mm is equivalent to 1 L of liquid $H_2O \cdot m^{-2}$. Differences between the two snow types extends to the chemistry of the snowpack.

The HCHs and ΣPCB are the most abundant OC compounds reported for the Amituk Lake snowpack (Table 3.11). Due to changes in analytical methodology in 1994, the number of PCB congeners detected increased from 69 to 116 with an obvious increase in ΣPCB from ~700 to 1200 pg·L^{-1}. OC snowpack concentrations were consistently higher in the deeper valleys than in shallow windswept snowpacks. To explain this observation, it is hypothesized that the shallow snow is older, accumulating during early fall and then subject to wind scouring throughout the winter. In contrast, it is suggested that the valley snow is fresher having fallen during late spring, which is consistent with its greater specific gravity and SWE relative to the shallow snow. Thus, concentration differences can be explained by volatilization of OCs from the aging shallow snowpack during winter, as Gregor (1991) has hypothesized (see Section 2.4.1). Alternatively, differential scavenging and atmospheric conditions prevalent during the two peak snow falls may account for the concentration difference.

The results in Table 3.12 show a decrease in HCHs in deep valley snow from 1992 to 1994. The decline is in accordance with reports of decreasing concentrations of HCH in arctic air (Bidleman *et al.* 1995a, Jantunen and Bidleman 1995, Falconer *et al.* 1995a). Shallow snow concentrations do not reflect this change, possibly due to the hypothesized post-depositional loss processes during winter.

The extreme heterogeneity in snowpack water and chemical content precluded an exact measurement of the snowpack OC burden prior to the spring melt. However, using both field surveys and photographic imagery, a basin-wide SWE was estimated using the grid method of Woo and Rowsell (1993). Using estimated SWE and mean shallow/valley OC concentrations, the snowpack OC burden was calculated for 1992–1994 (Table 3.12). For comparison, the results of an earlier snowpack survey in the Arctic completed by Gregor (1989) are also presented (Table 3.12).

At Amituk Lake, the shallow snowpack was sampled on May 14, 1993 and again on June 13, 1993. As seen in Figure 3.21a, air temperatures between these two sampling dates changed from sub-zero conditions to about 5°C . Concentrations of most OCs were lower in the June than May samples (Figure 3.21 b–e). Three possibilities can account for this difference. First, analytical uncertainty may be responsible since this pattern was evident for the least abundant OCs, but not HCHs that occur at the highest concentrations (for further discussion see Section 2.4.1). Second, the June snowpack may have been depleted of OCs due to volatilization from the aging snowpack. Third, OCs could be conveyed downwards in the snowpack with freshly melted snow as temperatures rise above 0°C in early June.

TABLE 3.11

OC concentration ranges (pg·L^{-1}) in Amituk Lake snowpack at Shallow Valley. For each range, the first and second numbers refer to shallow and deep snowpack, respectively.

	YEAR		
	1992	**1993**	**1994**
Dieldrin	106–181	47–111	31–59
Endosulfan	135–466	95–734	217–605
ΣChlord	114–309	75–290	105–309
ΣHCH	525–6392	250–4106	651–1943
ΣDDT	71–113	29–56	174–347
ΣPCB (69)	1112–763	785–1615	705–706
ΣPCB (116)	—	—	1188–1317

ΣChlord = heptachlor + heptachlor epoxide + *cis*-chlordane + *trans*-chlordane+ *trans*-nonachlor
ΣHCH = α-HCH + β-HCH + γ-HCH
ΣDDT = DDD + DDE + DDT

TABLE 3.12

A comparison of basin average snowpack burden (ng·m^{-2}) of OCs at Amituk lake in June (1992, 1993, and 1994) with those measured throughout the Arctic in April/May by Gregor (1989).

	Amituk Lake Snowpack			Arctic Snow
	1992	1993	1994	1986–1987
Heptachlor Epoxide	11.1	9.2	13.6	10–16
Trans-Nonachlor (T-N)	2.9	2.8	1.6	
cis-Chlordane	5.2	2.9	3.5	1–20
cis + *trans*-Chlord+T-N	10.1	5.8	7.9	
ΣChlord	21.2	18.3	24.5	
Dieldrin	15.1	8.3	5.5	30–40
Endosulfan I	29.5	39.6	48.9	30
α-HCH	203.9	141.3	95.2	100–300
β-HCH	13.2	8.1	8.1	
γ-HCH	95.4	52.2	50.1	70–100
ΣHCH	312.4	201.5	153.4	
ΣDDT	9.8	4.6	32.2	2–10
ΣPCB (69)	100.9	128.6	94.4	70–100
ΣPCB-(116)			165.5	

The processes of chemical fractionation and preferential elution of chemicals excluded from ice crystals in the initial snowmelt have been well documented for ions in temperate regions (Semkin and Jeffries 1988, and Trantor *et al.* 1986). In Figure 3.22, stream flow and the concentrations of OCs and PCBs in Gorge Creek (the major tributary of Amituk Lake) are shown for the study period. Chemical fractionation of the more water-soluble OCs (HCH, endosulfan) is readily apparent in the initial stream flow. This is in contrast to less fractionation observed for the less water soluble PCBs and DDTs. While this process occurs once melt commences, the June snow samples were taken prior to melt suggesting that this explanation is unlikely. The high HCH concentrations in June snow are inconsistent with the latter two explanations and remain puzzling.

About 95% of the snowpack in Arctic catchments melts within three weeks during spring, resulting in a more intense runoff period than in temperate systems (Woo 1980, Semkin 1996). Initial meltwaters can flow over the ice cover of the lake. As a moat is formed at the lake margins, the influx of cold (0°C) meltwater into the lake with water at 4°C, results in the meltwater flowing through in a stable layer under the ice and above the water column (Adams and Allan 1987). Whole-lake mixing occurs when the lake is free of ice and the water column overturns, which is after most of the meltwater (with its chemical burden) has been transported through the lake (Figure 3.23). Consequently, little of the early meltwater, with its enriched concentrations of the more soluble ions and OCs, enters the lake. As stream flow and lake ice recedes, an increasing fraction of the later, more dilute meltwater mixes with the lake (Bergmann and Welch 1985, Semkin 1996). Finally, up to 5% of the total water input to the lake enters as shallow groundwater flow (Kinney, pers. comm.). This water is able to interact with catchment soils and dissolved major ions and, possibly, accumulated contaminants.

The fraction of meltwater that mixes with the lake is lake-specific and dependent on yearly climatic conditions. An average value can be obtained by using input/output data collected during the study (Table 3.13). The fraction, as well as information on within lake chemical dynamics, can be obtained from a mechanistic mass balance model based on the Amituk Lake data (Freitas *et al.* 1997, Diamond 1993, 1994). A schematic of observed and model-predicted mass balances of α-HCH and ΣPCBs (69 congeners) is given in Figure 3.24.

The mechanistic mass balance model, based on the QWASI fugacity/aquivalence model of Mackay (1991) and Mackay and Diamond (1989), was developed for high Arctic lakes and applied to Amituk Lake and nearby Char Lake. Model details are provided by

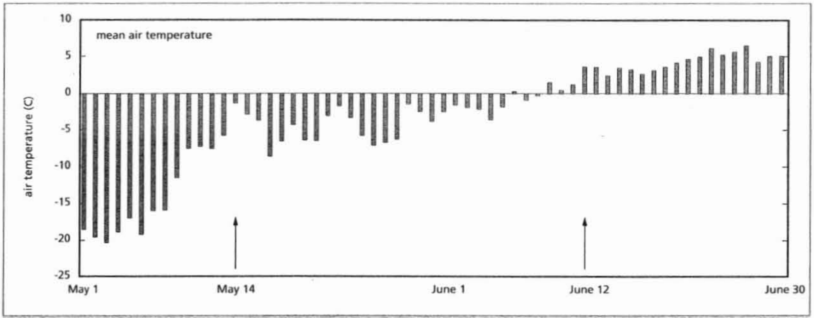


FIGURE 3.21A

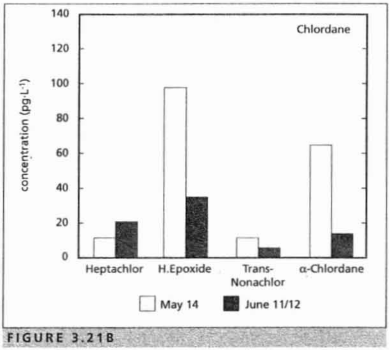


FIGURE 3.21B

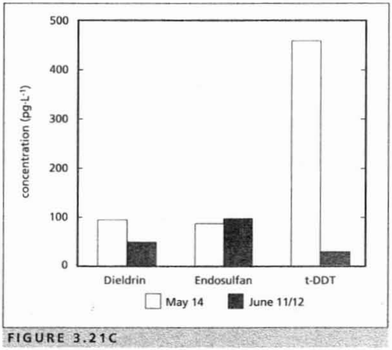


FIGURE 3.21C

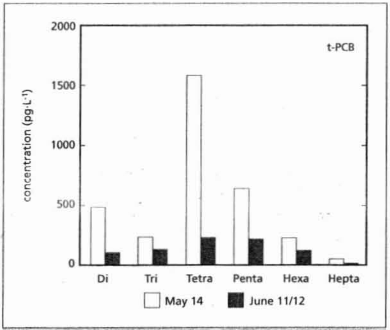


FIGURE 3.21D

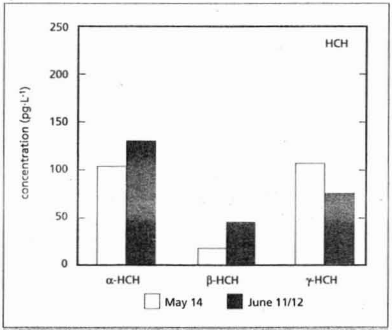


FIGURE 3.21E

Air temperature (A) and changes in OC burden of the snowpack (B to E) during spring warming at Amituk Lake in 1993.

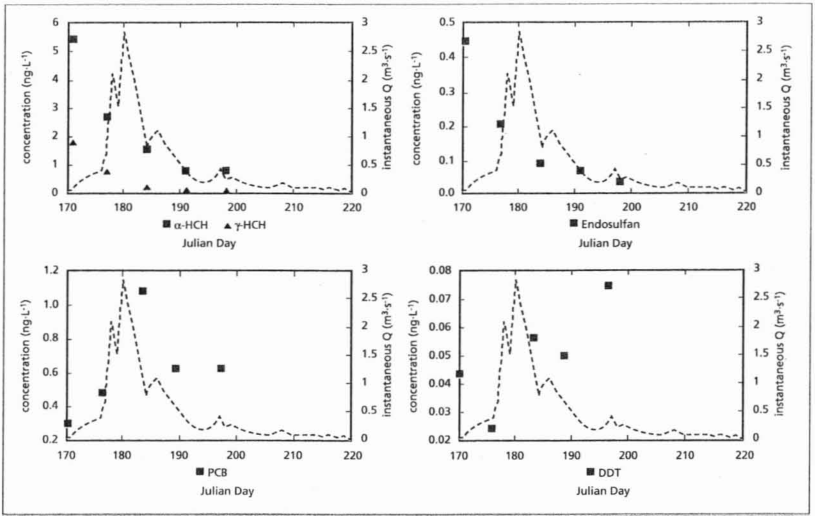


FIGURE 3.22

OC concentrations (ng·L⁻¹) in Gorge Creek in the Amituk Lake basin from June to August 1993. The dotted line is stream flow (m³·s⁻¹).

Freitas *et al.* (1997). Several versions of the model were developed, based on an understanding of Arctic limnology obtained during the Char Lake project (e.g., Rigler 1975, Schindler *et al.* 1974). These versions include steady- and unsteady-state simulations, and models to account for single chemicals and those that either speciate (e.g., mercury) or exist as a parent chemical with transformation products (multi-species model). The steady-state version of the model applied to Char Lake was calibrated with mass balance data for phosphorus; model results agree to within a factor of two. The steady-state version of the Amituk Lake model was calibrated using 1992 ΣDDT data. Results presented here illustrate Amituk Lake contaminant dynamics obtained with the steady- and unsteady-state versions of the model using 1992 and limited 1993 data. For 1992, measured and estimated water and sediment concentrations obtained with the steady-state version were generally within a factor of five for the six OCs modelled (Figure 3.24). For 1993, there was a greater difference between measured and predicted total sediment PCB concentration (Figure 3.24). Current efforts are underway to more extensively calibrate and test the Amituk Lake model using 1993 and 1994 data.

Model results suggest that in Amituk and Char Lakes, respectively, 50% and 30% of the meltwater passes through without mixing. The mass balance data presented in Table 3.13 and model estimates suggest that a very small fraction of total OC inputs are retained in the lake, and that the lake may currently be a source to the atmosphere of some chemicals (e.g., ΣPCB, Figure 3.25). The latter however, is difficult to verify since the amount that either volatilizes or is retained in the lake may be masked by analytical uncertainty at the prevailing low concentrations. Steady-state model estimates suggest that from 80% to more than 99% of most OCs contained in the snowpack are exported from Amituk Lake, with most of the remainder being buried or retained in the sediments. Inputs from atmospheric deposition and absorption directly to the open lake are negligible. Despite most chemicals being predominantly in the dissolved phase, losses due to volatilization are generally less than 5% of loadings due to cold temperatures and extended ice cover. PCBs may be an exception with up to 10% loss due to volatilization. These results indicate that Arctic lakes convey, rather than retain, chemicals deposited in the catchment area, unlike lakes in temperate regions that are sinks for particle-sorbed chemicals such as OCs. Thus, Arctic lakes behave similarly to rivers with respect to contaminants, with the Arctic Ocean acting as the recipient of most loadings.

Arctic lakes retain minimal contaminants not only because of the hydrologic regime, but because of their low productivity that results in very low suspended particle concentrations and sorptive capacity for contaminants. Suspended particle concentrations in Amituk Lake are 0.5 mg·L^{-1}, with an organic carbon content of 5% and 1.2% in suspended and bottom sediments, respectively. The low suspended particle concentration limits the amount of contaminant conveyed to the sediments, where the residence time is long as minimal fresh sediment is added annually. Because of the low organic carbon content,

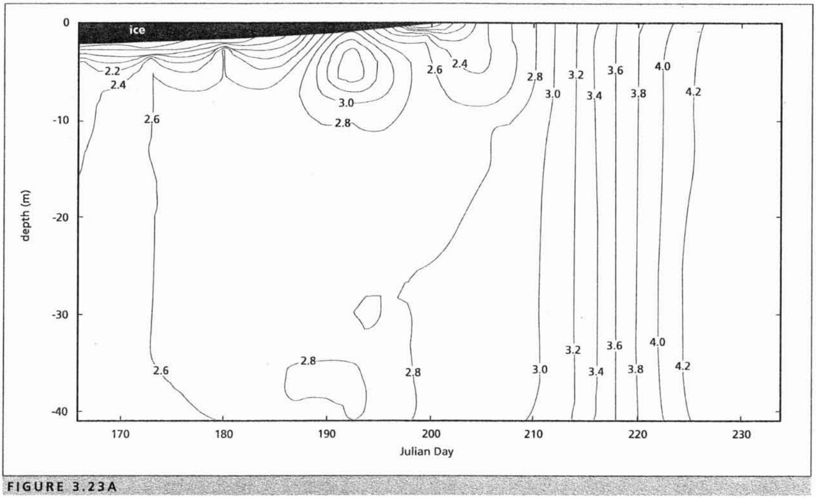


FIGURE 3.23A

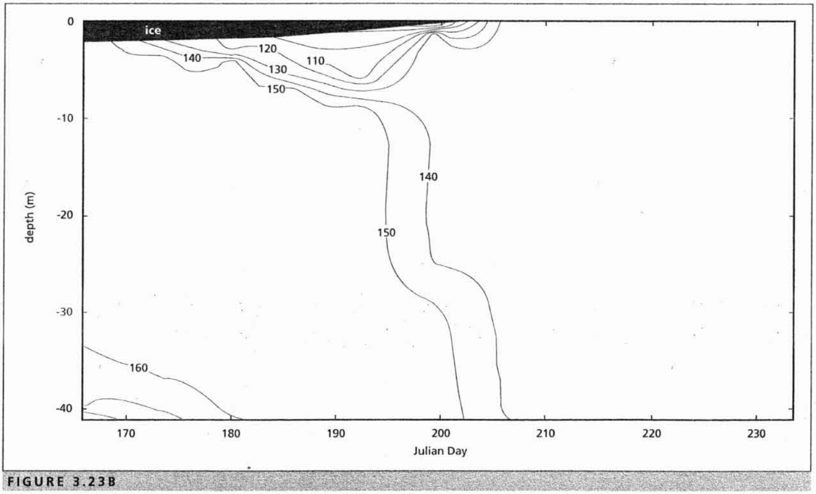


FIGURE 3.23B

Temperature (A) and conductivity (B) isopleths in Amituk Lake 1994. Note the development of isothermal conditions by day 210 (10 July 1994) and mixing of the water column.

it appears that relatively more contaminant sorbs to mineral matter than in systems with higher organic carbon levels. Thus, predicting partitioning between particulate and dissolved phases based on organic carbon (by using K_{oc} can give erroneous results. Similar partitioning behaviour has been noted in groundwater aquifers that typically have very low organic carbon, and is most pronounced for chemicals with low K_{ow} such as HCH (e.g., Calvert 1989).

A steady-state version of the multi-species model was used to consider PCB homologues, DDT and metabolites, and mercury that occur in different proportions in air, snow, lake water and sediment. For OCs, the analysis suggests three possibilities, of which all probably pertain. First, intermedia differences among proportions may be due to analytical uncertainty and differences among and within laboratories. Different proportions of PCBs and DDT are seen between laboratories, but also in similar media analysed at the same laboratory (e.g., snow, meltwater and lake water). At extremely low concentrations, such uncertainty would not be unexpected. Second, different proportions may reflect the differing scavenging and retention capacities of the media, with sediments having the highest sorptive capacity for the most heavily chlorinated PCBs, whereas snowmelt and water retain the less chlorinated, more soluble homologues. Third, the differences may be due to year-to-year variability, with the response to variability depending on the medium. For example, chemical proportions in snow change year-to-year and should be coupled to proportions in air, according to chemical-specific scavenging rates. Proportions in sediments integrate yearly variability and thus may differ from proportions in air, snow, and meltwater that respond rapidly to compositional changes. The water column is intermediate in its response time. This explanation should pertain to, for example, HCHs for which air concentrations are declining relatively rapidly. This explanation may also pertain to DDT and PCBs that have decreased in concentration, but more slowly than HCHs. For these chemicals,

TABLE 3.13

The mass budget of OCs dissolved ions, and DOC in Amituk Lake and surrounding snowpack for the period 18 June to 8 August, 1993.

	Snowpack[1]	Snowpack Cl^- Normalized[2]	Stream Input to Lake[3]	Ungauged Lake Input[4]	Lake Ice[5]	Snow on Lake Ice[6]	Sum Input to Lake[7]	Lake Outflow	Output-Input[8]	Bottom Sediment[9]
H_2O /10^6 (m^3)	3.085	4.864	4.998	0.516	0.766	0.091	5.605	55.358	−1.013	—
Cl^- (kg)	12 755	20 110	17 755	1830	—	435	20 020	20 110	90	—
DOC (kg)	4 190	6 605	1 665	170	—	75	1 910	1 715	−195	—
α-HCH (mg)	3 040	4 795	7 245	865	—	10	8 120	9 295	1175	5
β-HCH (mg)	200	315	170	20	—	5	195	220	25	—
γ-HCH (mg)	1 140	1 795	1 605	190	—	5	1 800	1 475	−325	10
ΣPCB[10] (mg)	3 200	5 045	3 120	370	—	70	3 560	2 545	−1015	180
HCB[11] (mg)	80	125	195	25	—	2	222	205	−17	25
Chlordane[12] (mg)	135	215	530	65	—	2	597	330	−267	22
ΣHept[13] (mg)	300	475	355	40	—	5	400	420	20	—
ΣChlord (mg)	430	680	885	105	—	7	997	750	−247	—
Dieldrin (mg)	205	325	345	40	—	5	390	300	−90	25
Endo I (mg)	890	1 405	655	80	—	10	745	500	−245	—
ΣDDT[14] mg	115	180	205	25	—	5	235	330	95	—

Notes:

[1] Snowpack SWE was calculated as follows. The watershed was divided into 200m x 200m grid cells. Using field surveys and photographic imagery, the relative snow distribution was discretized into the grid map (see Woo and Rowsell 1993). Using grid counts, the mean SWE for the overall watershed was estimated at 116.5mm in 1993. This compares to a winter precipitation (Sept 92–June 93) value of 100.8mm recorded by a standard, Nipher-shielded snow gauge in Resolute Bay. The snowpack chemical loading was a blend of deep valley snow and shallower snow on lake ice and on the upland plateau areas. The grid map results indicated that the deep valley snow, although less than 7% of the basin surface area, contributed about 30% of the basin SWE.

[2] The snowpack SWE and chemical burden are increased by a factor of 1.58. This value is the ratio of the Cl^- loading in the lake outflow to that in the pre-melt snowpack. The assumption is that Cl^- behaves conservatively in the basin and that the snowpack loading should equal the lake output.

[3] Stream and lake discharge were calculated from flow measurements and continuously- operating stream level recorders. Chemical loadings were calculated by: (i) summing the product of daily discharge and daily stream chemistry (major ions, nutrients), (ii) multiplying the cumulative daily stream/lake discharge by the discharge-weighted chemistry (OC compounds).

[4] Ungauged lake input = F x gauged stream input, where F = ungauged lake area/gauged stream basin area = 2.44/23.67.

[5] Lake ice thickness was measured at 2.2m on June 16, 1993. Ice melt volume = lake area x ice thickness x 0.92 where lake area = 0.38 x 10^6 m^2, ice = 2.2m and 0.92 is the ice density. Assume chemical content is negligible because of "freeze-out" effect during ice formation.

[6] Mean SWE (n = 8) in lake ice snow was 242mm. Chemical loading = SWE x lake area x mean chemical content.

[7] Sum Lake Input = gauged stream input + ungauged lake input + snow on lake ice + lake ice.

[8] A positive difference indicates an unmeasured input to the lakewater, such as groundwater input for Cl^- and atmospheric input for α-HCH. A negative difference implies a net loss for incoming chemicals relative to the lake output and would encompass such processes as sedimentation, biological uptake and/or increasing chemical content in the lake water column. The excess input of water relative to the lake outflow would contribute to lake storage and/or lake evaporation.

[9] Sediment fluxes from Muir *et al.* (1995).

[10] ΣPCB = sum of 67 congeners.

[11] HCB = pentaCB + hexaCB.

[12] Chlordane = *cis*-chlordane + *trans*-chlordane + *trans*-nonachlor.

[13] ΣHept = heptachlor + heptachlor epoxide.

[14] ΣDDT = DDT + DDE + DDD.

the sediments are likely not at steady-state with respect to inputs and the water column. This explains why, model-estimated α-HCH and ΣPCB sediment concentrations illustrated in Figure 3.24 underestimate measured sediment concentrations by about an order of magnitude.

The response time of the system to loading changes is illustrated by results obtained from an unsteady-state analysis. (Figure 3.26a, b). The results indicate that the water column, which is estimated to hold most of the recent chemical in the system, would clear itself within five years of the termination of loadings. The surficial sediments would take more than a decade to decline. These results indicate that these Arctic lakes generally will respond relatively rapidly to loading changes as a function of lake hydrology, because of the high chemical burden contained in the water column. Alternatively, the sediments chronicle major changes in loadings, lagging those changes by a decade or more.

The unsteady-state model was also used to estimate intra-annual changes in concentration (Figure 3.26c). Results of the analysis and field investigations suggest that cryoconcentration could increase contaminant concentrations in water by up to 12% to 15% during early May when ice thickness is maximal (Freitas *et al.* 1997, Semkin *et al.* 1996). Concentrations of contaminants in water then decline sharply as water from the melting ice and snow dilute the water column in late June and July.

Hg is the most important metal in Arctic lakes from a toxicology viewpoint. Similar to the OCs, it enters surface waters via snowmelt, with a negligible amount entering via direct atmospheric deposition to the lake during the brief (six-week) summer ice-free period. Most Hg entering the lake is presumed to be in a form (s) of Hg (II) that, because of its solubility, is readily scavenged by precipitation. As with other chemicals, a comparison of the amount of Hg that enters the lake with the amount exported depends on the extent of meltwater mixing in the water column. The model predicts that of the total Hg that enters Amituk Lake approximately 37% is retained in the sediments and 46% is exported downstream. The difference (17%) is likely lost to the atmosphere by gas exchange (see Section 2.4.1). These estimates are based on results obtained with the steady-state, multi-species model that was calibrated by correspondence of measured concentrations of Hg in Arctic char (Lockhart *et al.* 1993) and an estimated char concentration obtained using the modeled water concentration and a bioconcentration factor of 4.4×10^6.

Estimates of volatilization losses agree with research conducted on temperate lakes where they vary from 10% to 50% of annual inputs (Vandal *et al.* 1991, Watras *et al.* 1994). Hg cycling in the lake is maintained by species interconversion within the water column and surficial sediments. Specifically, the volatilization flux is supported by within lake reduction of Hg (II) to Hg° through, possibly, photochemical or biological reactions (Xiao *et al.* 1991, Silver 1984).

The magnitude and direction of the flux can be estimated simply by using the fugacity ratio:

$$f_w/f_a = C_w \cdot H/C_a R T_a \tag{3.1}$$

A value greater than one indicates net water-to-air exchange whereas a value less than one indicates net adsorption from air to water. For Amituk Lake, the influent stream concentration is 0.76 $ng \cdot L^{-1}$ and lake outflow is 0.52 $ng \cdot L^{-1}$ (Semkin, R.G., pers. comm.). We can assume an air concentration of 1.5 $ng \cdot m^{-3}$ that was measured at Alert (Schroeder 1996). Further, if we assume that between 1% and 3% of total Hg in water is Hg° (Watras *et al.* 1994), and using a Henry's

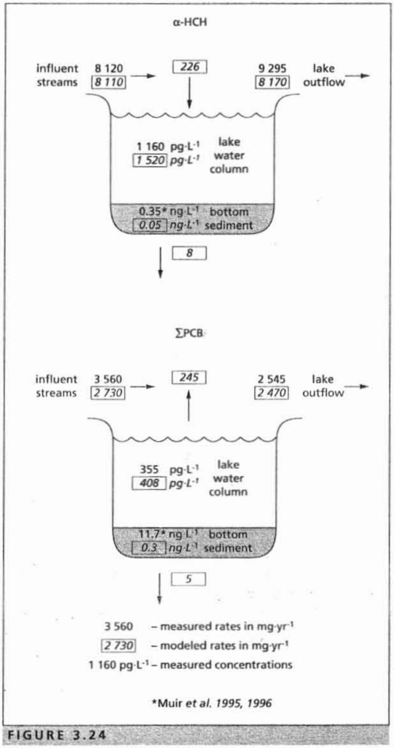


FIGURE 3.24

A comparison of measured and predicted OC concentrations in Amituk Lake water and sediment for 1993. Predictions were obtained using empirical water-suspended sediment partition coefficients.

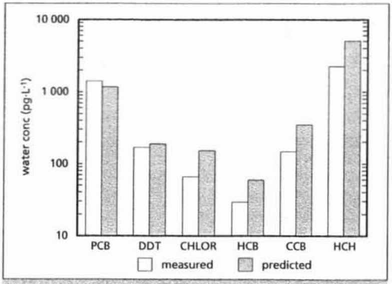


FIGURE 3.25A

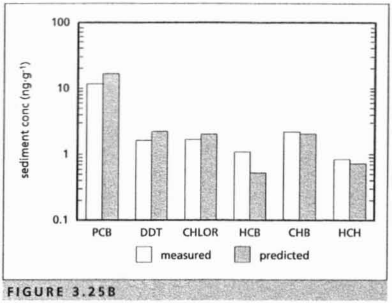


FIGURE 3.25B

A comparison of measured (1992) and predicted OC concentrations in Amituk Lake water and sediment. Predictions were obtained using empirical water-suspended sediment partition coefficients.

Law constant of 427 $Pa \cdot m^3 \cdot mol^{-1}$ at 3°C, we obtain a fugacity ratio of 2, that is consistent with net water-to-air exchange estimated from the whole lake mass balance.

The contaminant dynamics discussed above, have various implications for biota. In turn, the biology of the lakes has implications for contaminant dynamics. Since productivity in the lakes is extremely low, most contaminants are in the dissolved phase and a minimal amount is conveyed to the sediments. The high dissolved fraction suggests that bioavailability is enhanced (for example, uptake across the gills of fish). The effect of cryoconcentration could further enhance bioavailability since contaminants are excluded from ice that consumes up to 20% of the lake volume. Contaminants that enter the sediments (the more hydrophobic OC chemicals and mercury) are available to Char via benthic fauna, which is their major food source. Concentrations in benthos will respond slowly to changes in loadings due to the slow response time of sediments.

Yukon River Basin Study

In the Yukon aquatic system study, relatively high concentrations of toxaphene and some OCs have been detected in fish from Lake Laberge and a few other lakes in the Yukon River basin (Kidd *et al.* 1995b). This has led to a consumption advisory for the local fisheries (see discussion in Chapter 4). Studies from other lakes in southern Yukon, including Lake Hanson where toxaphene was applied, indicate lower concentrations of toxaphene in fish (Eamer 1994). There are several possible causes of elevated concentrations. First, Kidd *et al.* (1995a) hypothesized that differences among lakes are attributable to trophic structure, with fish feeding at higher trophic levels having elevated concentrations compared with those with lower concentrations. They substantiated their hypothesis with ^{15}N data illustrating differences in trophic level coincident with contaminant residues (see Chapter 3). However, the trophic level hypothesis was not able to explain all observed differences.

A second possibility for higher contaminant concentrations is the local use and disposal of organochlorine contaminants in the region. Records of historical use of organochlorine compounds that have been documented by the Yukon Contaminants Committee indicate that compounds such as DDT were used in aerial spraying between 1949 and 1969 (Nordin *et al.* 1993). Unknown quantities of household organochlorine pesticides and electrical components containing PCBs were abandoned in various sites along the Yukon River basin (Davidge 1994).

Finally, a third explanation is that inter-lake differences in limnological or physical processes may result in different contaminant concentrations among lakes and hence fish. Support for this hypothesis comes from the role that physical factors exert on contaminant dynamics (e.g., Diamond *et al.* 1996, Diamond, in press). The two latter possibilities were tested through an examination of field data and two mass balance studies.

Alaee *et al.* (1995) compared contaminant levels in water, suspended solids, and precipitation at Lake Laberge, Atlin, Bennet Fox and Kusawa lakes. Samples were collected during 1993 and 1994 during various seasons. Results for 1993 are presented in Table 3.14. α-HCH (0.3 to 0.48 $ng \cdot L^{-1}$) and γ-HCH (0.07 to 0.23 $ng \cdot L^{-1}$) isomers are the most abundant OCs in the Yukon River System. The observed concentrations are lower than those found in 1992/93 by Gregor and Eamer (1994) and by Alaee and Gregor (1994) during the winter of 1992/93. They are also lower than 1992 values reported for Amituk Lake by Falconer *et al.* (1995b) (1.3 $ng \cdot L^{-1}$ for α- and 0.28 $ng \cdot L^{-1}$ for γ-HCH) and 1990 samples in Lake Ontario (1–2 $ng \cdot L^{-1}$ for α-HCH and 0.5–0.75 $ng \cdot L^{-1}$ for γ-HCH) reported by Strachan *et al.* (1995).

Toxaphene concentrations in water and suspended sediments are also presented in Table 3.13. Water

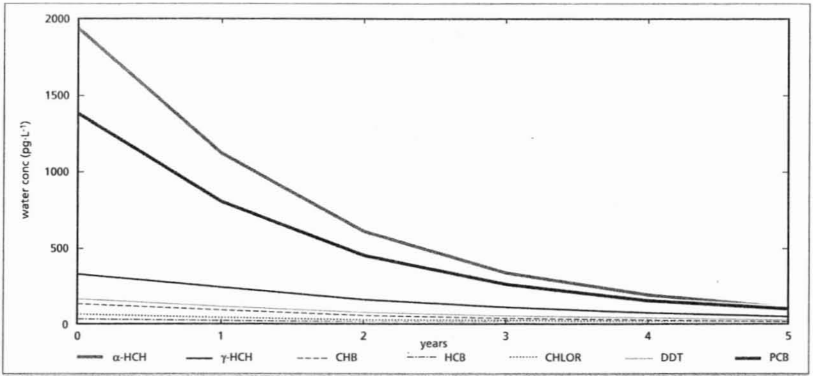


FIGURE 3.26A

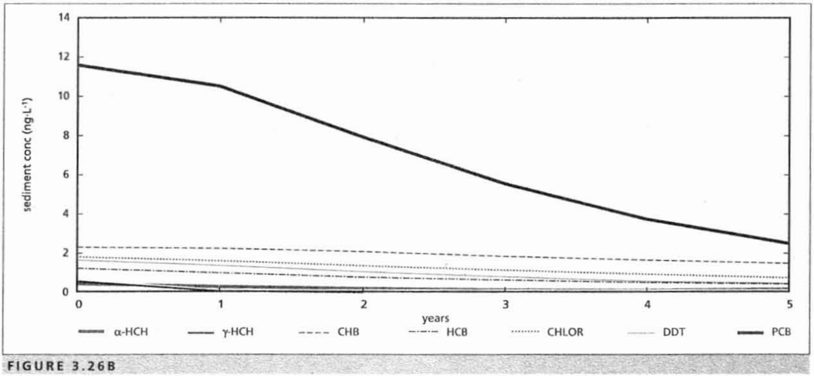


FIGURE 3.26B

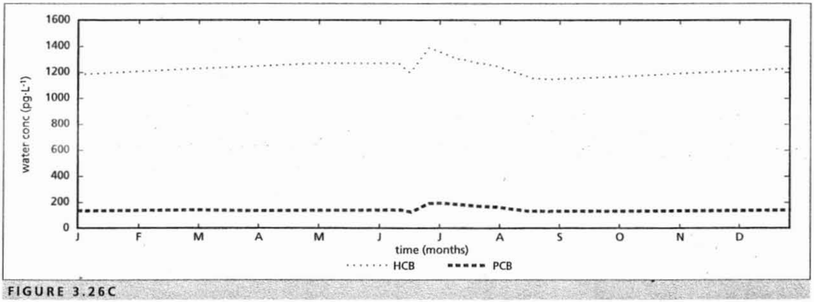


FIGURE 3.26C

Model predicted changes of the concentrations of ΣHCH and ΣPCB in (A) water, (B) sediment and (C) seasonal water column after chemical inputs are shutoff. An unsteady-state model was used (Freitas et al. 1997).

concentrations range from 0.02 to 0.27 ng·L^{-1} and were higher than those found in a preliminary survey done during 1992/93 (Alaee and Gregor 1994), but are closer to those observed by Bidleman *et al.* (1995b) for Amituk Lake (0.15 ng·L^{-1}) and by Muir *et al.* (1994a) for northern Ontario lakes (0.07 to 0.2 ng·L^{-1}). Toxaphene concentrations in suspended sediment from Lake Laberge are considerably higher (9 ng·g^{-1}) than those of the Takhini River (0.12 ng·g^{-1}). However, since the concentration of suspended sediments in Lake Laberge (0.61 mg·L^{-1}) is lower than in the Takhini River (460 mg·L^{-1} in June and 62 mg·L^{-1} in August), total water concentrations in the two areas are similar.

The similarities among toxaphene and HCH concentrations in the Yukon River basin lakes suggest that the main contaminant source is atmospheric and that there is no major local source to the lakes. To test further for the possibility of local sources, soil samples were collected from the Range Road dump site, a suspected point source for Lake Laberge. Soil samples from near Kusawa Lake and elsewhere near Lake Laberge were taken for comparison (Sumi 1995). The Range Road site samples had significantly higher concentrations of PCBs (77 ng·g^{-1}) compared with those from Lake Laberge (12 ng·g^{-1}) or Kusawa Lake (10 ng·g^{-1}). Those from Kusawa Lake and Lake Laberge had PCB congener profiles typical of atmospheric samples — a dominance by the less chlorinated congeners. Samples collected from the Range Road site, on the other hand, were dominated by the higher chlorinated congeners, particularly the penta- through hepta-substituted ones and one sample had an almost perfect match to Aroclor 1260 — a commercial mixture of PCBs (see Figure 3.27). This pattern is usually found in unweathered material. These observations indicate that PCBs have been released at the Range Road site and could be a local source, however samples collected from the lake sites indicate atmospheric deposition is the dominant source.

Two mass balance studies were conducted for the Yukon. Figure 3.28 illustrates results for HCHs in Lake Laberge that were compiled from measured inflow and outflow rates, estimates of air/water exchange (Mackay *et al.* 1994) and sediment burial (calculated from measured bulk sediment burial measurements reported by Lockhart *et al.* 1994). Lake Laberge receives over 90% of HCH loadings from the Yukon and Takhini rivers. Direct atmospheric deposition to the lake contributes a negligible amount. Nearly all chemicals are exported downstream via the Yukon River. Due to the low particle-reactivity of HCHs, minimal chemical is retained in the sediment. Air-water exchange is also minimal. Model results indicate that Lake Laberge receives its loadings from riverine sources. The results do not allow us to distinguish between atmospherically deposited chemicals and those that may come from local sources upstream of the lake, such as the Range Road site.

A second mass balance study was conducted for Tagish, Atlin, Bennet and Marsh lakes (Kawai 1995, Diamond *et al.* 1996). The model is based on the QWASI fugacity/aquivalence approach. The hydrology of the system was obtained from Environment Canada (1990), Kirkland and Gray (1986) and Carmack *et al.* (in prep). Air-water mass transfer coefficients were taken from Mackay *et al.* (1994) but adjusted to reflect the ice-free fraction of the year from May to November. Sediment burial rates for the lakes were estimated from those for Laberge, Little Atlin, Fox and Kusawa lakes. They ranged from 81 to 812 g·m^{-2}·y^{-1} (Lockhart 1994). Because the lakes have not been well characterized (e.g., particle budgets), the model is preliminary. Running the model with contaminant concentrations in snow (Swyripa and Strachan 1994) and comparing model results with measured lake concentrations (Alaee and Gregor 1994, Gregor and Eamer 1993), the results indicated similar chemical concentrations in the lakes, with Bennet Lake having slightly higher water concentrations as it receives the accumulated inputs from the three upstream lakes. Similarly with Lake Laberge, system hydrology largely controls loadings from the catchment areas to, and dynamics within, these low

TABLE 3.14

Average OC concentrations in waters of the Yukon River basin observed in 1993.

Location	α-HCH mean (ng·L^{-1})	γ-HCH mean (ng·L^{-1})	α/γ ratio	Toxaphene		ΣPCB Soil (ng·g^{-1})
				Water	Sus. Sed.	
				(ng·L^{-1})		
Llewellyn Glacier	0.32	0.15	2.1	0.06	—	—
Atlin Lake	0.39	0.2	1.9	0.27	—	—
Tagish Lake	0.39	0.15	2.6	0.02	—	—
Bennett Lake	0.34	0.11	3.1	0.11	—	—
Marsh Lake	0.48	0.2	2.4	0.18	—	—
Kusawa Lake	0.42	0.07	6	0.20	—	10
Takhini River	0.39	0.19	2.1	0.10	29 (June) 6.2 (Aug.)	— —
Lower Yukon River	0.44	0.23	1.9	0.16	—	—
Yukon River above Whitehorse	0.48	0.21	2.3		—	77
Yukon River below Whitehorse	0.45	0.16	2.6		—	Range Rd
Fox Lake	0.39	0.15	2.6	0.11	—	—
Lake Laberge	0.4	0.17	2.4	0.21	5.6	12

productivity lakes; air-water and sediment-water exchange are minimal (Figure 3.29). Overall, the results did not support the explanation that different OC concentrations in fish could be due to major physical or limnological differences among lakes, although, as mentioned above, limited limnological data are available for the system. The analysis suggests that it may be useful to investigate the role of glaciers as "cold traps" since Atlin Lake, which has elevated toxaphene concentrations in burbot, receives 90% of its inflow from the Willison and Llewellyn Glaciers located in the Coast Mountains, unlike the remaining three lakes studied that do not have elevated concentrations in fish (Diamond *et al.* 1996).

Radionuclide concentrations in northern surface waters have been summarized by Lockhart *et al.* (1992) and are presented in Table 3.15. The larger water bodies of Baker Lake and Great Slave Lake show radionuclide concentrations typical of atmospheric fallout across Canada. In the vicinity of mining activities (Sherman Lake and Great Bear Lake near Port Radium), some radionuclide concentrations in the water exceed the Canadian background levels (Table 3.15) It has not been determined whether this is due to mining activities or naturally enriched background levels.

Lake Sediment Observations

The final set of studies related to arctic and sub-arctic lakes concerns a geographic survey of contaminant concentrations in lake sediments. Since lake sediments integrate chemical inputs from catchment areas, they

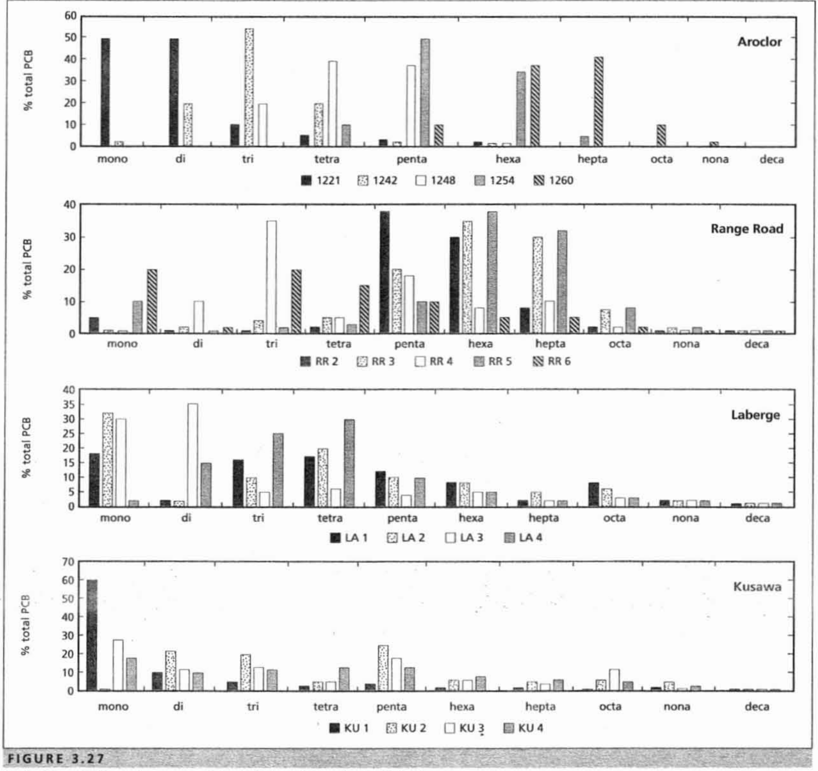


FIGURE 3.27

The fractional PCB homologue composition in Yukon soil samples (Sumi 1995)

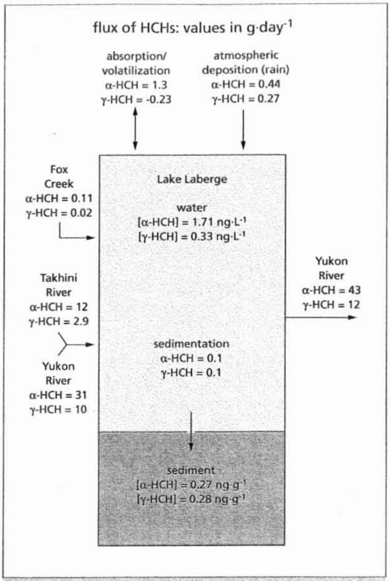


FIGURE 3.28

The mass budget of ΣHCH (α plus γ) in the Lake Laberge aquatic system during summer of 1993.

have long been used as indicators of chemical status (for example, elemental analysis of lake sediments is used as a geological prospecting tool). Sediments from the deep portion of lakes provide the best indication of chemical status since fine particles, with their high surface area (and hence ability to sorb chemicals), accumulate there. Sediments provide a useful indication of temporal trends of contaminant inputs to a lake, however, the temporal resolution of the sediment chronology is limited by the extremely low rates of sediment accumulation. Care must also be taken when comparing sediment concentrations lake-to-lake, since the proportion of total loadings to the system, which accumulates in sediment is lake-specific. As seen for Amituk Lake, this can be a very small fraction of total loadings. Sediments provide a reasonable estimate of water column concentrations for chemicals that have had relatively constant inputs over several decades (i.e., the water column and sediments are near steady state).

Muir *et al.* (1996) conducted a north-south transect of OCs in six NWT lakes — two near Chesterfield Inlet, two from Cornwallis Island and one each from southwest Axel Heiberg and northern Ellesmere Islands. In addition, Mudroch *et al.* (1992) examined sediments from Great Slave Lake for the same compounds. PAHs have been measured in sediments of several lakes from both territories (Lockhart *et al.* 1995). The temporal aspects of contaminant history are discussed in Section 2.5 of this chapter; comments here are restricted to the surficial, relatively recent record of concentrations reflected in the top 1 to 2 cm of sediments. This sediment section represents a

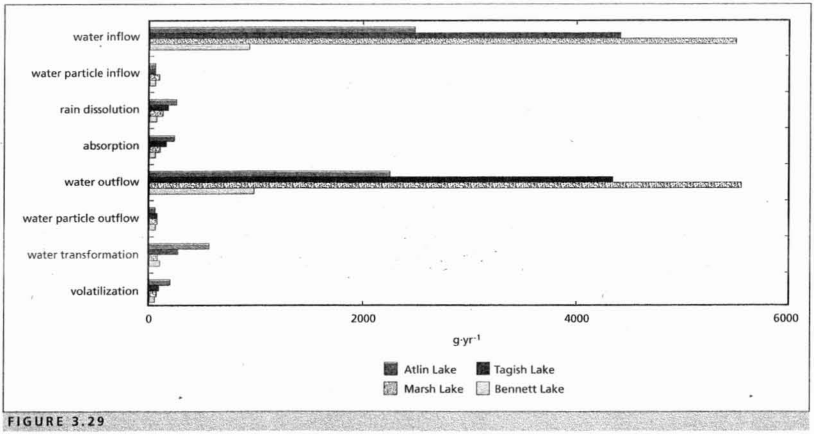


FIGURE 3.29

Rates of movement of α-HCH in lakes of the Yukon River basin estimated by a steady-state mass balance model (Kawai 1995).

range of years dependent on the lake-specific sediment accumulation rate.

As seen in Table 3.16, most sediment contaminant concentrations are similar over the large geographic range of sampling. The concentrations of ΣHCHs in sediment (mean: 1·0 ng·g^{-1} ± 70% rsd) is one-tenth to one-quarter of Lake Ontario measurements (1980s: 4 ng·g^{-1}, Eisenreich *et al.* 1989, 1990s: 11 ng·g^{-1}, Strachan *et al.* 1995). Mean ΣDDT (0.9 ng·g^{-1}) and mean HCB (0.8 ng·g^{-1}) concentrations in Arctic lake sediments are 1% to 2% of the Lake Ontario values, respectively. Muir *et al.* (1995) reported concentrations for HCHs, DDTs, chlordanes and HCB for several lakes in the Experimental Lakes Area (ELA), northwestern Ontario that were 1.3, 10, 2 and 0.5 times greater than those of Arctic lake, respectively.

Summary

The results of studies of Arctic and subarctic lakes conducted under the NCP indicate that most lakes receive contaminants by long-range transport, with snow accumulation in catchment areas capturing and conveying contaminants to the lakes. The lakes retain only a small fraction of the total OC inputs to the catchment area, the specific fraction being a function of lake hydrology (e.g., the timing of snowmelt vis-a-vis lake turnover) and productivity (e.g., the fraction of contaminants conveyed to the sediments by suspended particles). They act more as rivers, channelling the year's accumulated contaminant inputs downstream. These simple systems point to the interaction between biological productivity and contaminant dynamics: the low levels of biological productivity and organic carbon result in minimal retention of contaminants (especially the more hydrophobic OCs and mercury). In more productive systems, a higher proportion of contaminant loadings are retained in the system and are, in turn, available for uptake through the food web. The effect of higher contaminant retention is tempered somewhat by dilution by the large pool of organic matter and biomass. In Arctic and Subarctic lakes however, the OCs and mercury retained in the lake will tend to accumulate in the limited organic and biotic mass. It still remains to estimate the fraction of contaminants that remain in the snowpack during winter as melt proceeds. In addition, the amount of contaminants transported downstream during spring melt remains poorly known. In these Arctic lake systems, the snowpack and the lake act as temporary sinks for contaminants in the winter. Over the winter, a fraction of the contaminants may volatilize to the atmosphere; however, the majority of contaminants pass through the lakes and downstream during spring/summer melt.

- *The suspended sediment load of the Mackenzie River is the major source of PAHs to the Beaufort basin of the Arctic Ocean. The PAHs are primarily natural in origin.*
- *In arctic rivers most of the burden of OCs is carried in the dissolved phase. The contribution of the suspended solid fraction to the total OC concentration was less than 10%. The relative abundance of OCs in arctic rivers is* $\Sigma HCH \approx \Sigma\, PCB > \Sigma DDT \approx \Sigma Chlordane$.
- *A detailed basin study of OCs and metal pathways for Amituk Lake in the high Arctic indicates that unlike lakes in temperate regions that are major sinks for particle-sorbed chemicals such as OCs, individual arctic lakes retain little of the inflowing OCs. However, systems of arctic lakes may well represent a substantial sink when their cumulative effect is taken into account.*
- *Hg is the most important metal in Arctic lakes from a toxicology viewpoint. Observations show and models confirm that of the total Hg that enters a high arctic lake (e.g. Amituk Lake) about one third is retained in the sediments, half is exported downstream and the rest is lost to the atmosphere.*
- *The main source of toxaphene, ΣPCB and HCH to Yukon River basin lakes is atmospheric rather than local dumping.*
- *Lake Laberge receives over 90% of HCH loadings from the Yukon and Takhini rivers.*
- *For Tagish, Atlin, Bennett and Marsh lakes in the Yukon, system hydrology largely controls loadings from the catchment areas to — and dynamics within — these low productivity lakes; air-water and sediment-water exchange are minimal. Overall, the results did not*

TABLE 3.15

Radionuclide concentrations in water of northern lakes.

Water body	Titium (Bq·L^{-1})	^{137}Cs (mBq·L^{-1})	^{226}Ra (mBq·L^{-1})	Total U (µg·L^{-1})	Reference
Background values (Canada-wide)	5–12	4	1–13	0.2–5	Baweja *et al.* 1987
Baker Lake, NWT	12 ± 5	4 ± 2	1.6 ± 0.7	0.16 — 0.08	Baweja *et al.* 1987
Great Slave Lake, NWT	—	1.3–3.6	0.8–2.8	—	Elliot *et al.* 1981
Port Radium Area (Great Bear Lake)	—	—	~10	1–400	Kalin 1984
Sherman Lake, NWT (Rayrock Mining Site)	—	—	5–230	1–5	Veska and Eaton 1991
Beaverlodge Lake, Sask. (uranium mining site)	—	—	40–80	200–400	Swanson 1983
Maximum allowed in drinking water		60 000	1000	100	Federal/Provincial Working Group 1978

A value reported as 12 ± 5 indicates a mean ± 1 SD in a series of measurements.

A value reported as 5–12 merely indicates a range; there were inadequate data to calculate a meaningful average.

A value reported alone indicates only one measurement was reported.

TABLE 3.16

Concentrations of persistent OCs[1] ($ng \cdot g^{-1}$ dry wt) and lead ($ug \cdot g^{-1}$ dry wt) in surface sediments (cores or grab samples) from Canadian arctic lakes (Muir *et al.* 1995a,b, 1996, Evans *et al.* 1996, Lockhart and Muir 1996).

Lake Name	Region	lat N	long W	Type	N		% Org C	HCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene[2]	Lead
Peter	NWT	63°05'	93°47'	grab	5	Mean	6.5	0.26	0.26	0.27	0.39	7.2	—	—
Hawk	NWT	63°38'	90°42'	Core	1		12.8	1.8	2.9	4.0	5.1	38	—	47.6
Far	NWT	63°42'	90°40'	Core	1		3.0	0.29	2.5	0.82	1.5	19	—	31.9
Ste. Therese	NWT	64°35'	121°20'	Core	1		3.1	1.0	0.56	0.32	0.91	12	—	10.5
Belot	NWT	66°50'	126°16'	Core	1		5.0	0.11	0.27	1.3	0.91	27	4.9	14.2
Amituk	NWT	75°00'	93°46'	Core	1		1.4	1.1	0.91	1.8	1.7	12	—	20.7
Sophia	NWT	75°00'	93°35'	Core	1		3.0	0.09	0.05	0.53	0.34	4.0	—	12.4
Buchanan	NWT	80°00'	89°00'	Core	1		1.3	1.3	3.1	0.69	0.77	16	—	25.9
Hazen	NWT	81°45'	71°30'	Core	1		1.7	1.0	0.57	0.11	0.10	2.4	—	20.4
Yaya	NWT	69°10'	134°38'	Core	1		—	1.4	0.23	0.30	0.56	13	—	27.5
Great Slave – 19B	NWT	61°25'	114°25	Core	1		1.5	1.1	0.31	0.26	0.41	8.8	1.3	1.3
Great Slave	NWT	61°14'	113–114°	grab	10	Mean	—	0.39	0.40	0.30	0.38	4.1	—	—
– Slave River Delta	NWT					SD	—	0.18	0.12	0.15	0.19	2.0	—	—
Great Slave	NWT	62°10'	110–113°	grab	15	Mean	—	0.29	0.09	0.10	0.12	1.0	—	—
– east basin	NWT					SD	—	0.18	0.05	0.06	0.08	0.3	—	—
Little Atlin	Yukon	60°15'	133°57'	Core	1		—	0.86	0.23	3.8	3.5	28	—	9.1
Kusawa	Yukon	60°20'	136°22'	Core	1		1.7	0.08	0.15	0.03	0.05	3.1	0.1	21.6
Laberge	Yukon	61°11'	135°12'	Core	2	Mean	—	0.61	0.35	0.38	0.93	7.9	0.2	17.5
Fox	Yukon	61°14'	135°28'	Core	1		—	0.33	0.93	0.05	0.89	31	1.3	5.0
Lindeman	Yukon	59°47'	135°02'	Core	1		—	0.10	0.62	0.29	0.90	12	0.0	44.9

[1] ΣHCH = Sum of α-, β-, and γ-HCH. ΣCHL = Sum of *oxy-*, *cis-*, *trans*-chlordane, heptachlor epoxide and *cis-*, *trans*-nonachlor.
ΣDDT = Sum of *o,p'*-DDT, *p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE.
ΣPCB = Sum of CB 8/5, 18, 17, 16/32, 22 , 31, 28, 33, 52, 49, 47/48, 44, 61/41/71, 40, 64, 70/76, 74, 66/95, 60, 91, 101, 99, 83, 97, 87, 85, 110, 118, 151, 149, 146, 153, 141, 138, 158, 178/129, 187, 183, 174, 185 , 177 , 180, 170, 200/157, 198, 199, 196, 195, 194, 206

[2] Toxaphene determined by high resolution negative ion mass spectrometry.

support the explanation that different OC concentrations in fish could be due to major physical or limnological differences among lakes. This is similar to the Lake Laberge situation.

- *The order of abundance of OCs in northern lake surface sediments is ΣPCB > ΣDDT > ΣHCH ≈ toxaphene ≈ ΣChlordane.*

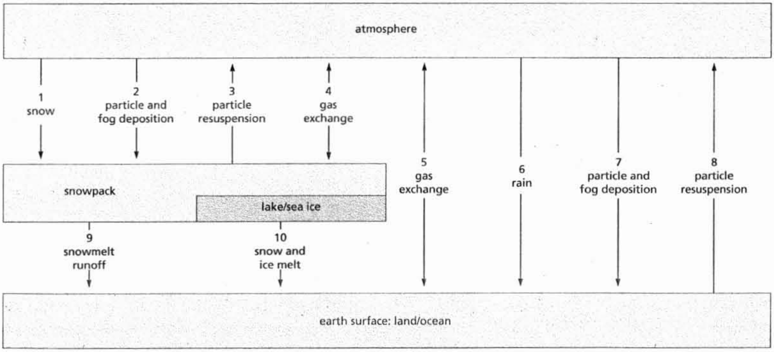
2.4 Inter-compartmental Exchange Processes

2.4.1 Atmosphere-Surface

Figure 4.1 summarizes schematically the major pathways for contaminants exchange between atmosphere and surface, as well as the current state of knowledge of flux equations for these pathways. Transfer from atmosphere to the Earth's surface in the polar regions is complicated by the presence of seasonally varying intermediate media, namely, snowpack and ice. In the Arctic Ocean, the marginal seas undergo the greatest variations in percentage of ice cover (see Section 2.3.3).

The snowpack plays an important role in atmosphere-surface exchange. The snowpack accumulates contaminants deposited by fresh snow during winter, plus those delivered by particle dry deposition and gaseous exchange. Gaseous exchange may be negative (surface loss) or positive (surface gain). Currently, there is no way of measuring it directly. However, there are indirect ways that can be used to infer gas exchange (see below). The net exchange between the atmosphere and the Earth's surface is the result of all these processes. A specific example of this is discussed in Section 2.6.1 on HCHs.

A major experimental effort to understand partitioning of organic contaminants to snow and rain has been undertaken in the NCP by Hoff *et al.* (1993a, 1995). Partitioning of volatile compounds in air to water and ice was investigated over a range of temperatures using inverse gas chromatography, monitoring vapour concentrations by gas chromatography or by solid phase micro-extraction. The concentrations in snow were determined by purging the meltwater with a carrier gas. It was concluded that sorption by snow can be predicted by extrapolating adsorption at the air-water interface to lower temperatures, and a formula was proposed for estimating the interfacial partition coefficient (K_{ia}) values as a function of temperature. An accurate method for measuring the specific surface area (SSA) of snow was also developed based on adsorption of nitrogen. Preliminary data for relatively wet snow in southern Ontario suggest that the SSA can be estimated from the size and geometry of ice crystals in snow. Taken together, these findings indicate that falling snow efficiently removes organic contaminants from the atmosphere, and that snow acts as a temporary storage medium for contaminants on the ground. Examples are given in the following sections to illustrate the relative scavenging efficiency of snow, rain and particles in the atmosphere for OCs, as well as the changes in OC content of snowpack in sorption



flux equations

1. $C_s \times P_s$
2. $V_d \times C_p$
3. ?
4. ?
5. $K(C_g - C_w/H)$ water
   ? land
6. $C_r \times P_r$
7. $V_d \times C_p$
8. ?
9. $C_{runoff} \times P_{runoff}$
10. $C_{runoff} \times P_{runoff}$

FIGURE 4.1

Schematic of processes of atmosphere-surface exchange (pathways TF-A and A-TF in Figure 1.2).

equilibrium with atmospheric OCs during spring warming. It should be emphasized that these calculations are speculative in that they involve extrapolating data for volatile compounds (10^{-1} to 10^{-4} Pa) to a lower range of vapour pressures (10^{-5} to 10^{-10} Pa). An experimental program is currently underway to test the accuracy of the K_{ia} formula for low vapour pressure contaminants and to define the SSA of Arctic snow and its rate of decrease during ageing.

We consider each pathway in turn according to its numerical sequence in Figure 4.1. The flux (F) of material between the atmosphere and surface is defined as the product of a measured contaminant concentration (C) or a concentration difference (ΔC) and an exchange coefficient (e.g. mass transfer coefficient K, dry deposition velocity V_d, or precipitation rate P). NCP research has largely focused on determining contaminant concentrations, then estimating inputs from atmosphere to ecosystems using exchange coefficients, available from the scientific literature.

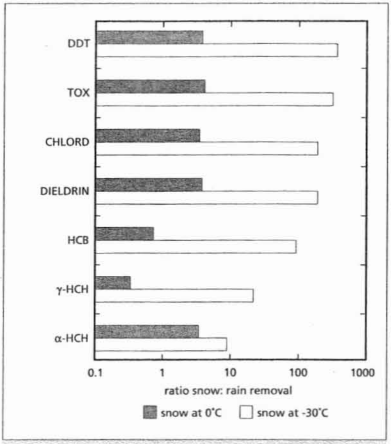


FIGURE 4.2A

The ratio of OC mass-scavenged from the atmosphere by snow at (0°C and -30°C) to that by rain at 0°C when the OC is allowed to partition between the gas and particle phase at conditions typical of arctic haze in April to May.

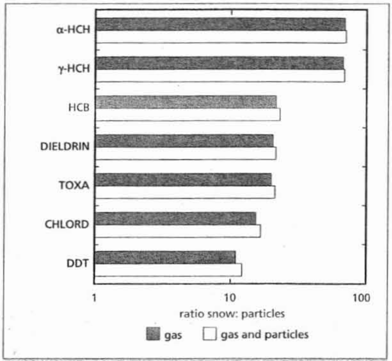


FIGURE 4.2B

The ratio of OC mass deposited by snow at -30°C compared with that by particle dry deposition when snow scavenges: (i) gaseous OC only (ii) gaseous and particulate OC. Parameters used in the calculation were surface area to volume ratio of rain: 2.4×10^4 and snow: 1×10^5 m^2 (hydrometeor)·m^{-3} (air); aerosol atmospheric surface area: 3×10^{-5} $m^2 \cdot m^{-3}$ (air); precipitation rate: 10 mm $H_2O \cdot month^{-1}$; particle dry deposition velocity 0.1 $cm \cdot s^{-1}$. Particle scavenging by rain and snow calculated according to methodology of Gong et al. (1996) assuming drizzle raindrops and platelet ice crystals.

2.4.1.1 Precipitation Scavenging

Deposition of contaminants by precipitation is assumed to be a one-way process from atmosphere to surface. The deposition flux (F) is the product of the concentration of contaminant in snow (C_s) or rain (C_r) and the rate of precipitation (P_s). Contaminants are incorporated into snow and rain in two forms — as particles or as a gas (Barrie 1991). Gas sorption by snow is a process that only recently, with support of the NCP, has been quantified. The work of Hoff *et al.* (1995) mentioned above was used to estimate gas exchange between the atmosphere and snowpack. The relative importance of rain and snow scavenging (Figure 4.1, steps 1 and 6) of OCs is predicted from model calculations and illustrated in Figure 4.2a. The ratio of OCs scavenged from the atmosphere by snow (at 0°C and -30°C) to that by rain at 0°C was calculated using the theories of Hoff *et al.* (1995) for gas to snow sorption, Pankow for particle-gas partitioning (Pankow 1987, see Section 2.2.3), and Gong *et al.* (1997) for particle scavenging by hydrometeors. With the exception of γ-HCH and HCB at 0°C, snow is much more efficient than rain in scavenging OCs at both temperatures. At -30°C, increased sorption of gaseous OCs makes snow a very much more efficient scavenger than rain at 0°C. This is consistent with the conclusions of Sparmacher *et al.* (1993).

The fate of OCs in snow after deposition, when snow surface-area decreases as a result of snow pack metamorphosis processes is unknown. There are no detailed studies of the variation in C_s on the ground within hours to days following deposition. The only indication that such processes may lead to dramatic changes in C_s are the observations of Patton *et al.* (1989) on the Canadian Ice Island in June 1986, which showed concentrations of γ-HCH and α-HCH in surface snowpack that were lower than in fresh snow by a factor of 2 and 6, respectively.

Measurements of C_s have been made in the Arctic at many locations on a weekly basis (see Section 2.3.2). Post-depositional changes on time scales of hours to days have not been resolved because the sampled snowfall remains in an open bucket for a week. Post-depositional losses during the week cannot be excluded. Where possible, snowfall sampling sites were co-located at sites where weekly observations of air concentrations (C_a) were taken (Alert, Tagish and Cape Dorset, Figure 1.1).

The cumulative flux from atmosphere to surface (ΣF_i) via snow is calculated as follows:

$$\Sigma F_i = C_{vwm,i} \cdot \Sigma P_i \qquad (4.1)$$

where $C_{vwm,i}$ is the precipitation amount weighted mean concentration of the contaminant in the snowmelt and ΣP_i is the total precipitation amount for the period. If the units of $C_{vwm,i}$ and P_i are ng·L^{-1} and mm snow water equivalents, respectively, the units of F_i are ng·m^{-2} for the period. Assuming that a large fraction of the winter snowfall is sampled and that the snow is sampled frequently enough that losses are not significant, cumulative flux (ΣF_i) is an estimate of the cumulative input of a contaminant to the snowpack by snow (Figure 4.1, step 1). ΣF_i based on observations at various locations during the winters of 1991/92 through 1993/94 are shown in Table 4.1. Observations at Fraser have not been included in determining the overall mean, since the air mass contributing to snowfall at this location comes mainly from the Pacific and because annual snowfall is many times greater than at the other sites. In addition, two anomalously high values (HCB at Alert in 1993/94 and ΣDDT at Whitehorse in 1991/92) were omitted from the calculation of the mean. With these values excluded, the means are as shown. The contributing values for ΣHCH fall into two groups — results for Alert and Eureka (the farthest north), and, all others (the mean of the fluxes without these three northern values is almost halved, to 0.18 μg m^{-2} and the standard deviation is 0.06 μg m^{-2}.) Since the concentrations are elevated for these locations, and the snowfall deposition is actually lower than the other locations, the flux differences can be attributed to either an improved scavenging and retention at the colder temperatures or to greatly different concentrations in the air masses. Since atmospheric concentrations of HCH at Alert (Figure 3.1) and Tagish (Figure 3.2) are comparable, the former explanation is the most probable one and is consistent with the modelled predictions (Figure 4.2) that snow is a more efficient scavenger of OC contaminants at lower temperatures. Temperatures in the high Arctic are generally colder for a longer period than in the mainland Arctic. This may explain higher observed deposition of Σ-HCH at Alert and Eureka (Table 4.1).

Observed deposition of contaminants in arctic snowfall can be compared to the wet deposition in the Great Lakes (Hoff *et al.* 1996). For example, in the Lake Superior region the winter ΣHCH and HCB fluxes are similar to those in the Arctic while ΣDDT and ΣPCBs fluxes in the Arctic are somewhat higher (but not significant statistically) than those observed for Lake Superior.

Even though snow may scavenge OCs more efficiently than rain (Figure 4.2), the role of rain as a sink for atmospheric OCs should not be neglected. In the Arctic on average, rain makes up about 45% of the total annual precipitation (Barrie *et al.* 1992) but this ranges (Table 4.2). Measurements of C_r for OCs, PAHs or metals have not been made in the Canadian Arctic. C_r is likely highest in summer, as might be expected for some volatile contaminants. In the

TABLE 4.1

Cumulative flux (ΣF_i, equation 4.1) of contaminants from the atmosphere to the Earth's surface (pathway 1, Figure 4.1) for the snow season (see Figure 3.12, Section 2.3.2). Units are (μg·m^{-2}·season^{-1}). Bold values are outliers. For a definition of OC group constituents see the footnote of Table 3.4.

Station/Sample Year	Deposition				
	ΣHCH	ΣChlord.	ΣDDT	HCB	ΣPCBs
Alert, NWT (93/94)	0.50	0.01	0.05	**0.11**	0.37
Alert, NWT (92/93)	0.88	0.03	0.05	0.06	0.19
Eureka, NWT (91/92)	0.78	<0.01	0.01	0.01	0.26
Mould Bay, NWT (93/94)	0.25	<0.01	0.03	0.02	0.23
Cape Dorset, NWT (93/94)	0.22	0.01	0.06	0.01	0.51
Snare Rapids, YK (93/94)	0.18	0.01	0.03	0.02	0.33
Dawson City, YK (93/94)	0.19	0.03	0.04	0.01	0.41
Whitehorse, YK (93/94)	0.12	0.02	0.02	0.01	0.20
Whitehorse, YK (92/93)	0.07	0.04	0.05	<0.01	0.43
Whitehorse, YK (91/92)	0.16	0.06	**0.35**	—	0.75
Tagish (93/94) (Atlin, BC)	0.20	0.03	0.04	0.01	0.35
Tagish (92/93) (Atlin, BC)	0.28	0.09	0.01	0.01	0.24
Arithmetic Mean	0.31	0.03	0.04	0.02	0.35
Standard Deviation	0.25	0.02	0.02	0.02	0.15
Fraser, BC (93/94)	1.0	0.11	0.13	0.02	1.1
Fraser, BC (92/93)	1.4	**0.91**	**0.51**	0.05	2.1

TABLE 4.2

Mean arctic precipitation and temperature data for 1990 to 1995 for the summer (June to Sept.) and winter (Oct. to May) portions of the year.

	Snow Depth (cm)	Total Pptn. (mm)	Rain (%)	Average Termperatures (°C)	
				Summer	Winter
Mould Bay, NWT	86	102	23	–0.1	-25.5
Cape Dorset, NWT	261	359	35	+3.4	-17.3
Dawson City, YK	190	327	61	+11.9	-13.7
Whitehorse, YK	182	278	55	+11.8	-5.9
Tagish[1], BC	183	363	49	+11.6	-4.7
Fraser, BC	900	1264	29	+9.1	-6.5

[1] *data from Atlin (closest Atmospheric Environment Service weather station).*

absence of measurements of C_r, precipitation scavenging of particulate and gaseous organic compounds can be estimated using air concentrations and the scavenging relationships discussed in Section 2.6.2.

2.4.1.2 Particle and Fog Deposition (Figure 4.1, Steps 2 and 7)

The flux of contaminants from the atmosphere to the Earth's surface by particle dry deposition is the product of the particle deposition velocity (V_d), the particle concentration in air (C_a) (Tables 6.3 and 6.4), and the fraction of the compound sorbed to aerosols. V_d depends on particle size and physical properties such as hygroscopicity (ability to absorb water). This flux measurement is being studied intensively in other research programs such as European and North American acid rain research programs. A rigorous treatment requires a knowledge of C_a and of V_d for each particle size. However, in practice, a knowledge of the size distribution of particulate matter in the Arctic can be used to obtain an average V_d to estimate dry deposition. For the Arctic, most anthropogenic aerosols that reach the region are smaller than 1 µm in diameter (Barrie 1986). In this case for snow, a V_d of 0.05 to 0.1 $cm{\cdot}s^{-1}$ has been determined in field studies for sub-micrometer particles typical of those carrying most contaminants (Ibrahim *et al.* 1983, Davidson 1989). This value was applied to estimate contaminant deposition (Jantunen and Bidleman 1995, McVeety and Hites 1988). An illustration of the relative importance of the scavenging of OCs by snowfall and by particle dry deposition of Arctic haze aerosols is given in Figure 4.2b. Results are based on the sorption studies of Hoff *et al.* (1995). It can be seen that for all OCs examined, snow scavenging of particles and gases at -30°C is always at least 10 times as important as particle dry deposition.

Another atmosphere-surface exchange pathway is fog water deposition (Barrie and Schemenauer 1986, 1989), which is a hybrid of precipitation scavenging and dry deposition processes. Fogs are essentially clouds at the ground. In some cases this is literally true, for example, when a layer of cloud passes over a mountain. In other cases, the fog is formed by processes (different from those of cloud formation) that take place near the Earth's surface. If air is cooled sufficiently, the water vapour in it will condense into liquid drop. The cooling can be produced by several different meteorological situations leading to different types of fogs. At night, the Earth's surface loses heat through the transmission of terrestrial infrared radiation to the atmosphere above. Sufficient surface cooling leads to saturation of the near-surface layer and "radiation fog." Light winds (<10 km/hr) promote fog development by mixing cool, moist surface air to moderate heights. At higher wind speeds, mixing with the drier air aloft tends to inhibit fog formation.

"Advection fog" is produced when warm moist air is transported over a colder surface. Most coastal fogs are of this type. "Upslope fog" is produced when air is forced to rise over a topographical barrier decreasing atmospheric pressure that produces expansion and cooling of the air. Other types of fogs are less important vehicles of deposition. "Steam fog" or "arctic sea smoke" is produced by the condensation of water vapour from a relatively warm surface in much colder air aloft. This type of fog is observed when Arctic Ocean ice cracks in winter, forming open leads, and the atmospheric temperature is much below 0°C. Ice fog is produced at temperatures below about -30°C when water vapour sublimes directly onto atmospheric ice nuclei (particles) thus forming tiny ice crystals. Fogs remain liquid (containing supercooled water drops) down to temperatures as low as -20°C, and occasionally to much lower temperatures, a factor that that is important in the scavenging of contaminants from the atmosphere.

The means by which particles and gases are incorporated into fog droplets of 1 to 20 micrometers in diameter are the same as for cloud droplets. They are then deposited to the Earth by processes similar to that of dry deposition of particles: sedimentation for droplets larger than 5 micrometers, and impaction on surface roughness elements for smaller particles. The microstructure of fogs is strongly influenced by the characteristics of atmospheric cloud condensation nuclei (i.e. hygroscopic aerosol particles dominated by sulphates and sea salt). Polluted air masses with high aerosol concentrations generally produce fogs with high fog droplet number concentrations and smaller drop sizes. Sea fogs generally have lower total droplet concentrations and larger droplets. Knowledge of the chemical composition of fogwater as a function of droplet diameter is important in calculating the deposition of contaminants in fogs. Unfortunately, reliable size-dependent chemical composition data are not available for fogs in the polar regions.

2.4.1.3 Particle Resuspension (Figure 4.1, Steps 3 and 8)

It is well known that particulate matter can be suspended to great heights in the atmosphere and then transported long distances. For instance, dust from the Gobi desert, Mongolia has been detected in Hawaii in the central Pacific, and occasionally in the Canadian Arctic (Barrie 1994, 1996, Welch *et al.* 1991). Similarly, in high wind conditions in the Arctic, considerable quantities of snow, together with particulate matter, can be suspended in the atmosphere. This occurs when surface wind speeds exceed 3 $m{\cdot}s^{-1}$. Atmospheric aerosol aluminum is a good indicator of airborne soil dust. Air concentrations of soil aluminum measured at Alert during a blizzard in April 1986 were five to seven times higher than those during low wind conditions (Barrie *et al.* 1989). Thirteen years of routine observations at Alert show much higher concentrations of suspended soil during windy weeks than during low-wind weeks (Figure 4.3). However, little is known quantitatively about the resuspension of snow-bound contaminants during blowing snow conditions.

The erosion of surface snow occurs when the shear force at the air/snow interface exceeds inter-particle cohesive forces and particle inertia. The two primary modes of snow transport are saltation and suspension. Saltation is a near-surface phenomenon that occurs at low wind speeds, while suspension can disperse snow to heights of tens of metres. Suspension is the dominant process in high winds. Both are important phenomena in the Arctic, especially in the tundra and barren lands. Blowing snow grains become fragmented during collision with other snow particles or surface snow resulting in a more amorphous particle shape (Pomeroy *et al.* 1991, Pomeroy and Gray 1995) and an associated decrease in surface area. The effect of surface area changes during blowing snow on the associated contaminant content is not well understood. Pomeroy and Gray (1995) have shown that wind velocity, temperature and relative humidity are important factors in sublimation of airborne ice crystals. Sublimation results in increased concentrations of involatile major ions and decreased concentrations of volatile OCs that are released together with water vapour.

2.4.1.4 Gas Exchange Between Atmosphere and Snowpack/Ice (Figure 4.1, Step 4)

Gas exchange is a two-way exchange process between the atmosphere and the surface. The partial vapor pressure gradient of an OC between ambient air and interstitial air within the snowpack determines the potential for exchange. This is strongly dependent on temperature. In general, compounds with relatively high vapor pressures, such as HCB and α- and γ-HCH, can volatilize substantially from the snowpack while PAHs and many of the PCB congeners with lower vapor pressures may not. Supporting evidence for volatilization from snow comes from past and present studies in the Arctic. For instance, on the Canadian Ice Island HCB and chlordane were detected in snow collected in May and June, 1986 (33 and 91 $pg \cdot L^{-1}$, respectively) but not in snow collected from the same area in August and September (<1 $pg \cdot L^{-1}$).

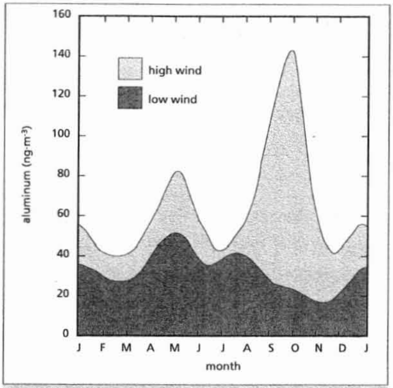


FIGURE 4.3

Seasonal variation of airborne windblown dust at Alert indicated by concentrations of aluminum during weeks of low and high wind (Sirois and Barrie 1997).

Several observational studies in the NCP have allowed us to examine the process of post-depositional gas exchange between the snowpack and atmosphere. These include: (i) Agassiz Glacier studies (Gregor 1991, and new results reported here), (ii) Amituk Lake basin studies (Semkin *et al.* 1996) and (iii) snowpack-snowfall chemistry surveys. On the Agassiz Ice Cap, Ellesmere Island, Canada (80° 40′N 73° 30′W), replicate samples were taken from annual snow layers in late winter of the year of deposition (approximately May), and the same deposit in one or more subsequent years. The original (year of deposition) and residual (following years) OC concentrations were determined for these samples (Table 4.3). As noted by Gregor (1991) average summer melting on the ice cap is limited to about 3% of the past winter snow accumulation. The data confirms the original conclusions from this sampling location (Gregor 1991). The greatest decrease in OC concentrations in the fresh snow layers was observed for α- and γ-HCH (>65%), endosulfan-I (>90%), and dieldrin (>80%).

At Amituk Lake (Figure 3.21, Section 2.3.4), there was a loss of most OC contaminants from the shallow snowpack between May 13 and pre-melt June 13. In particular, the di- and tri-chlorobenzenes, heptachlor epoxide, α-chlordane, ΣPCB and ΣDDT showed marked decreases in concentration possibly as a result of volatilization losses. The exception was the HCH isomers, which were constant. This latter finding is not consistent with the Agassiz glacier observations or the theory of gas volatilization.

Data from the snowpack chemistry survey (Section 2.3.2) undertaken in March–May 1992 yielded the following estimates (Table 4.4) of snowpack burden (B) defined as follows:

$$B = C\rho_{snow}\, d_{snow} = C\ SWE \qquad (4.2)$$

where C is the contaminant concentration, ρ_{snow} is snowpack density, d_{snow} is the snowpack depth, and the product of ρ_{snow} and d_{snow}, SWE, is snow water

TABLE 4.3

Original and residual concentrations (pg·L⁻¹) of pesticides in annual snowpack layers from the Agassiz Ice Cap[1]

Year of Snowfall: Sampling Year: Organochlorine Pesticide	1985/86 1986 Mean (SD) (n=5)	1985/86 1987 Mean (SD) (n=3)	Percent Residue[2] in 1987 (%)	1988/89 1989 Mean (SD) (n=3)	1988/89 1990 Mean (SD) (n=3)	1988/89 1992 Mean (SD) (n=2)	Percent Residue[2] in 1990 (%)	Percent Residue[2] in 1992 (%)	1989/90 1990 (n=1)	1989/90 1992 Mean (SD) (n=2)	Percent Residue[2] in 1992 (%)
Hexachlorobenzene	10 (1)	10 (1)	100	51 (15)	128 (14)	62 (13)	100+	100+	174	54 (22)	31
Heptachlor	—	—	—	—	240 (279)	21 (6)	—	—	137	91 (3)	66
α-HCH	6580 (1440)	497 (90)	8	437 (73)	207 (51)	115 (5)	47	26	856	109 (3)	13
γ-HCH	4080 (909)	128 (43)	3	435 (113)	70 (3)	143 (28)	16	33	561	136 (17)	24
Heptachlor epoxide	358 (55)	47 (15)	13	56 (49)	68 (11)	—	100+	0	151	0	0
γ-Chlordane	394 (51)	17 (15)	4	—	—	—	—	—	158	0	0
α-Endosulfan	1094 (156)	ND	0	298 (124)	—	35 (23)	—	12		46 (1)	—
α-Chlordane	366 (53)	23 (15)	6	—	—	—	—	—	146	29 (41)	20
Dieldrin	1350 (240)	213 (75)	16	530 (161)	98 (4)	99 (71)	18	19	286	180 (35)	63
o,p'-DDT	—	—	—	—	—	—	—	—	159	59 (17)	38
p,p'-DDT	—	—	—	—	—	—	—	—	490	160 (24)	33
o,p'-DDD	—	—	—	—	—	—	—	—	227	144 (204)	63
p,p'-DDD	—	—	—	—	—	—	—	—	128	93 (10)	72
o,p'-DDE	—	—	—	—	—	—	—	—	151	ND	0
p,p'-DDE	—	—	—	—	—	—	—	—	284	24 (5)	9
p,p'-Methoxychlor	—	—	—	—	—	—	—	—	697	150 (212)	21

[1] *Franz* et al. *1997*

[2] *Residue is the amount (expressed) as a percent of pesticides measured in original snowpack layer remaining when resampled in subsequent year(s) as noted.*

equivalent. B is in units of mass per unit area. Comparison of regional mean seasonal deposition (ΣF_i) (Table 4.1) with regional mean snowpack burden (B) (Table 4.4), provides little insight into post-depositional loss of contaminants from the snowpack due to the high variability of B in space and time which, as indicated in equation 4.2, is determined by variance of snowpack water equivalent (SWE) and, to a lesser extent, of contaminant concentration (C).

It is more appropriate to compare ΣF_i and B for individual sites. There were a limited number of sites at which both these parameters were measured simultaneously. The ratio of $\Sigma F_i/B$ for ΣHCH and ΣPCB is shown in Figure 4.4. Concentrations for other OCs were too close to detection limits to allow a meaningful comparison to be made. In general, ΣHCH is more abundant in fresh snowfall than in the snowpack (ratio range 1 to 4.5), while ΣPCB was not significantly different (ratio range 0.3 to 2.2). Thus, as might be expected, the more volatile HCHs showed evidence of post-depositional loss while PCBs did not. These results are consistent with those for Agassiz Glacier (Table 4.3).

To compare the above observations to the theory of gas to snow partitioning of semi-volatile organic compounds of Hoff *et al.* (1995), we calculated the ratio of the snowpack burden (B, equation 4. 2) for an OC that is in equilibrium, assuming a constant concentration of total OC (gas + particle) at -30°C to that at 0°C (Figure 4.5). The atmospheric concentration of many OCs is relatively constant (variations less than 50%) in the month of May when temperatures in the Arctic rise rapidly toward 0°C (for instance, observations from week 16 to 21 in Figure. 3.1 and 3.2 for Alert, Tagish and Dunay, as well as at Spitzbergen in the European Arctic (Oehme *et al.* 1996). For the most volatile constituents (α-HCH, HCB and γ-HCH), the theoretically predicted ratio of initial to final snowpack burden (Figure 4.5, bars) is greater than 5. In other words less than 20% of the initial burden remains after the snowpack warms as a result of volatilization to the atmosphere. In contrast, the less-volatile more particle-bound constituents (dieldrin, chlordane, toxaphene and DDT) are predicted to change little in concentration. The reason for this is that as temperature rises, the shift in OC mass from particle to gas phase exceeds the decrease in sorption of the gas by the snowpack. The results in Figure 4.5 are theoretical predictions that assume atmospheric concentrations are constant. They serve to illustrate the complexity of pathways 4 and 5 in Figure 4.1.

Observations of snow pack losses by Semkin *et al.* (1996) at Amituk Lake (see Section 2.3.4) and from the Agassiz glacier (Table 4.3) are also included in Figure 4.5 as bars with arrows and square, respectively. Reality differs considerably from the simple model discussed above. However, conditions in the studies at Amituk Lake are closer to those assumed in the model than those at Agassiz. Losses from the Agassiz Glacier occurred over a period of a year from one May to the next May as the surface snow pack warmed, drifted and was buried by more snowfall. Atmospheric concentrations may not have remained constant over this period. They probably dropped from beginning to end. This would tend to increase losses even more than predicted.

The re-volatilization of OCs to the atmosphere from the snowpack may have an impact on atmospheric OC concentrations. The concentration of many OCs in air in the high Arctic at Alert peak in the spring. For instance at Alert between February, April and June, the average total HCH concentration increases from 85 to 112 to 120 pg·m^{-3}, respectively (Figure 3.2). The change between February and springmelt in June is likely caused in part by increasing import of HCH from mid-latitudes as usage and emissions to the atmosphere increase.

Let us examine whether there is sufficient HCH in the arctic snowpack to have an impact on atmospheric concentrations between April and June as the snowpack warms up. From Table (3.3), the average snowpack HCH concentration in late winter of 1991 in the Canadian Arctic was 0.76 ng·L^{-1}. Assuming that the snowpack is 30cm deep with a density of 0.3 g·cm^{-3} (typical of snowpack on the Arctic Ocean in April), the corresponding average snowpack burden of HCH is 68 ng·m^{-2}. Assuming an atmospheric concentration of total HCH of 110 pg·m^{-3} typical of early May, the total amount of HCH in an atmospheric column of air 1 km deep is 110 ng·m^{-2} compared with 68 ng·m^{-2} in the snowpack. Thus, there are comparable burdens of HCH in the late winter arctic snowpack and in the atmosphere. This means that increases in atmospheric HCH concentrations due to volatilization from the snowpack during spring warming are indeed possible. Since more than 99% of each of the HCH isomers leave the snowpack during spring warming, there is no fractionation of isomers expected in this process. Thus, observed peaks during May of the γ/α ratios of atmospheric HCH (Figure 3.2) are likely from import of HCH from sources outside the Arctic.

2.4.1.5 Gas-Exchange Between Atmosphere and Soil, Water and Vegetation (Figure 4.1 Step 5)

This process involves the reversible exchange of gases to water (fresh or salt), vegetation or soil. Most is known about the exchange to water, which is usually described by the "two-film" model (Liss and Slater 1974). In this process, the rate of exchange is assumed to be limited by molecular diffusion across thin air and water films at the interface. The net gas flux can be estimated by:

$$F = K_l[C_w - C_aRT/H] \tag{4.3}$$

$$1/K_l = 1/k_l + RT/Hk_g \tag{4.4}$$

In these equations C_w and C_a are the concentrations of chemicals in water (dissolved) and air (gaseous), k_l and k_g are the mass transfer coefficients for the individual water and air films, H is the Henry's Law constant at the water temperature and T is the air temperature (K). K_l is the overall mass transfer coefficient that takes into account the resistance to transfer offered by both the air and water films. These relationships have been applied to the exchange of OCs by others (Bidleman and McConnell 1995, and Cotham and Bidleman 1991) as well as in this review

TABLE 4.4

Snowpack burden (B) of OCs (mg·m^{-2}·season^{-1}) from 1992 snow survey. For site locations see Figure 3.11. For a definition of OC group constituents see Table 3.3. Bold values are anomolous.

	Date	Depth (cm)	Density (g·cm^{-3})	Snowpack Burden at end of Season ΣHCH	ΣChlord	ΣDDT	HCB	ΣPCBs
Great Bear Lake	Apr. 24	27	0.32	0.01	0.01	0.01	0.02	0.32
Nahanni Butte	Mar. 17	51	0.30	0.06	—	0.01	0.04	0.32
Slave River	Mar. 20	63	0.097	0.01	—	<0.01	<0.01	0.11
Kakisa River	Mar. 17	27	0.23	0.01	—	<0.01	0.02	0.26
Reid Lake	Apr. 4	54	0.24	0.03	<0.01	<0.01	0.02	0.44
Akasta River	Apr. 25	46	0.31	0.03	—	<0.01	0.02	0.27
Back River	Apr. 30	38	0.43	<0.01	<0.01	0.01	0.03	0.14
Baker/VOR Lake	Apr. 30	28	0.49	0.12	<0.01	0.01	0.02	0.45
Yathkyed Lake	May 6	26	0.51	0.03	<0.01	<0.01	0.05	0.22
Thelon River	Apr. 30	36	0.37	0.07	—	0.01	0.03	0.43
Brown River	Apr. 29	37	0.36	0.11	<0.01	0.01	0.03	0.41
Lorrilard River	May 2	34	0.27	0.02	—	<0.01	0.02	0.30
Seal Hole Lake	Apr. 10	38	0.45	0.12	0.02	0.01	0.02	0.37
Ellice River	Apr. 21	22	0.42	0.04	—	0.01	0.03	0.43
Hayes River	Apr. 29	83	0.43	1.30*	0.03	0.05	0.06	**1.27**[2]
Stanley Fletch. Lake	Apr. 2	50	0.24	0.23	0.02	0.05	0.01	0.81
Chartrand Lake	May 6	26	0.24	0.13	0.01	0.03	—	0.56
Lady Melville Lake	Apr. 2	27	0.27	0.06	<0.01	0.02	<0.01	0.94
Penny Ice Cap	May 3	106	0.23	**0.55**[1]	—	0.03	0.01	**2.24**[1]
ARITHMETIC MEAN	—	—	—	0.16	—	0.017	0.025	0.54

[1] *High due to above average snowfall at Penny Ice Cap. Concentrations were average.*

[2] *Hayes River site was subject to locally generated wind blown sediment, which is incorporated into the snowpack.*

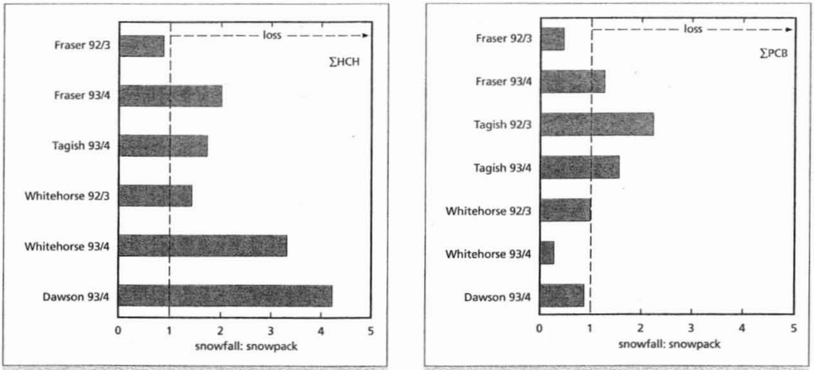


FIGURE 4.4A

FIGURE 4.4B

The ratio of precipitation-amount-weighted-mean OC concentration in snowfall for the snow accumulation period, to the snowpack concentration at the end of the snow season for selected sites. A ratio greater than one indicates post-depositional loss of OC from the snowpack. For site locations see Figure 3.11.

to calculate Arctic Ocean HCH budgets (Figure 4.8, Section 2.6.1). It is important to recognize that gas exchange takes place even under equilibrium conditions when invasion and evasion are balanced and the net flux is zero. Thus the mass of material transferred by gas exchange is much larger than is apparent from the net flux. For this reason, Murphy (1995) suggested that gas exchange be calculated as two separate terms, one for volatilization and the other for deposition.

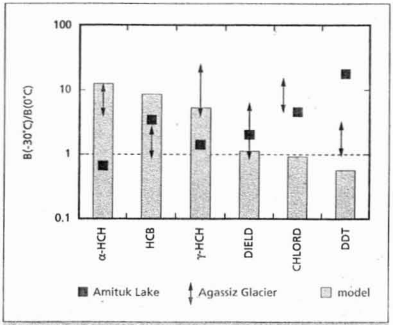


FIGURE 4.5

The ratio of initial to final snowpack burden (B) of an OC in snowpack as temperature rises from −30°C to 0°C calculated using the theory of Hoff et al. (1995) for gas sorption to snow and particles (depicted as bars). Atmospheric gas-particle partitioning is included in the calculation (for details see Hoff et al. 1995). Snowpack depth assumed was 30 cm at a density of 0.3 g·cm^{-3}. Atmospheric parameters for aerosols were those listed in the caption for Figure 4.2. Observations of snowpack losses by Semkin et al. (1996) at Amituk Lake (black squares, see section 2.3.4) and from the Agassiz glacier (Table 4.3) (range depicted as double-headed arrows) are also included.

Gas exchange to soils depends on the sorption of contaminants in the gas phase to soil surfaces. In forested areas (leaf: ground area ratios of 3–19), leaves or needles may represent the primary terrestrial contact for air-borne chemicals (Schulze 1982) and thus for gas-phase transfers. Uptake and loss of OCs to arctic plants such as lichens and frozen soils are poorly understood necessitating further research if the air-plant-terrestrial food chain pathway is to be better quantified.

2.4.1.6 Snowmelt Runoff (Figure 4.1, Step 9)

A key question in the inputs of contaminants to eco-systems during winter is: "How much of a contaminant present in the snowpack just before spring melt actually reaches the water as runoff?" In the previous two sections, evidence of post-depositional loss of OCs from snowpacks was given. The process is significant in affecting the snowpack burden immediately prior to melt. The Amituk Lake study in the high Arctic (Section 2.3.4) has shown that most OCs left in the snowpack pass through the lake in meltwater streams, interacting very little with the lake system.

2.4.1.7 Spatial Variations in the Snowpack Burden

The inventory of contaminants in the snowpack before the onset of snow melt can be mapped, as illustrated for HCHs and PCBs in Figure 4.6 and 4.7, respective-

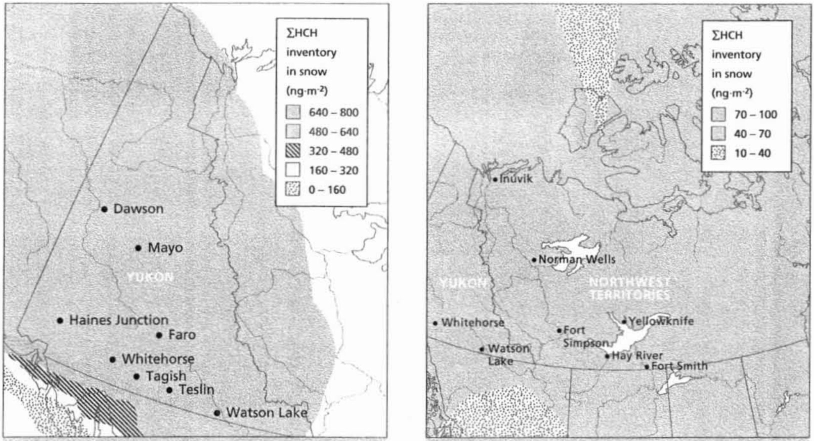


FIGURE 4.6A **FIGURE 4.6B**

The estimated spatial distribution of snowpack burden (B) of ΣHCH in the Yukon/NWT.

ly (Gregor and Williams 1996). The figures are based on available snow chemistry data, and estimates of snow water equivalents (SWE) and are mapped using a combination of satellite imagery and ground observations. Although a number of compounds have been mapped in this way (see Gregor and Williams 1996), ΣHCH and ΣPCB have been selected to illustrate the spatial pattern representing volatile and particle-associated compounds, respectively. (The map is based on snowpack chemistry data from 1988 to 1993 for the NWT, and from 1992 to 1993 only for the Yukon). The variance in B is mostly due to variance in SWE. The range in concentration between sites for the compounds considered was relatively small (coefficient of variance 120% for HCH and 71% for ΣPCB).

For the NWT, SWE data from satellite snow surveys were used (B. Goodison, Atmospheric Environment Service and PhD Associates). A total of 3114 observations were interpolated to a 25 km grid covering the territory. The algorithms run to determine SWE values accounted for forest cover in different regions. The final SWE values produced from the satellite surveys were mean values and incorporated values from 1979 to 1993. In addition, 53 NWT sites from snow surveys undertaken by the Department of Indian Affairs and Northern Development (DIAND)–Yellowknife region were also incorporated into the SWE data base as mean SWE for the period of record (variable by station). In comparison, the snowpack chemistry data are relatively limited in number (a maximum of 86 samples collected between late March and early May in 1989, 1990, 1992 and 1993) although widely distributed across the NWT.

In the Yukon, satellite imagery is not reliable due to mountainous terrain. Instead, a total of 60 snow survey ground observations were used (including two BC sites and three Alaska sites). The SWE data for the Yukon were multi-year averages for the entire period of record for each site (DIAND 1995). Two springtime snowpack SHCH concentrations were used for the Yukon (0.24 ng·L^{-1} for areas with SWEs less than 321 mm and 1.35 ng·L^{-1} for areas with SWE greater than 321 mm (Gregor and Williams 1996). This was required because of the evidence for higher concentrations of the volatile compounds in deeper snowpacks from the Yukon (Palmer 1996, Semkin 1996). In the NWT, the average of all sites (0.886 ng·L^{-1}) was used.

ΣPCB concentrations used were 3.7 ng·L^{-1} in the Yukon (the mean of bulk samples excluding Whitehorse) and 3.6 ng·L^{-1} in the NWT (based on data from 1991/92, 28 samples). Due to the reduced volatility of PCBs, there was no evidence for PCB concentration differences with snowpack depth in the Yukon. There is very good agreement for the concentrations used for PCBs in the two territories. The snow chemistry data suggest that extensive snowpack chemistry sampling is not required for estimating contaminant inventories since the major source of variance is the SWE. In future studies, it may be preferable to have fewer sites and more within-site sampling in order to arrive at a more accurate estimate of a regionally representative snowpack contaminant burden and to

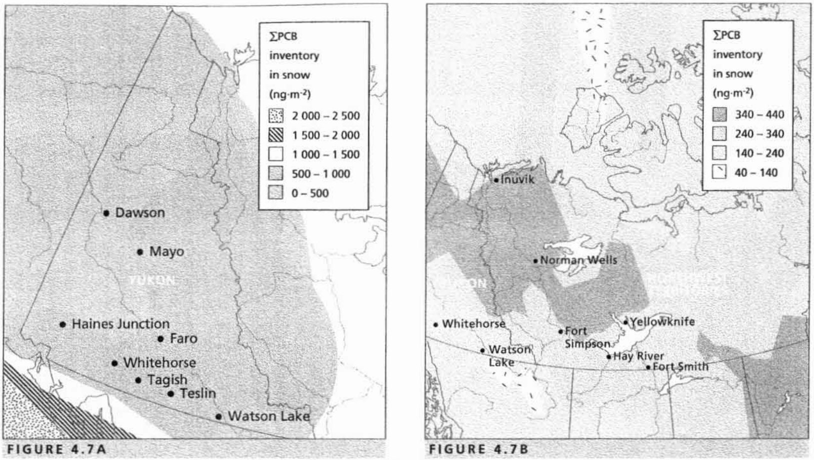


FIGURE 4.7A

FIGURE 4.7B

The estimated spatial distribution of snowpack burden of ΣPCB in the Yukon/NWT.

provide a better understanding of the inventory of contaminants potentially delivered to the aquatic system. Greater emphasis should be put on analytical sensitivity and spatial chemical variability as well as in improving the SWE estimates.

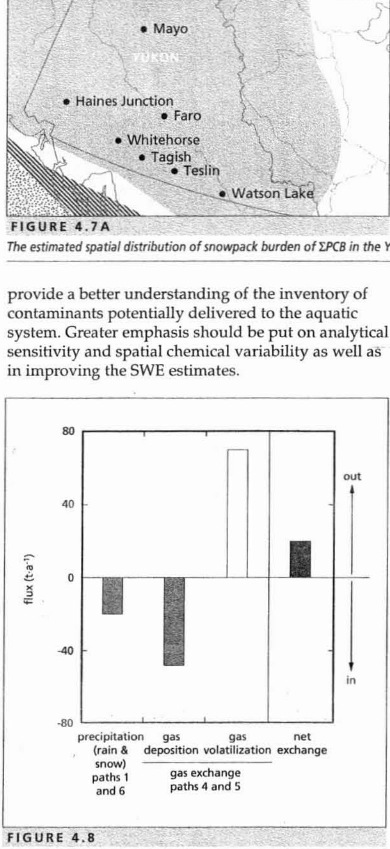


FIGURE 4.8

A comparison of the estimated annual inputs and outputs of ΣHCH between the atmosphere and Arctic Ocean (see section 2.6.1). Dry deposition was negligibly small. Fog water deposition was neglected. Refer to Figure 4.1 for a description of potential pathways.

2.4.1.8 A Synthesis of Atmosphere Ocean Exchange for HCH

Using techniques discussed above and in some detail in Section 2.6.2, the relative magnitude of atmosphere-ocean exchange pathways (Figure 4.1) can be estimated for HCH. Since HCH is a relatively volatile constituent, particulate matter plays little role in the process. In Figure 4.8, it can be seen that the main controlling factor of atmosphere-Arctic Ocean exchange is gas exchange, which is larger than inputs by precipitation and particle dry deposition. Most of the deposition in precipitation is due to sorption of HCH isomers by snow, which was assumed to be a one-way process.

- *Atmosphere-surface exchange of contaminants involves many processes including scavenging by rain or snow, deposition in fog and particles, gas exchange and particle resuspension. Generally, snow is a much more efficient scavenger of OCs than rain.*
- *There is considerable observational evidence confirmed by theory showing that many volatile OCs deposited in snow in the cold winter return to the atmosphere from the snowpack as it warms in spring and is ventilated by wind. ΣHCHs tend to leave more readily than ΣPCBs. Less loss occurs from deep snowpacks than from shallow ones.*
- *Fogwater deposition of contaminants is very probable in summer over the Arctic Ocean but no information exists to quantify this pathway.*

- *For the Arctic Ocean, atmosphere-surface exchange of ΣHCH is dominated by gas exchange between air and open water, snow scavenging and possibly volatilization loss from snow pack to atmosphere. This latter step needs to be studied more. Atmosphere-surface exchange of toxaphene is dominated by gas exchange, precipitation scavenging and, in contrast to HCH, dry deposition because these compounds associate more with particles.*

2.4.2 River-Ocean

The amount of contaminants transferred in a year to the Arctic Ocean by rivers ($F_{r/o}$) is estimated from the following integral:

$$F_{r/o} = \int (C_w + C_{ss} S) V \, dt \qquad (4.3)$$

where C_w is the concentration of a substance dissolved in river water, C_{ss} is the concentration of the substance per unit mass of suspended sediment, S is the mass concentration in the river of suspended sediment and V is the volume flow rate of river water into the ocean. V and S for the Mackenzie River are shown in Figures 2.5a and b, respectively. All the terms in equation 4.3 are highly variable and in practice few measurements of the chemical concentrations are made during the year, especially during the spring melt period. Nevertheless, estimates of $F_{r/o}$ have been made (Table 4.5) using data on mean flows and OC concentrations for 1993 (Table 3.8). It is recognized that the chemical data were derived from samples taken during the high flow period and that these concentrations are probably not constant throughout the year. The proportion and amounts that will be transported via dissolved and adsorbed suspended matter will also change.

Concentrations of the OCs measured were variable and often undetectable (Table 3.8). Suspended solid burden (S) in the non-Mackenzie rivers was very low. For the Mackenzie River, the percent of ΣPCB on the suspended fraction was 13%–24% during the 1993 summer sampling period. Of the smaller Canadian rivers, only the Coppermine and Hayes rivers had significant suspended material. The former had 31% of the PCB burden in the suspended fraction while the Hayes River had 69%. PAHs in the Mackenzie River samples ranged from 25% to 94% in the suspended fraction (napthalene 25%–35%, phenanthrene 57%–68%, 2-methyl- naphthalene 45%–46%) while those in the Coppermine and Hayes rivers were much lower (less than 10% for the significant PAHs mentioned). The differences in phase distribution are attributed to a number of factors, including the low concentrations of OCs in all rivers (mainly <1 ng·L^{-1}) except for the major PAH compounds (>2–3 ng·L^{-1}), the higher suspended solid burden of the Mackenzie River (60–100 mg·L^{-1} vs. 26 and 9 mg·L^{-1} for the Coppermine and Hayes rivers, respectively) and higher particulate organic carbon concentrations for the Mackenzie River suspended matter (approx. 2.0 vs. 0.4 mg·L^{-1}).

One particular purpose in making the estimates shown in Table 4.5 was to determine whether the sum of contaminant inputs via rivers other than the Mackenzie River is a significant fraction of the total Canadian riverine input to the Arctic Ocean. The annual flows and drainage areas of the sampled non-Mackenzie Arctic rivers represent roughly 15% and 33%, respectively, of the flow and drainage area of the Mackenzie River. For ΣDDT and ΣPCB, the six rivers collectively have loadings relative to the Mackenzie River in proportion to their flows. In contrast, the concentrations of ΣHCHs and Σchlordanes, are 4 to 10 times more than expected on the basis of flows. However, when this comparison is made on the basis of relative drainage areas rather than flows, these other rivers are under-delivering ΣDDT and ΣPCB by 50% and over-delivering ΣHCH and Σchlordane by 50% and 400%, respectively. In contrast, results for Pb (Table 4.5) indicate that on the basis of flow and area the group of smaller arctic rivers under-delivers by a factor of two and five, respectively.

Inputs of OCs to Hudson Bay by northern rivers are also included in Table 4.5. They are comparable to the sum of inputs to the Arctic Ocean of the Arctic rivers sampled, excluding the Mackenzie River.

The particulate flux of PAHs in the Mackenzie River exceeds the dissolved fraction by two orders of magnitude and has a strong seasonal flux; winter contributes less than 0.6% of the annual total. The PAH data available is summarized by Yunker and Macdonald (1995). The authors examined the suspended solids and dissolved concentrations and fluxes of the alkanes and PAHs in the Mackenzie River at several times during the seasons of 1987. Compared to assessments elsewhere (Hites *et al.* 1980), the Mackenzie River is considered to be an area with a high natural baseline of PAHs. Phenanthrene, anthracene and naphthalene, including the methylated naphthalenes were observed to dominate the PAH burden and an assessment of the PAH compounds and their relationship to known tracers of combustion and plant origin, led the authors to conclude that there was little contribution from anthropogenic or long-range transport sources. Using a PAH mean of approximately 1000 ng·g^{-1} and an annual flux of suspended solids of 2.2 x 10^{11} kg·a^{-1} (Yunker and Macdonald 1995), the total PAH flux to the Arctic Ocean (Beaufort Sea) is 220 t·a^{-1}. This is a tremendous mass when compared with the fluxes observed for the OCs. The environmental significance is unknown. Ecosystems have probably been historically exposed to elevated levels of PAHs and it is likely that biota have evolved detoxification mechanisms. It may be, however, that a threshold exposure level has been reached. Therefore, any additional contaminants inputs to the area as a result of petroleum exploration and development is of concern.

Canadian river inputs of OCs to the Arctic Ocean can be compared with those from Russian rivers. Estimates were made of the riverine transfer of OCs to the Arctic Ocean from Russian rivers using the OC concentrations from ROSHYDROMET (1995) (see Section 2.3.4). When combined with the annual flows from Table 2.2, inputs of ΣHCH for Russian rivers were estimated at 32, 23 and 15 $t \cdot a^{-1}$ for 1992, 1993 and 1994, respectively. The net input of Canadian rivers of total HCH is 0.15 $t \cdot a^{-1}$. Thus, if the reported HCH concentrations in Russian rivers are accurate they are much greater sources of HCHs to the Arctic than North American rivers. More measurements and inter-laboratory comparison of measurement and analysis methodologies are required.

2.4.2.1 Delta Processes

Because deltas, estuaries and large shelves tend to trap most of the sediments brought in by rivers (GESAMP 1987, 1993), particle-reactive contaminants entering in this manner will tend also to be initially trapped in these places. However, dissolved (conservative) contaminants will be free to travel further, undergoing dilution as they are entrained into sea water. Many rivers entering the Arctic Ocean form deltas on the shelf proper, but two of the largest, the Yenisey and the Ob, discharge via long estuaries (Pfirman *et al.* 1995b). This has a large impact on the trace metal pathways (Dai and Martin 1995). It is to be expected that the delivery efficiency of particles and particle-associated contaminants to the ocean, as well as their trapping in coastal sediments depends on exactly how a river enters the sea. Sediment loads that are discharged by rivers into shallow coastal waters (<20 m) may settle initially but will be vulnerable to further transport by resuspension, direct entrainment of turbid river water into ice and the entrainment of bottom sediments into ice either by direct bottom contact or by suspension freezing (see Section 2.3.3).

In the Canadian Arctic, the Mackenzie River forms the largest delta, and probably has the best data base from which to estimate contaminant fluxes and budgets (Macdonald et al. 1997). This river discharges onto a wide, shallow shelf and therefore, ultimately, the deposition of sediments will be considerably affected by storms and ice-related processes. Macdonald *et al.* (1997) estimate that about half of the sediment load coming down the Mackenzie River (127×10^6 $t \cdot a^{-1}$) is trapped within the delta. However, this estimate is subject to considerable uncertainty. In addition to the difficulty of determining the rate of accumulation within the delta directly, or the error in calculating it by difference, there is considerable reworking of deposits in the lower river. New sediments may settle, only to be replaced by the entrainment of existing sediment within the delta. Storms frequently produce wholesale resuspension in the nearshore (<10 m) transporting sediments out onto the shelf (Hill and Nadeau 1989). Other rivers in the Canadian Arctic are far smaller than the Mackenzie River in their discharge and in their sediment load. For the Mackenzie River, the present estimate is that about half of the particle-associated contaminants coming down the river stop in the delta and most of the remainder becomes trapped on the shelf with only a small amount (10% to 15%) exiting the shelf (Macdonald *et al.* 1997).

- *Inputs of OCs to the Arctic marine system by Canadian rivers is not always dominated by the Mackenzie River even though it carries 75% of the Canadian river water that flows annually to the Arctic seas. Small rivers flowing to the Archipelago carry a higher fraction of total inputs of ΣHCH and ΣChlordane to the Arctic Ocean than they do for water. However, fractional*

TABLE 4.5

Estimates of the annual transfer of OCs and Pb to the Arctic Ocean ($F_{r/o}$) (kg·a⁻¹) by Canadian Arctic rivers.

	V ($m^3 \cdot s^{-1}$)	ΣHCH (kg)	ΣDDT	ΣPCB	ΣChlor	Pb
Mackenzie at Arctic Red	9094	90.7	29.6	185	2.4	509
Mackenzie East	133	2.4	1.7	33.9	0.6	—
Mackenzie West	982	10.7	0.1	150	2.3	20
Anderson below Carnwath	146	—	—	—	—	4.6
Coppermine above Copper C.	266	6.7	0.9	8.6	—	—
Burnside at mouth	125	4.6	0.2	2.8	0.3	—
Ellice at mouth	83	4.5	0.1	2.1	0.2	2.6
Back above Hermann R.	494	24	1.9	9.7	2.5	16
Hayes above Chantrey In.	129	6.7	0.4	2.2	0.8	5.3
Total Non-Mackenzie to Arctic Ocean	**1244**	**46.5**	**3.6**	**25.3**	**3.8**	**28**
Fraction of Mackenzie at Arctic Red	**14%**	**51%**	**12%**	**14%**	**156%**	**6%**
Thelon below Shultz Lake	995	28.3	1.2	22.3	2.6	—
Thelon above Beverly Lake	253					—
Dubawnt at Marjorie LakeOut.	352	12.7	0.3	13.3	0.7	—
Kazan above Kazan Falls	434	19.3	0.3	10	0.5	—
Quoich above St.Clair Falls	176	4.3	0.2	4.3	0.4	—
Thlewiaza above Sealhole Lake	217	—	—	—	—	—
Total to Hudson Bay	**1605**	**51.9**	**1.7**	**36.6**	**3.6**	**—**

inputs of ΣPCBs and ΣDDT are similar to the fraction of water flow.

- *The relative abundance of OC inputs by the Mackenzie River to the Arctic Ocean is: ΣPCB > ΣHCH > ΣDDT > ΣChlordane.*
- *If reported HCH concentrations in Russian rivers are accurate, Russian rivers are much greater sources of HCHs to the Arctic than North American rivers. More measurements and inter-laboratory comparisons of measurement and analysis methodologies are required between Canadian and Russian laboratories.*
- *River-ocean exchange is strongly influenced by loss to sediments as water flows from deltas to estuaries to continental shelves. For the Mackenzie River, about half of the particle-associated contaminants coming down the river stop in the delta and most of the remainder becomes trapped on the shelf with only a small amount (10 to 15%) exiting the shelf. For OCs that are least particle-associated in water (e.g. HCH) , most of the contaminant burden is in the dissolved phase and escapes the sediment sink. However, for substances such as PCB and DDT, considerable total loss can occur.*

2.4.3 Ocean-Ocean

The Arctic Ocean should not be considered as a single oceanic entity (Aagaard 1989). Regional differences within the Arctic Ocean are evident both between the two major basins in the interior ocean (i.e., the Canadian Basin and the Eurasian Basin) and between the shelves and their respective basins. The oceanographic differences between the Eurasian and Canadian basins are due partly to large-scale circulation patterns (Figures 3.15 and 3.16) and partly to differences in water sources as shown by the distribution of Pacific and Atlantic origin water (Figures 3.14 and 3.16). Generally, the residence times of the surface water and ice are longer in the Canadian Basin than in the Eurasian Basin and this difference shows up in oceanographic properties such as the standing stock of fresh water (Aagaard and Carmack 1989) and silicate concentrations (McLaughlin *et al.* 1996). The partition between basins is also clearly important to the large-scale distribution and exchange of contaminants as shown by radionuclides from Sellafield (see Section 2.6.5) and surface HCH (Section 2.6.1). Historically, the demarcation between basins has been considered to lie approximately along the Lomonosov Ridge where it is seen as an oceanographic front (Rudels *et al.* 1994). Recently, however, the position of this front has moved toward the alpha ridge (Figure 3.14) signaling a large displacement of the Pacific waters by those of Atlantic origin (Carmack *et al.* 1994, McLaughlin *et al.* 1996, Macdonald 1996). This "invasion" has been especially dramatic in the top several hundred metres and is clearly marked by the Sellafield radiotracers (see Figure 6.10). How frequently such invasions might occur and their role in the transfer of properties from one basin to the other is currently not well understood.

The Arctic Ocean is unique for its disproportionately large fractional area covered by shelves (30%) that act as a buffer between the coast and the interior ocean. They are seasonally exposed to the atmosphere and receive continental runoff. They comprise a significant component of oceanic contaminant budgets. The residence time of water on the shelves is an important feature because it influences how the shelves respond to inputs from the land (runoff, sediments, contaminants) and how long it takes, on average, for conservative water properties to escape the shelf. For the large Russian shelves, residence times are thought to be of the order of 1 to 3 years (Schlosser *et al.* 1994) but for the largest Canadian arctic shelf (Beaufort Sea), it is probably less than a year (Macdonald *et al.* 1989, 1995, Omstedt *et al.* 1994). Residence times for the narrow shelves north of the Archipelago are not known, but it is probably accurate to assume that they are less than a year because of flushing by shelf edge currents and outflow through the Archipelago. In terms of contaminant movement between the shelf and the interior ocean, the most important feature of shelves is that they capture particles (GESAMP 1993). As mentioned previously, only 10% of the particle load delivered to the coast by the Mackenzie River and coastal erosion escapes the shelf edge either as suspended sediment or in ice (Macdonald *et al.* 1997). Shelves, therefore, act to remove contaminants that are strongly particle-associated while allowing dissolved contaminants to pass through to the interior ocean with a delay of less than three years.

Shelves are often considered as "boxes" into which coastal runoff enters and undergoes exchanges at the shelf boundary (200 m). This "compartmentalization" facilitates the calculation of residence or flushing times for the shelves; however, the shelves cannot be viewed simply as uniform, well-mixed zones. Processes on the shelves (Figure 3.17) contribute to the redistribution of water masses (and their contaminant burdens) within the ocean. Freshwater entering during periods of rapid runoff will tend to transit the shelf in a buoyant surface layer. On the other hand, wind-driven upwelling will import deep, denser water from the interior ocean (200–400 m) up onto the shelf, and some of this water will be entrained into shelf waters and exported nearer the surface (Macdonald *et al.* 1987, Aagaard and Roach 1990). In winter, brines formed on the shelf move surface water and associated contaminants into deeper layers of the interior ocean (see Section 2.3.3). Because of coastal upwelling and brine production in flaw leads and polynyas, the shelves are much more important than the interior ocean for water-mass communication between surface and deeper water. Even though budgets can be constructed for runoff, ice melt and brine production (Macdonald *et al.* 1995), these processes are sufficiently complex that the rates of exchange between shelf and interior ocean are still subject to considerable uncertainty. Because shelves tend to be net exporters of ice (Reimnitz *et al.* 1994), shelves also have the potential to export freshwater, particulates and contaminants in the ice itself. This flux, while possibly important, has not been well studied for Canadian shelves (Macdonald *et al.* 1997) or any of the arctic shelves.

Within the Canadian Arctic, the Archipelago must be considered as a distinct oceanographic regime. It receives a predominantly one-way exchange from the Canadian Basin with a flow estimated at 1.7 Sv (Table 2.1). This flow together with the area of the Archipelago (0.7×10^6 km^2) and an average assumed depth of about 150 m implies a residence time for the water transiting the Archipelago of about half a year.

- *Regional differences within the Arctic Ocean are evident both between the two major basins in the interior ocean (i.e., the Canadian Basin and the Eurasian Basin). Historically, the demarcation between basins has been considered to lie approximately along the Lomonosov Ridge where it is seen as an oceanographic front. Recently, however, the position of this front has moved toward the alpha ridge signalling a large displacement of the Pacific waters by those of Atlantic origin.*
- *The Arctic Ocean is unique for its disproportionately large fractional area covered by shelves (30%) which act as a buffer between the coast and the interior ocean.*

2.4.4 Ocean-Sediment

Sediments are an important sink for contaminants. Substances that become strongly attached to particles (e.g., Pb, Pu, highly chlorinated PCBs, high molecular weight PAH) are removed from the water column by attachment to particles that settle, often as biologically-produced aggregates, and accumulate in sediments. Provided the contaminants remain attached to particles, and provided there are no processes to disturb the sediments, contaminants will ultimately be buried and prevented from recycling. The rate at which particle-associated contaminants are removed to the sediments depends on the particle sedimentation rate and the concentration of contaminant on the particles. This latter property depends in part on sediment properties like the organic carbon content and particle size distribution (see Macdonald and Thomas 1991). In addition to the settling of particles, some contaminants, like Cd, may enter sediments through diffusion into them from bottom water.

There are few measurements of particle fluxes in the Arctic Ocean. Hargrave *et al.* (1989b, 1994) used sediment traps to measure fluxes characteristic of regions under permanent ice cover. Peaks in particle sedimentation were observed during late summer (August–September) and mid-winter (December–January). Particle fluxes were low (1.1 g·m^{-2} a^{-1}) and not enough material was collected to analyse for contaminant content, but the seasonal pattern of deposition suggests that scavenging of particle-associated compounds from the water column is greatest during the late summer months. O'Brien *et al.* (1991) measured particle fluxes in seasonally ice-covered locations at the edge of the Canadian Beaufort Shelf. They found fluxes one to two orders of magnitude greater than those observed at the Ice Island, with flux maxima occurring during both spring and fall. Honjo *et al.* (1995) report August peaks in surface water suspended sediments in the Beaufort Gyre and TransPolar Drift (Figure 3.15). Particle fluxes in the Arctic Ocean, therefore, vary widely between the marginal seas where strong seasonal inputs of terrestrial and marine particles occur, and the interior ocean whose surface water is isolated from both terrestrial and atmospheric inputs, and where productivities, and therefore particle fluxes, are much lower (Subba Rao and Platt 1984, Macdonald *et al.* 1993).

Bottom sediments can also act as a source of contaminants. Particle-associated contaminants may be released to pore water and diffuse back into the water column, or they may be ingested by biota foraging in sediments and thus enter or re-enter the food chain. Sediment resuspension during storms, which occurs on shallow parts of the shelves (<20 m), is also an effective way to inject bottom sediments back into

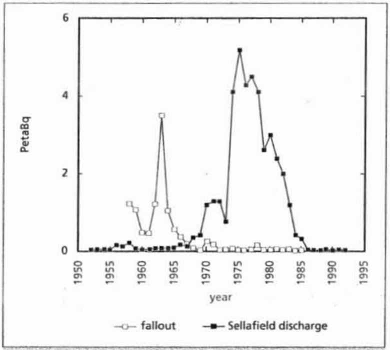


FIGURE 4.9

A comparison of the fallout record for ^{137}Cs from atmospheric weapons testing for the latitude band from 70°N–90°N (^{137}Cs fallout was estimated using available ^{90}Sr measurements from 1958–1990 given in Environmental Measurements Laboratory and the Health and Safety Laboratory reports and scaling these numbers upward by a constant factor of 1.6; data provided by M. Monetti) with the discharge of ^{137}Cs to the ocean from Sellafield nuclear reprocessing plant for the period 1951–1992 (data taken from Gray et al. *1995).*

the water column (Hill and Nadeau 1989). While these processes are clearly important to the ocean-sediment interaction, few quantitative measurements are available for the Canadian Arctic Ocean.

The disturbance and transport of shallow-water sediments by ice is unique to seasonally ice-covered seas (Reimnitz *et al.* 1993a, Rearic *et al.* 1990, Pfirman *et al.* 1995a). Turbulence and freezing in the water column during ice formation (suspension freezing) can effectively entrain bottom sediments by frazil and anchor ice formation (Pfirman *et al.* 1990, Riemnitz *et al.* 1994 and references therein). Generally, sediments incorporated into the ice by the above processes are then transported off the shelves to be deposited elsewhere (Figure 3.17). The rates of these processes are not well known for the Canadian shelves. According to Reimnitz *et al.* (1993a), suspension freezing in a coastal polynya off Alaska produced sediment burdens in ice of over 289 $t \cdot km^{-2}$ with subsequent export of sediment estimated at 6.7×10^4 t during a three-month period. Ice also scours the sediment in shallow water (0–20 m) forming furrows up to 6 m deep. Studies of scouring on the Alaskan Shelf suggest that in 6–14 m of water depth complete sediment reworking takes place to an average depth of 20 cm within 50 years (Reimnitz *et al.* 1977). In terms of frequency and volume of sediment suspension, most of the present ice scour is generated by ice shoves and pressure ridges formed at the outside edge of the ice landfast zone (Hequette *et al.* 1995).

In contrast to the wide distribution of pressure ridges in the Arctic Ocean, the formation of icebergs and tabular ice occurs at specific locations. Icebergs are derived from glaciers, whereas tabular ice is formed from ice shelves. In the Canadian Arctic, these ice formations originate from the Ward Hunt Ice Shelf off Ellesmere Island and fiords on the north coast of Axel Heiberg Island (Jeffries 1987, Jeffries *et al.* 1989). The Ward Hunt Ice Shelf has been the source of a number of tabular ice-islands of deep draft (i.e., 40–50m), including the Canadian Ice Island. Upon breaking off from the main body of the shelf, ice-islands tend to circulate with the Beaufort Gyre, often completing several circuits before leaving the Arctic Ocean in the Transpolar Drift (Jeffries and Shaw 1993). Some of the ice-islands may enter the Canadian Archipelago where they eventually disintegrate; however, there is only one observation of an ice-island drifting eastward after calving (Jeffries and Shaw 1993). Compared with pressure ridges, the role of icebergs and ice-islands in disturbing or moving sediments is minor and the main concern is their potential to collide with structures such as those used by the offshore oil industry (Sackinger *et al.* 1985).

Biomixing of surface sediments can also play a crucial role in sediment — ocean interaction and contaminant distribution in sediments. Biota that live in or on the sediments, and forage for food there, tend to mix the sediment surface. As a result, a surface mixed layer is often found in the one to ten centimetres of the sediment. Biomixing may move contaminants deeper into the sediments and initially dilute sedimenting contaminants. However, if contaminant input is reduced, biomixing also acts to maintain contaminant burdens in surface sediments by reintroducing deeper, contaminated material to the surface (Smith *et al.* 1994). The relative importance of biomixing can be estimated using a biodiffusion

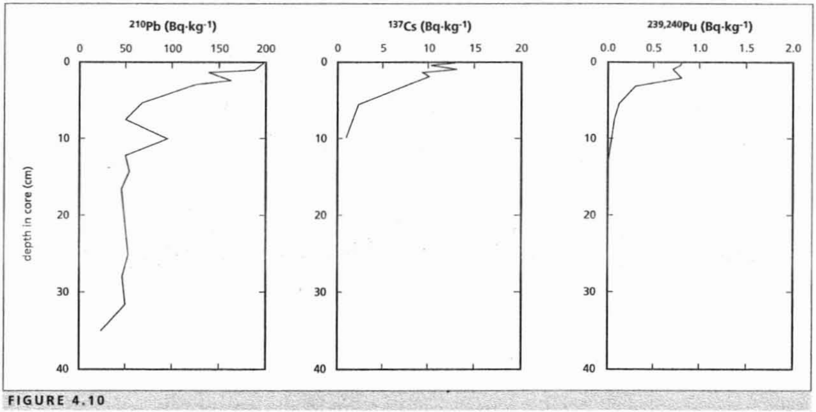


FIGURE 4.10

Vertical profiles of ^{210}Pb, ^{137}Cs and ^{239,240}Pu in a sediment core (SS-4) collected in the Beaufort Sea (^{210}Pb data are from Gobeil et al. 1991 and ^{137}Cs/^{239,240}Pu are from J.N. Smith unpublished).

model, which simulates the downward transport of particle-associated contaminants introduced at the sediment-water interface by a process analogous to diffusion. The relative importance of mixing versus burial can be evaluated using the contrast between fallout radionuclides, which came as a pulse centred in the early 1960s (Figure 4.9), and ^{210}Pb, which enters the sediments at a more or less constant rate and decays away to background after about 120 years. For example, a core collected in the Mackenzie Trough on the Canadian Beaufort Shelf (Figure 4.10) clearly exhibits strong surface mixing. First, fallout radionuclides (^{137}Cs and ^{239,240}Pu) penetrate to approximately the same depth (10 cm) where ^{210}Pb decays to background. Second, the top few cm of the sediments have uniform, elevated fallout radionuclide concentrations with no sign of the dramatic decrease in fallout that occurred after 1963 (Figure 4.9). These two observations can only be accounted for by mixing, which has redistributed fallout radionuclides to depth, and completely blurred the 1963 pulse. Since mixing affects the distribution of contaminants and not their total amounts, sediment inventories can still be calculated. For the core in Figure 4.10, these are ^{210}Pb–13 000 Bq·m^{-2}, ^{137}Cs–390 Bq·m^{-2} and ^{239,240}Pu–19 Bq·m^{-2}. The activity for ^{137}Cs falls near the average determined by Baskaran and Naidu (1995) for the eastern Chukchi Sea, suggesting that the sediment flux at this location can be explained entirely by radioactive fallout. Sediment mixing and burial rates determined from radionuclide tracers can therefore be used to predict the spatial distribution of sedimentation of other particle-associated contaminants such as metals (Pb) and organics (PAHs, PCBs).

Sediments and sedimentation rates are not evenly distributed within the Arctic Ocean. As discussed in Section 2.3.3, the shelves receive and capture most of the inorganic sediments introduced to the sea by rivers or coastal erosion. A regional sediment budget is a first step toward the understanding of ocean-sediment interactions. Such a budget should account for the sources of suspended solids, the main locations where sedimentation occurs (e.g., shelves, slopes and basin interiors) and the interactions between them (Stein and Korolev 1994, Stein *et al.* 1993). Once a reliable budget has been constructed for the solids, other components such as organic carbon or contaminants can be incorporated. Presently, such budgets are in a very primitive state. For the Canadian Beaufort Shelf, Macdonald *et al.* (1997) have calculated a budget for sediments and organic carbon. They suggested that about 50% of the 127 x 10^6 t of sediment delivered annually by the Mackenzie River is captured in the delta, 40% on the shelf and 10% escapes to the slope or interior.

A calculation of the flux of particle-bound contaminants to sediments requires a knowledge of sedimentation rate as well as the particular geochemical (e.g., redox chemistry) and biological (biomixing) processes within the core that might affect contaminant distribution after it arrives in the sediment. A knowledge of the latter is gained only by detailed compound- or element-specific geochemical studies of carefully collected sediment cores. Few such studies have been carried out for Canadian arctic marine sediments.

A number of factors conspire to make it difficult to estimate the sedimentary fluxes of contaminants in the Arctic Ocean using sediment cores:

(i) shelf sediments, which are the important locations for sedimentation of terrestrial inputs, are often subject to scour, resuspension and bio-mixing (Figure 3.17),
(ii) the flux of ^{210}Pb (a commonly used dating tool) tends to be low in the Arctic limiting the potential of this dating technique,
(iii) basin sediments accumulate slowly and the surface mixed layer often incorporates the entire industrial era, and
(iv) contaminant burdens in arctic marine sediments are often below detection limits.

Despite these difficulties, contaminant profiles from sediment cores can often be used to calculate the contaminant inventory (see above for radionuclides) and fluxes.

Few data sets exist that can be used to estimate sediment fluxes of contaminants. One example, the Pb concentration profile given in Figure 3.19, provides an opportunity to illustrate the difficulty in estimating the sediment flux component of the contaminant Pb budget. The excess Pb in the top of this particular core implies a contaminant Pb inventory of about 35 µg·cm^{-2} (Gartside *et al.* 1995). If, as a crude approximation, we consider that most of the anthropogenic Pb has been deposited after Pb was first put into gasoline, the implied average flux over that time period (1927 to 1991) for this location is about 0.5 µg·cm^{-2}·a^{-1} . This is actually a very large flux. For example, if it is applied to the whole of the Canada Basin (3 x 10^6 km^2), we obtain a flux of 15 000 t·a^{-1}, which is much larger than the 2400 t·a^{-1} total atmospheric contaminant Pb input to the Arctic estimated for 1979/80 by Akeredolu *et al.* (1994). The contrast is even larger considering that only 11% to 14% of this input is estimated to be actually deposited within the Arctic. Conversely, an annual rate of delivery of 300 t of Pb distributed evenly over the Arctic Ocean (10^7 km^2) implies an "average flux" of 0.003 µg·cm^{-2}·a^{-1}. Such an average flux, therefore, would be hard to detect in regions of active biomixing, rapid sedimentation or variable natural Pb flux. The main difficulty with this basin-wide flux estimate based on a single core is that we do not know what area of the basin the sediment core fairly represents. The contradiction between the calculated sediment flux for the core data in Figure 3.19 and the source function can be reconciled if, for example, the marginal sediments (perhaps 10% of the area) are relatively much more important for contaminant Pb fluxes than the interior basin sediments. This seems highly plausible because scavenging processes and ice contaminant delivery processes probably converge at the ocean margins. Currently, we lack sufficient aerial coverage for any contaminant in sediments to determine how fluxes are distributed and thereby to make realistic sediment flux calculations.

Cd is captured in sediments by diffusion and precipitation at depth in authigenic sulphide phases (Pedersen *et al.* 1989, Rosenthal *et al.* 1995). To estimate the capture rate requires profiles in sediments of pore-water Cd concentration for which none presently exist in the Arctic Ocean. However, as shown in Section 2.6.4, Cd fluxes can be estimated from other marginal seas. While it is possible to derive a reasonable estimate of total Cd flux to sediments, it appears highly unlikely that contaminant Cd fluxes can be measured from sediment core data because they are small relative to the natural background fluxes.

- *The disturbance and transport of shallow-water sediments by ice in shallow waters of continental shelves is unique to seasonally ice covered seas. Turbulence and freezing in the water column during ice formation can effectively entrain bottom sediments into frazil and anchor ice. Generally, sediments incorporated into the ice by the above processes are transported off the shelves to be deposited far away. The rates of contaminant resuspension and export in ice are not well known for the Canadian shelves.*

2.4.5 Lake-Sediment

Lake-sediment exchange comprises the processes of deposition, resuspension of particulate matter, and bidirectional sediment-water diffusion of dissolved chemicals. In arctic lakes, more than 90% of contaminants are dissolved due to the extremely low concentrations of suspended particles. Organic carbon, hypothesized to affect OC and PAH partitioning, also occurs in very low concentrations because of low biological productivity in arctic and subarctic lakes.

Results of mass balance studies from Amituk Lake suggest that the partitioning of OCs on particles is not related to the organic carbon content of particles (Freitas 1994). Rather, mineral matter, especially the clay fraction, is a major factor in sorption. This has been demonstrated in other low organic carbon systems such as groundwater aquifers (Curtis *et al.* 1986, Grundl and Small 1993). This effect is most pronounced for low K_{ow} chemicals such as HCHs and toxaphene. The discrepancy between empirical results and partitioning predicted from K_{oc} is greatest for low K_{ow} chemicals.

Less than 10% of the total loadings of PCBs and DDT in Amituk Lake are deposited to the sediments (Table 3.12, Figure 3.25). For chemicals that are less associated with particles (e.g. HCH, chlordane, PCB) less than 2% is deposited to sediments. The opportunity for wind-driven resuspension is minimal because the lake is ice-covered for 48 weeks of the year.

Depending on the chemical, sediment-water diffusion may play a role in sediment-water exchange. However, diffusive rates are slow compared with rates of particle movement, particularly at the cold temperatures in arctic lakes. Model estimates for Amituk Lake suggest that for the low K_{ow} chemicals such as HCHs, there is minimal *net* diffusion between sediments and water. For the high K_{ow} chemicals, the net diffusion rate is from sediments to water.

In arctic lakes, chemicals that enter the sediments will persist due to minimal resuspension, low rates of sediment-water diffusion and low rates of biological activity that could transform the chemicals. Muir *et al.* (1995) and Lockhart (1994) have calculated burial rates for a series of arctic and subarctic lakes. These are discussed in detail in the next section (Table 5.2, Figs. 5.6 to 5.9) on contaminant trends.

2.5 Contaminant Trends in the Arctic

2.5.1 Atmosphere

Since the early 1980s, many measurements of the atmospheric concentration of HCHs have been made at locations in the Canadian and Norwegian Arctic, and the Bering and Chukchi seas. These observations and recent ones made in the NCP program at Alert (Fellin *et al.* 1996, Figs. 3.1 and 3.2) and at Resolute Bay (Bidleman *et al.* 1995a), show a significant decrease in both α-HCH and γ-HCH concentrations between 1978 and 1990 (Bidleman *et al.* 1995b) in all seasons. Between June and November, ΣHCH declined from 880 $pg \cdot m^{-3}$ in 1979 to approximately 100 $pg \cdot m^{-3}$ in 1992/93. The decline was not linear but occurred in steps, decreasing after 1982 and between 1990 and 1992. Seasonal measurements (winter–spring defined as December–May, and summer–fall defined as June–November) correspond roughly to the haze and non-haze seasons, respectively. In both seasons, atmospheric concentrations of α-HCH (Figure 5.1) declined significantly ($p < 0.01$) with a 50% decrease over 4 years in summer-fall and over 6 years in winter-spring. Concentrations of γ-HCH also showed a decline in the Canadian Arctic and the Bering and Chukchi seas region. No time trend was apparent in the Norwegian Arctic. A likely consequence of the decrease in air concentrations is a reversal in the direction of exchange of HCH from air-to-ocean in the early 1980s to ocean-to-air in the 1990s (Figure 4.8). As yet, however, little temporal change has been observed in surface Arctic Ocean HCH concentrations (Bidleman *et al.* 1994).

Heavy metals such as Pb, Cu, Zn and non-soil V (x-V) have been routinely measured in the Arctic. x-V is anthropogenic in origin. It is calculated by subtracting soil V (derived from the product of the ratio of V to Al mass concentration in soil and aerosol Al concentration) from the concentration of atmospheric V measured. Downward trends since the late 1960s have been reported for concentrations in glacial ice at Summit, Greenland for Pb and Zn, but not for Cu (Boutron *et al.* 1991). In glacial ice, Pb and Zn are definitely of anthropogenic origin while Cu originates in part from soil. At Alert for the period from 1982 to 1993, the concentrations of these metals in air show a distinct seasonal variation with a maximum from January to March and a minimum in summer (Sirois and Barrie 1997). The long term trend at this high arctic station during winter (Figure 5.2) shows a marked decrease in Pb and in x-V between 1982

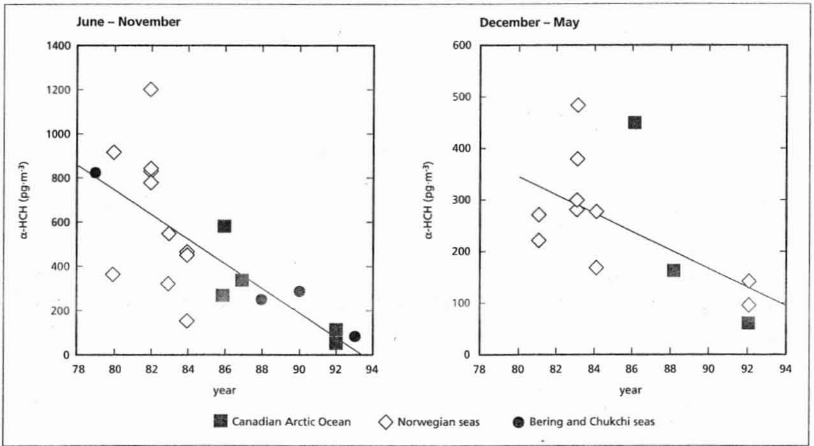


FIGURE 5.1

Summer-fall (June–Nov.) and winter-spring (Dec.–May) concentrations of α-HCH in air over the Canadian Arctic Ocean (black squares) and Norwegian (diamonds), Bering and Chukchi seas (black circle) reported by Bidleman et al. *1995b. The trends are plotted on a linear scale.*

and 1993. In contrast, long-term variations of Zn and Cu are less clear. Copper concentration decreased markedly in 1993 after a recent peak in 1990 and 1991. Whether the Cu trend reflects the economic state of the smelting industry in Russia, or whether it is due to meteorological variations remains an open question. In contrast to the situation in Greenland snow (Boutron *et al.* 1995), the concentrations of Cu in winter (January–March) aerosols at Alert are much higher (75 to 230) than in soil indicating that this element is anthropogenic in origin in the atmospheric aerosol in winter. There are few measurements of atmospheric Hg in the Arctic. The first observations made routinely for a year at Alert are shown in Figure 3.9. No information on temporal atmospheric trends in the Arctic is available. Long-term observations are needed.

Long term-trends of anthropogenic radionuclide concentrations in the atmosphere have been reviewed by Barrie *et al.* (1992). The trend of annual ^{137}Cs fallout in the Arctic (Figure 4.9, previous section) typifies that of atmospheric nuclear bomb test products. It peaked in the early 1960s and is now close to natural background levels.

2.5.2 Snow/Ice

Ice and snow cores can proved a history of the deposition of atmospheric contaminants from the pre-industrial revolution to present day. At the Agassiz Ice Cap on Ellesmere Island Peters *et al.* (1995) and Gregor *et al.* (1995b) have measured the concentration of PAHs in glacial snow and ice back to 1803. The time series for fluorene and phenanthrene (two PAHs typical of anthropogenic sources) are shown in Figure 5.3. Generally, concentrations were lowest in pre-industrial times, peaking in the 1950s and 1960s and then decreasing to a relatively constant but still elevated level in the period from the 1970s to the 1990s. The lack of change in the last 30 years does not reflect trends in global fossil fuel combustion, which has increased from 2.60 gigatonnes (Gt) in 1961 to 6.19 Gt in 1991 (Marland *et al.* 1994). Thus, PAH concentrations in arctic snow are not simply related to total combustion but are likely a complex function of the type of fossil fuel consumption.

Gregor *et al.* (1995a and b) examined long-term trends in PCB deposition to the Agassiz Ice Cap (Figure 5.4). PCBs show little post-depositional loss of from the snowpack (Section 2.4.1, Table 4.3). Prior to 1957, PCBs were absent, which is consistent with the synthesis and production history of these chemicals. Then, in the period 1957 to 1963, they appeared in glacial snow in small amounts (mean annual deposition 9 ng·m^{-2}·a^{-1}). Thereafter, deposition to glaciers was much higher with a mean annual deposition in the 1963 to 1993 period of 406 (± 187) ng·m^{-2}·a^{-1}. No significant temporal trend was evident in this period. More measurements of PCBs in glaciers are needed with special attention paid to congener patterns and post-depositional effects.

Changes in heavy metal concentrations in the Greenland Ice Cap are summarized in Table 5.1

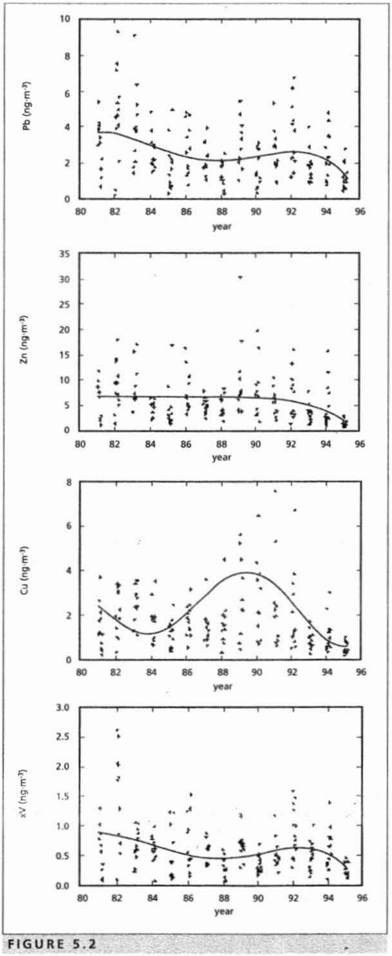


FIGURE 5.2

Long-term trends in weekly mean air concentrations of anthropogenic metals Pb, Zn, Cu and x-V (non-soil V) in aerosols at Alert, NWT (Figure 1.1) from Sirois and Barrie (1997).

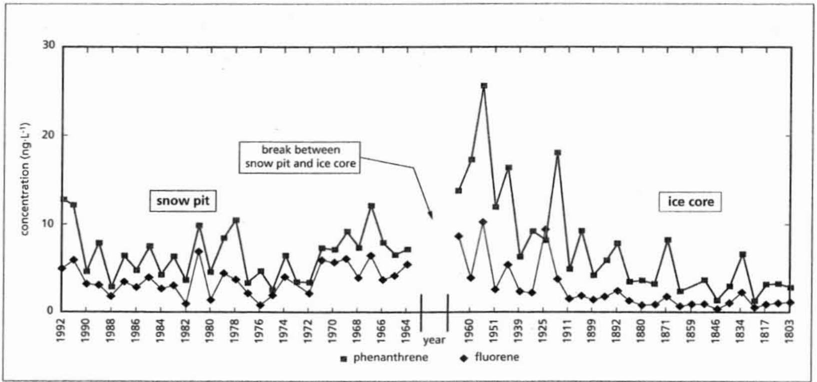


FIGURE 5.3

Long-term trend of fluorene and phenanthrene concentrations in glacial snow and ice on Agassiz Ice Cap, Ellesmere Island, NWT (Gregor et al. *1995b, Peters, A.J., pers. comm.)*

(Boutron *et al.* 1995). Pb concentrations increased from about 10 pg·g^{-1} in the second half of the 1700s and the early 1800s to about 200 pg·g^{-1} in the early 1990s, amounting to a 20-fold increase. However, this total increase is made up of a slow increase from the 1750s to the 1940s, followed by a much more rapid post-war increase through to the mid-1960s. This rapid increase was due to increasing use of alkyl-Pb additives in gasoline in the post-war era until the early 1970s. Initiatives to eliminate Pb in gasoline beginning in the 1970s has resulted in a marked decrease in the concentrations of Pb in Greenland snow by a factor of 7.5 between 1970 and the early 1990s. Present concentrations are roughly similar to those observed in the 1930s. Pb concentrations in air at Alert have decreased much less (by 50%, Figure 5.2) than those in Greenland Ice for the same period. This reflects the greater influence of Eurasian emissions compared to North American ones at Alert near sea level, than on Greenland at 2.5 to 3 km altitude. Pb isotopic measurements on Greenland confirm this conclusion. In Greenland snow, Cd, Zn, and Cu all increased greatly from pre-industrial times to a peak in the 1960s and 1970s decreasing somewhat thereafter.

TABLE 5.1

Mean ratio of change in Pb, Cd, Zn and Cu concentrations in the Greenland Ice Cap snow from the 1750s to early 1990s (Boutron *et al.* 1995).

Time interval	Pb	Cd	Zn	Cu
1750s–mid-1960s	+20	+5	+5	+3
Late 1960s–present	−7.5	-2.5	−2.5	−1.3

2.5.3 Lake Sediments

As part of the NCP, a geographically extensive investigation of contaminant trends of OCs and PAHs was undertaken in order to investigate the fate of OCs and metals in lake systems and the contaminant levels in biota. To examine latitudinal and temporal variations in contaminant deposition, Muir *et al.* (1996) measured OCs in sediment cores from eight lakes along a mid-continental transect from 49°N to 82°N (Figure 5.5). For all OCs, concentrations were highest in the top layer (median age 1960). In Figures 5.6 and 5.7, the depth profiles of various OCs are presented.

The three high arctic lakes (Sophia, Amituk and Hazen) showed DDT maxima appearing later than the other locations. Detectable concentrations in slices with median ages that pre-date the use of DDT, especially in Sophia Lake, may be due to sediment movement. Sedimentation rates were very low. In all lakes, HCB and pentachlorobenzene (PCBz) peaked in concentration after 1960 (Figure 5.6). The period of maximum production of chlorobenzenes in the USA was in the mid-1960s (Oliver *et al.* 1989) Thus, these mid-continental lake sediments suggest a 5 to 10 year lag in the peak in arctic lakes. The atmospheric input function of PCBs derived by Rapaport and Eisenreich (1988) is compared with the PCB sediment accumulation profile for four lakes in Figure 5.7. Studies of Lake Ontario sediment cores show that the sedimentary record accurately reflects the trend in global PCB sales (Eisenreich *et al.* 1989, Oliver *et al.* 1989). The mid-latitude and sub- arctic lakes (e.g., L375, Belot and St. Therese) all show an onset of PCBs in the 1940s (±10 years) in reasonable agreement with the source function. The high arctic cores, Amituk, Sophia and Hazen, differ markedly from the source function with significant PCB inputs not noticeable until the 1960s (±10 years), lagging the initiation of production by about 20 years.

High latitudes experience higher proportions of lower chlorinated PCB congeners. Airborne PCBs in the Arctic are dominated by the di-, tri- and tetrachlorinated congeners (Figure 3.4). In the Agassiz glacier snowpack (Gregor *et al.* 1995a) and in subsequent studies of snowfall and snowpack (Figure 3.13), a similar congener distribution was found. Thus, inputs to the aquatic environment of the high Arctic, either via snowmelt or gas exchange, differ from those in more temperate regions. Even though lower chlorinated congeners are more water-soluble and consequently tend to remain in the water column, they are also predominant in arctic lake sediments (Muir *et al.* 1996). This may in part be due to the decrease in the use of the more highly chlorinated Aroclors in US domestic sales between 1956 and 1974 (Muir *et al.* 1996) but it is more likely due to the preferential removal of the higher chlorinated compounds from air masses during transport from source area to the Arctic (Franz *et al.* 1997).

Recently, the mid-continental survey of lakes has been broadened to include lakes in the western NWT (Lockhart 1994) and the Yukon (Muir and Lockhart 1994). The data show that the sediment record is due to deposition and within lake processes. Also, it is clear that ΣPAH deposition varies regionally, and that recent inputs are lower than earlier in this century. Profiles in Lake Laberge in the Yukon have ΣPAH concentrations that fluctuate over a relatively narrow range for the past 60 years with highest concentrations in the pre-1900 slices. These early elevated levels are believed to coincide with woodburning during the gold rush at the end of the last century.

Lake Laberge in the Yukon has quite different ΣDDT profiles than other lakes in the Yukon and the NWT as illustrated in Figure 5.8 for the Yukon Lakes. Highest concentrations of ΣDDT in Lake Laberge are seen in slices from the period dated 1940–1946 while the other lakes show maximum deposition more recently (Muir *et al.* 1994b). The peak loadings of ΣPCB to Lake Laberge also occurred in the 1940s. The congener composition of ΣPCB changed down the core with the older 1945 to 1955 slices dominated by relatively heavy hexachlorobiphenyls whereas the younger surface slices are dominated by lighter di-, tri- and tetra-chlorinated biphenyls. The latter matches present atmospheric and snow congener patterns (Figure 3.13).

These results suggest that there were relatively local inputs of Aroclor oils and DDT upstream of Lake Laberge in the 1940s and early 1950s, whereas the current deposition is more indicative of long-range transported pollution. The other two lakes in the Yukon — Kusawa and Fox — do not show the dominant hexachlorobiphenyl pattern indicating that these lakes did not receive the proximal Aroclor oil input.

Small, low closure lakes (flooded annually as part of the annual freshet but otherwise connected by a distributary channel to the main river) in the middle Mackenzie delta have been investigated in association with the Mackenzie River suspended sediment study. Selected preliminary data for one lake with undisturbed sediments are presented in Figure 5.9 (Graf Pannatier *et al.* 1996). For the last three decades, there is an indication of a general, but slight increase in the sediment of the concentration of ΣPCB (~4.44 $ng \cdot g^{-1}$ over 30 years, significant at 95%) and the sum of pentachlorobenzene and hexachlorobenzene

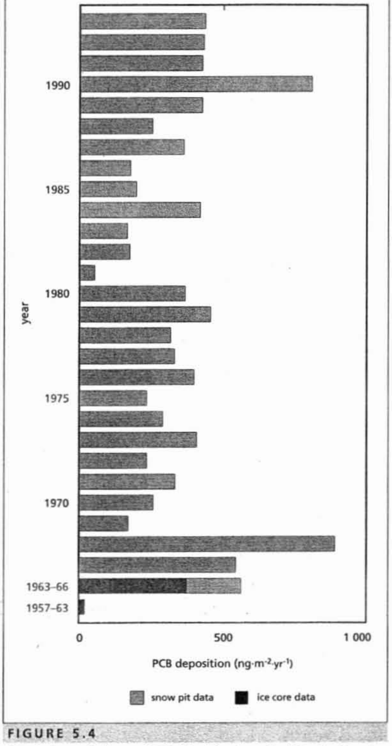


FIGURE 5.4

Historical record of annual PCB deposition ($ng \cdot m^{-2} \cdot yr^{-1}$) to the Agassiz Ice Cap, Ellesmere Island, NWT. Mean annual values are shown for the periods 1963–1966 and 1957–1963. PCBs were not detected in ice core samples prior to 1957 (Gregor et al. *1995)*

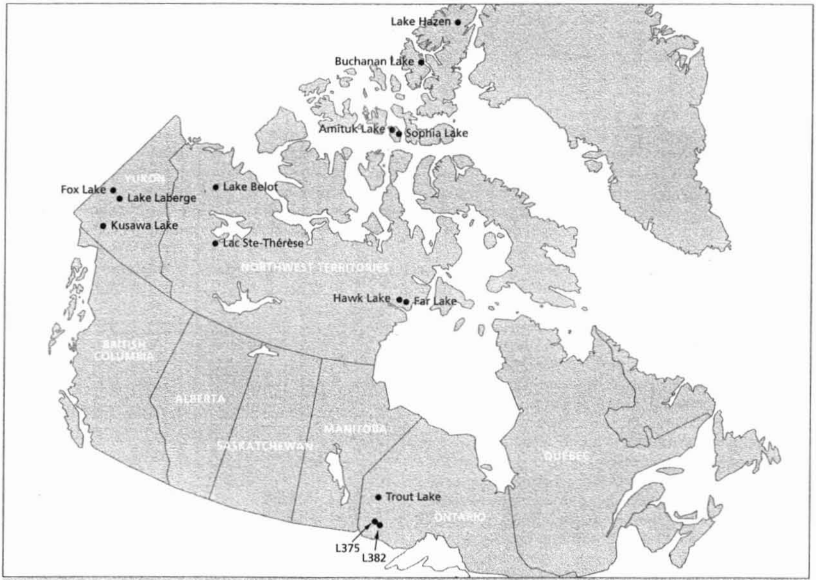


FIGURE 5.5

The location of freshwater lakes at which OCs and mercury were measured in sediment cores (Muir et al. 1996, Lockhart 1994).

(PeCB + HCB) (~0.12 ng·g^{-1} over 30 years, significant at 95%).

In 1994, cores were collected from YaYa Lake in the Mackenzie Delta (Lockhart 1997). Cores from this lake indicate that the deposition of Hg, Cd and Pb has not increased noticeably over the past several decades. Thus, it appears that, in this lake, geological sources of metals are more important than atmospheric sources. In contrast, Lockhart *et al.* (1994, 1995) reported that sediment cores from many NWT lakes (with the exception of Lakes Hazen and Buchanan) are receiving loadings of Pb and Hg that are considerably higher than those in the past (see Section 2.6.4). Surface and historic mercury fluxes and the associated enrichment factor (i.e., ratio of post-industrial to pre-industrial deposition) for Hg in the sediments of nine lakes are presented in Table 5.2.

TABLE 5.2

Comparison of surface and deep fluxes of mercury (µg·m^{-2}·a^{-1}) to core sites in arctic lakes and Hudson Bay. In addition, enrichment factors calculated as the ratio of the current flux to the historic flux (Lockhart *et al.* 1995) is given. See Figure 5.5 for site locations.

Location	Surface flux (µg·m^{-2}·a^{-1})	Pre-1800 flux (µg·m^{-2}·a^{-1})	Enrichment Factor
Lake 375	21.3	7.4	2.9
Hawk Lake	5.0	0.7	7.0
Far Lake	7.7	2.3	3.3
Amituk Lake	28.4	7.0	4.1
Lac Belot	3.8	3.5	1.1
Lac Belot	4.3	2.5	1.8
Lac Belot	3.9	3.3	1.2
Colville Lake	8.1	4.2	1.9
Fox Lake	8.1	5.3	1.7
Kusawa Lake	8.5	5.3	1.6
Lake Laberge	17.7	14.5	1.2
Hudson Bay	26.7	19	1.4
Hudson Bay	47.3	31.3	1.5

- *In parallel with trends in global usage of HCH, there has been a significant decrease in observed arctic atmospheric concentrations between 1978 and 1990. Sharp decreases occurred in the mid 1980s and in 1991. There has been an attendant reversal in the direction of exchange of HCH from air-to-ocean in the early 1980s to ocean-to-air in the 1990s.*
- *Between 1980 and 1995 at Alert, NWT, atmospheric lead has decreased significantly by about 50%. This is consistent with changes in atmospheric lead emissions in Europe and Russia but not with those in North America. The latter were decreased by more than a factor of ten while Eurasia has chosen to phase-out Pb usage in auto fuel more slowly.*

- *In Canada, historical records of contaminants in glacial ice have been reported for two classes of compounds ΣPCBs and selected PAH compounds and for one glacier (Agassiz Glacier on Ellesmere Island). PCBs are subject to mild volatilization losses during summer months. The record shows the appearance of PCBs in about 1960 after they were synthesized and commercially distributed. There is no trend evident after 1960. PAHs did show a trend. They peaked in the 1950s and 1960s and then settled down to a relatively constant concentration from 1970 to 1992. More measurements are needed of OCs, PAHs, Pb and Hg in northern glaciers.*

- *In lake sediments, there is a clear record of contaminant deposition history. Sub-arctic lakes show an onset of PCBs in the 1940s (±10 years) in reasonable agreement with synthesis and release history. In contrast, high arctic lakes show no significant PCB inputs until the 1960s (±10 years), lagging the initiation of production by about twenty years. This difference is consistent with large scale model predictions of the lag time in diffusion of PCBs released at mid-latitudes to the north and with glacial ice PCB records.*

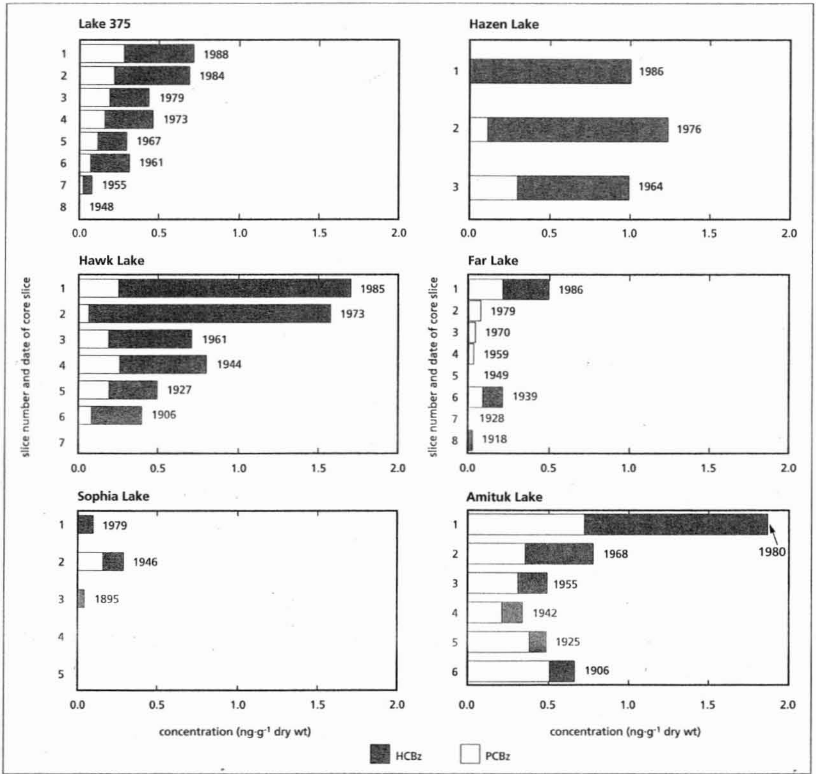


FIGURE 5.6

Profiles of HCB and PCBz (combined bars equal ΣCBz) in six sediment cores from lakes along a mid-continental transect from 49°N to 82°N (see Figure 5.5 for site locations). From Muir et al. *(1996).*

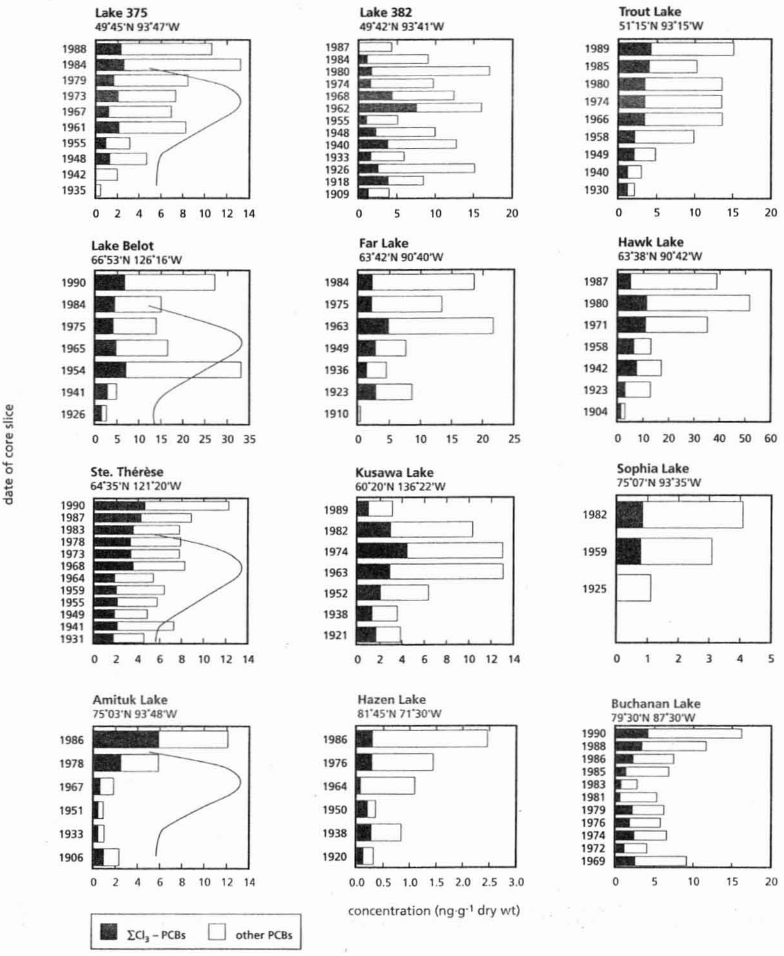


FIGURE 5.7

Profiles of tri-chlorinated PCBs and other PCBs in freshwater lake sediments (from Muir et al. *1996). See Figure 5.5 for site locations. The line is the PCB input to peat cores in northwestern North America (Rapaport and Eisenreich 1988).*

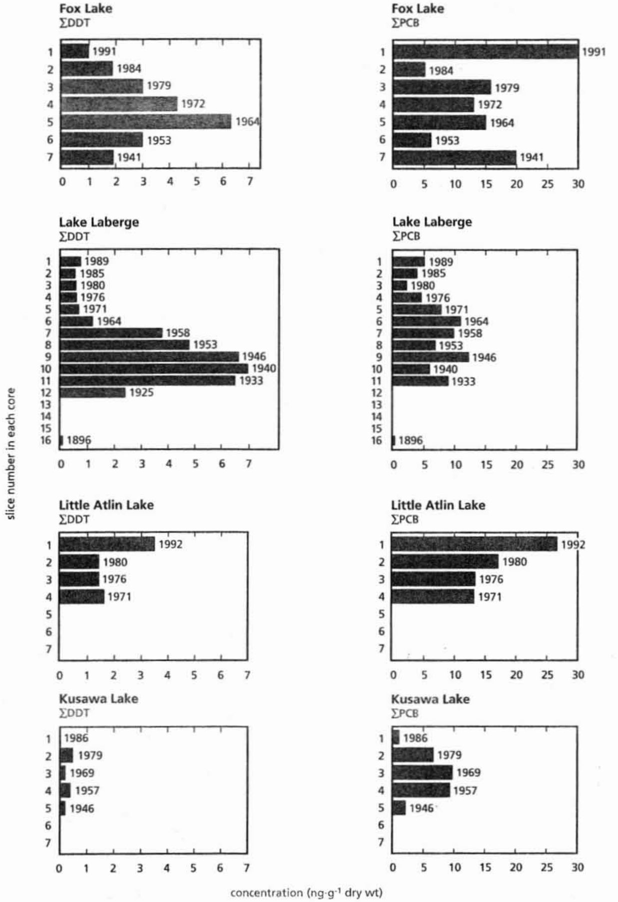


FIGURE 5.8

Profiles of ΣDDT and ΣPCB in dated (i.e. year beside bar) sediment cores from four Yukon lakes. The dates are preliminary estimates (Muir et al. 1994b).

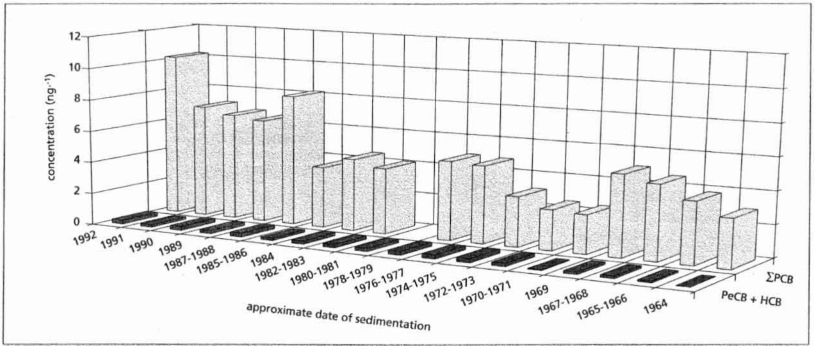


FIGURE 5.9

Historical record of ΣPCB and pentachlorbenzene (PeCB) plus HCB concentrations in a Mackenzie River delta lake core sediment for the period 1964 to 1992 (Graf Pannatier and Gregor 1996).

- *Lake sediment records of anthropogenic Pb and Hg show clearly that anthropogenic sources contribute substantially to lake inputs. Unlike synthetic organic compounds such as the OCs, these metals also have a natural geological source. The ratio of the current deposition flux to the natural, pre-industrial flux in numerous lakes sampled ranges from 1.1 to 7 for Hg and from 1.1 to 10 for Pb.*

2.6 Special Topics

2.6.1 Hexachlorocyclohexane Mass Balance in the Arctic Ocean

Hexachlorocyclohexane (HCH) has been produced since World War II and is one of the few organochlorine insecticides still in large-scale use today. Two types of HCH products are manufactured throughout the world. A technical mixture containing 60–70% α-HCH, 5–12% β-HCH, 10–15% γ-HCH and minor proportions of other isomers, was mainly used in Asian countries during the 1980s (Iwata *et al.* 1993a). Canada, the United States, and most countries in Europe and the Southern Hemisphere use pure γ-HCH (lindane), the only isomer with insecticidal properties. Global production has been estimated as 550 000 tonnes of technical HCH and 720 000 tonnes of lindane (Table 2.4).

HCH compounds volatilize soon after application, especially in the tropics (Takeoka *et al.* 1991), and are atmospherically transported to the Arctic and other

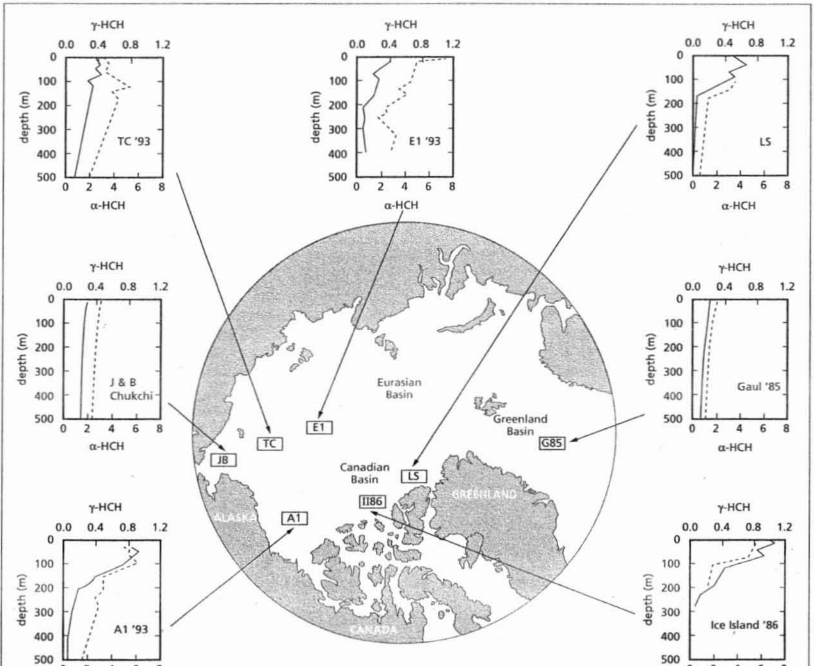


FIGURE 6.1

Vertical profiles of HCH for the Arctic Ocean. Data have been taken from Gaul (1992) (G85), Hargrave et al. *(1988) (Ice Island) (II86), Jantunen and Bidleman (1995) (Chukchi) (JB) Macdonald* et al. *(1996) (A1, TC, E1, LS).*

remote regions. The low Henry's Law constants of HCHs (Figure 2.11) favour partitioning from air into water, especially at low temperatures. At equilibrium the ratio of α-HCH concentration in seawater to that in air is 3000 at 25°C, but 22 000 at 0°C. The world's oceans are the major reservoir of HCHs. Global models estimate that approximately 20% of the HCH present in the environment is held in the ocean surface layer, which is taken to be 75 metres (Strand and Hov 1996) or 200 metres (Mackay *et al.* 1995). Even though the heaviest use of HCHs has been in tropical and subtropical regions (Section 2.2.2), levels in surface seawater are an order of magnitude higher in the Arctic (Iwata *et al.* 1993, Schreitmüller and Ballschmiter 1995, Figure 2.18). Concentrations of HCHs in tree bark increase with latitude, being relatively high in Canada, Nordic countries, Alaska and Russia, and lower in tropical countries (Simonich and Hites 1995). Thus, the transport and distribution of HCHs exemplifies the "cold condensation" effect (Wania and Mackay 1993a, 1995).

In particular, toxaphene has air-water partitioning characteristics similar to the HCHs and would be expected to follow similar transport pathways in this regard. The difference between HCHs and less volatile OCs (e.g. PCBs, chlordanes, DDTs and toxaphene) is that the fate of the latter substances is influenced to a greater extent by wet and dry deposition of particles from the atmosphere and sedimentation in the water column.

Barrie *et al.* (1992) formulated a mass balance for HCHs in the Arctic Ocean using air and water concentrations typical of the mid-1980s. The budget was based on limited data for HCHs in ocean water and rivers, and assumed that atmospheric levels remained constant over the year. Deposition of HCHs was calculated for two seasons, winter and summer. The estimated standing stock of HCHs in the upper 200 m of the water column was 8100 tonnes with a residence time of 20 to 30 years. Major inputs to the Arctic Ocean were by ocean currents (63%), atmospheric deposition (30%) and river runoff (7%). Losses were mainly by outflow of water to the North Atlantic through the Canadian Archipelago (78%), the East Greenland Current (16%), and to a lesser extent by other currents (4%) and ice export (2%).

Abrupt changes in the release of HCHs to the environment appear to have taken place in recent years, which have been manifested in declining atmospheric concentrations (Figure 5.1). This assessment report provides an opportunity to update the HCH budget, taking into account advances in knowledge. Improvements are made in constructing the new budget by:

i) using atmospheric HCH concentrations measured in the early 1990s,
ii) estimating atmospheric fluxes on a monthly basis rather than biannually,
iii) incorporating new data on the spatial variability of HCHs in the Arctic Ocean,
iv) making use of more detailed information on the circulation, ice cover and water budget of the Arctic Ocean, and
v) updating estimates on the input of HCHs by rivers and ocean currents. Details of the new budget calculations are presented in the following sections.

2.6.1.1 Budgets for Water and Dissolved HCHs in the Arctic Ocean

For purposes of the budget, the "Arctic Ocean" is defined as selected waters above 65°N: the Canada and Eurasian basins, the Canadian Archipelago, and the following regional seas: Barents, Kara, Laptev, East Siberian and Chukchi. Excluded are Baffin Bay, Hudson Bay, the Norwegian Sea and the Greenland Sea. Even though portions of these waters lie above 65°N, they are not included because their circulation patterns place them outside of the Arctic Ocean regime.

The HCH budget is constructed using the volumetric flows for freshwater, sea water and ice into and out of the Arctic Ocean (Table 2.1) together with the respective estimated concentrations of HCH. The results of the calculations and the sources of HCH data are summarized in Table 6.1 (see also Figure 6.1 and Table 6.2, which show vertical HCH profile data and integrated water column burdens for various regions). One of the problems in constructing this budget is that there are no synoptic surveys that include measurements of HCH in all the important water masses within a time frame of a few years. Values for the Eurasian Basin and the regional seas in the Russian Arctic are especially lacking. Therefore, the values selected for Table 6.2 incorporate data collected at various times during the past decade.

HCHs are not uniformly distributed within the Arctic Ocean (Figures 2.20 and 6.1). A large proportion of HCH is contained in the top 200 metres of the water column. The Canada Basin contains a pool of surface water where HCH concentrations are elevated by a factor of two or more compared with other regions of the Arctic Ocean, or oceans to the south. To estimate the burden of HCH, we partitioned the Arctic Ocean into two domains; the "North American Arctic Ocean" (NAAO = 2.57×10^6 km^2), which includes the Canada Basin and the Archipelago and accounts for approximately 25% of the total ocean area, and the "Eurasian Arctic Ocean" (EAO = 7.72×10^6 km^2), which includes the Eurasian Basin and the regional seas. Table 6.2 shows the burden of HCH estimated from available vertical profile data (Figure 6.1). For the two domains in the Arctic Ocean, we estimate it to be: NAAO = 2200–2600 t of α-HCH and

280–420 t of γ-HCH; EAO = 2200–2400 t of α-HCH and 500–630 t of γ-HCH.

For the removal of HCHs by ice flow through the Archipelago and the Fram Strait, we have relied mostly on the measurements of HCH levels in ice collected at the Canadian Ice Island (Hargrave *et al.* 1988). Representative HCH concentrations for the Bering Sea have been determined (Jantunen and Bidleman 1995) which, when combined with reliable estimates of inflow through the Bering Strait, makes this the best-determined component of the HCH budget. The estimate of HCH in runoff is uncertain, particularly for the Russian rivers. Backus *et al.* (1995) measured a fairly uniform, low range of HCH concentrations in the Mackenzie River and eleven other small Canadian arctic rivers. In contrast, data for the Russian rivers in 1992–1993 (Roshydromet 1995), show a number of sporadic very high points and average HCH concentrations well above those found in the North American rivers. In the budget, we have used 1994 α- and γ-HCH concentrations, which range from 0–8 ng·L^{-1}. Our estimation of the exchanges between the Atlantic and Arctic Oceans is based exclusively on Gaul's data (1989, 1992). For the East Greenland Current, we have partitioned the water column into two domains (Foldvik *et al.* 1988) and have chosen HCH values based on the vertical distribution given by Gaul (1989).

As previously discussed (Section 2.4.4), there are insufficient data at present to make a reliable determination of HCH flux to sediments as part of the budget. HCHs are neither very particle-reactive nor bioaccumulative, and therefore they are often below detection limits in suspended particles or in sediments (e.g., Hargrave *et al.* 1988, 1989a). For these reasons, HCH fluxes to sediments were assumed to be small in the previous HCH budget (Barrie *et al.* 1992). However, in a highly productive region of the Chukchi Shelf, Macdonald *et al.* (1996) measured α-HCH concentrations in shallow-water surficial sediments as high as 0.23 ng·g^{-1}, suggesting that fluxes in some locations might be important.

To assess the potential significance of sediments as a sink for HCH, we make the following simple estimates. Concentrations of α-HCH on suspended particles, estimated to be <1 ng·g^{-1} from Hargrave *et al.* (1989a), and average sedimentation rates for the Ice Island site (2 mg·m^{-2}·d^{-1}, Hargrave *et al.* 1989b) yield a sedimentation rate of α-HCH <2 pg·m^{-1}·d^{-1}. For the sake of comparison to other mass flow values in Table 6.1, this rate extrapolated to the entire Arctic Ocean implies a deposition to the sediments of only 0.008 t·a^{-1} — which is negligible by several orders of magnitude in comparison to the other terms in Table 6.1. Taking a different approach, let us assume that suspended sediments from the Mackenzie River scavenge HCHs and then accumulate at a

TABLE 6.1

HCH budget for the surface Arctic Ocean, representative of the early 1990s.

		Water Concentration			Flux t·a^{-1} (% of total)	
	Water Flow 10^4 km^3·a^{-1}	α-HCH µg·m^{-3}	γ-HCH µg·m^{-3}	Ref.	α-HCH	γ-HCH
INPUT						
Bering Strait	2.6	2.0 ± 0.48	0.45 ± 0.10	a	52 (22)	12 (20)
Norwegian Coastal Current	2.2	1.3 ± 0.3	0.5 ± 0.2	b	29 (12)	11 (18)
Barents Sea	3.78	1.0 ± 0.25	0.22 ± 0.08	b	39 (16)	8.3 (14)
W. Spitzbergen Current	6.3	1.0 ± 0.25	0.22 ± 0.08	b	65 (27)	14 (23)
Rivers:						
– N. American	0.33	0.1–1.3	0–0.3	c,d	4 (2)	1 (2)
– Asian	(total flow)	1–7	0–8	e		
Atmospheric					51 (21)	15 (24)
OUTPUT						
Atmospheric					−65 (17)	−7.5 (13)
E. Greenland Current (Polar Water)	3.15	0.9 ± 0.2	0.2 ± 0.04	b	−28 (7)	−6 (10)
E. Greenland Current (Atlantic Intermediate Water)	7.3	0.7 ± 0.2	0.16 ± 0.02	f	−51 (13)	−12 (20)
Canadian Archipelago	5.35	3.4 ± 0.5	0.49 ± 0.12	g	−181 (47)	−27 (45)
Ice	0.52	1.3 ± 0.3	0.19 ± 0.05	h	−6.8 (2)	−1 (2)
		0.27–1.0	0.1–0.46	f		
Hydrolysis				i	−55 (14)	−6 (10)
Sedimentation					negl.	negl.

[a] *Jantunen and Bidleman 1995.*
[b] *Gaul 1992.*
[c] *Backus* et al. *1995*
[d] *Jeffries, pers. comm.*
[e] *Roshydromet 1995.*
[f] *Gaul 1989.*
[g] *Hargrave, pers. comm.*
[h] *Hargrave* et al. *1988.*
[i] *Calculated from rate constants in Ngabe* et al. *1993*

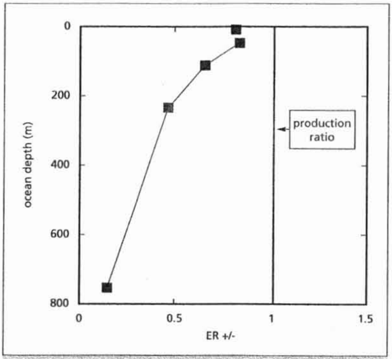


FIGURE 6.2

Profile of the (+) α-HCH/(-)α-HCH enantiomer ratio (ER) with depth at a station in the Greenland Sea. The pesticide is produced with a racemic (1.00) ratio. An ER less than 1.00 implies selective breakdown of the (+)α-HCH enantiomer.

concentration equivalent to that observed on the Chukchi shelf (0.23 ng·g^{-1}). Observed HCH concentrations on the Mackenzie shelf are actually far smaller than this assumed value. At a sediment delivery of 127 x 10^6 t·a^{-1} from the Mackenzie River, this still amounts to only 0.03 t·a^{-1} of HCH. Based on these lines of reasoning, it appears unlikely that sedimentation plays much of a role in the overall HCH budget for the Arctic Ocean, even though there may be locations where the vertical fluxes are enhanced by high primary productivity.

HCHs are hydrolyzed in basic solution to yield first pentachlorocyclohexenes and finally trichlorobenzene. Second-order rate constants for the initial reaction of α- and γ-HCH with OH^- were determined as functions of temperature by Ngabe *et al.* (1993). Using these constants the half-lives of α-HCH and γ-HCH in seawater at pH 8.1 are estimated to be 0.4 and 0.5 years at 25°C, but increase to 60 and 100 years at 0°C. The fraction of the initial concentration that is hydrolyzed in one year is 0.0110 for α-HCH and 0.0064 for γ-HCH. Taking the surface layer burdens to be 5000 t of α-HCH and 1000 t of γ-HCH, hydrolysis accounts for annual losses of 55 t of α-HCH and 6 t of γ-HCH — amounts comparable to removal by other processes such as water outflows and volatilization (Table 6.1).

Little is known about microbial breakdown of organochlorine compounds in arctic waters. Wania and Mackay (1993a) suggested that their persistence in the polar environment is increased due to low temperatures, limited biological activity and the relatively small incidence of sunlight. However sedimentation and hydrolysis are also slow, and microbial attack may compete in removing HCHs from the water column.

The first evidence that organochlorine pesticides can be microbially degraded in arctic waters has recently been obtained from the changing proportion of α-HCH enantiomers. Chiral pesticides like α-HCH are produced as racemates, which contain equal proportions of right- and left-handed enantiomers. In the environment these enantiomers are broken down at different rates by enzymatic activity. Selective degradation, which is expressed by the enantiomer ratio, ER = (+)α-HCH/(-)α-HCH, is not the same for all organisms. Preferential metabolism of either (+) or (-) α-HCH has been found among different species of birds, marine mammals and terrestrial mammals (Hummert *et al.* 1995, Hühnerfüss *et al.* 1993, Müller *et al.* 1992, Mössner *et al.* 1992), and in water from different regions of the North Sea (Faller *et al.* 1991). Selective breakdown of (+)α-HCH was found in water from Resolute Bay and in Amituk Lake on Cornwallis Island (Falconer *et al.* 1995a,b). ERs in these waters ranged from 0.77–0.93. These changes are assumed to be caused by microbial activity, since abiotic mechanisms such as hydrolysis and photolysis are not enantioselective.

Transects of the Bering-Chukchi seas (BERPAC-93) (Jantunen and Bidleman 1995) and the Arctic Ocean (AOS-94) (Jantunen and Bidleman 1996) provided an opportunity to investigate α-HCH enantiomers in more detail. Selective breakdown of α-HCH was found in the Canada Basin and the Greenland Sea,

TABLE 6.2

HCH burden in the top 200 m of the water column from vertical profiles of HCH (see Figure 6.1 for locations, Units are µg·m^{-2}).

Location	Date	α-HCH	γ-HCH	Source
N. Atlantic	1985	182	37	Gaul 1992
Ice Island	1986	800	97	Hargrave *et al.* 1988
Bering Sea	1993	341	78	Jantunen and Bidleman 1995
Siberian Chukchi Seas	1993	350	98	Jantunen and Bidleman 1995
Beaufort Sea (A1)	1992	680	87	Macdonald *et al.* 1996
Beaufort Sea (A1)	1993	780	130	Macdonald *et al.* 1996
Chukchi Sea (TC)	1993	460	120	Macdonald *et al.* 1996
East Siberian Sea (E1)	1993	375	130	Macdonald *et al.* 1996
Lincoln Sea (LS)	1993	510	90	Macdonald *et al.*-1996
Archipelago[1]	1993	440	98	Falconer *et al.* 1995a, Hargrave *et al.* 1997

[1] *Depths in the Canadian Archipelago are less than 200 m in many locations.*

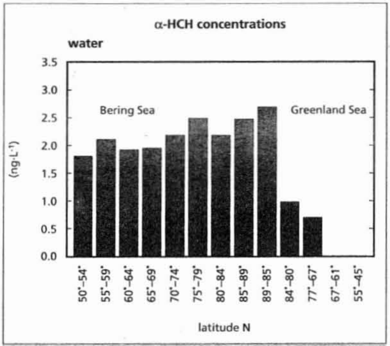


FIGURE 6.3A

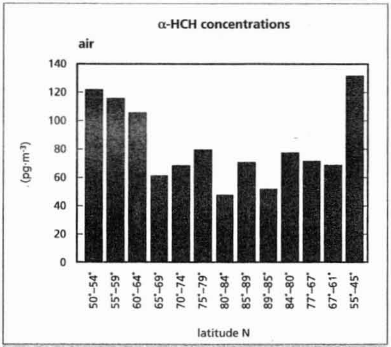


FIGURE 6.3B

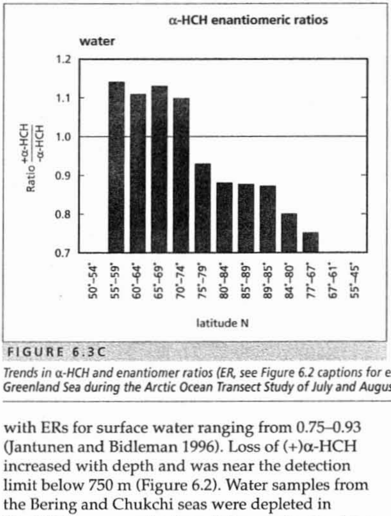


FIGURE 6.3C

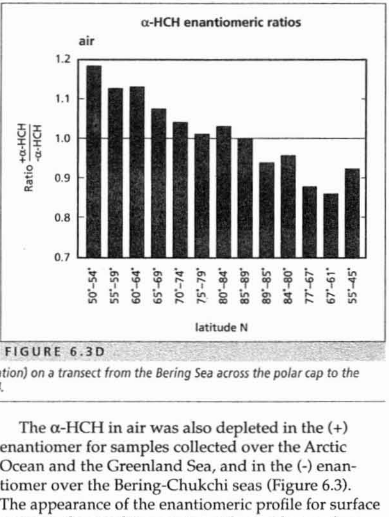


FIGURE 6.3D

Trends in α-HCH and enantiomer ratios (ER, see Figure 6.2 captions for explanation) on a transect from the Bering Sea across the polar cap to the Greenland Sea during the Arctic Ocean Transect Study of July and August 1994.

with ERs for surface water ranging from 0.75–0.93 (Jantunen and Bidleman 1996). Loss of (+)α-HCH increased with depth and was near the detection limit below 750 m (Figure 6.2). Water samples from the Bering and Chukchi seas were depleted in (-)α-HCH, an opposite degradation preference from the Arctic Ocean (Figure 6.3). Reasons for this are not known, but may be related to different microbial populations in these regions. Enantiomeric signatures alone cannot provide absolute rates of microbial breakdown, since only the ratio of compounds is measured. The kinetics might be estimated *in situ* by making use of other chemical tracers to determine turnover rates and lifetimes of different water masses and coupling this information with ER and HCH concentration data.

The α-HCH in air was also depleted in the (+) enantiomer for samples collected over the Arctic Ocean and the Greenland Sea, and in the (-) enantiomer over the Bering-Chukchi seas (Figure 6.3). The appearance of the enantiomeric profile for surface water in the overlying air provides evidence of sea-to-air volatilization (see below).

2.6.1.2 The Atmospheric HCH Budget

Concentrations of HCHs (particularly α-HCH) in arctic air have fallen over the last 15 years, from ~800 pg·m^{-3} in 1979 to ≤100 pg·m^{-3} in 1992–94. A three-fold drop has taken place since 1988–90 (Bidleman *et al.* 1995a, Jantunen and Bidleman 1995, 1996) (Figure 5.1). Although reliable production data are difficult to obtain, the atmospheric trend signals

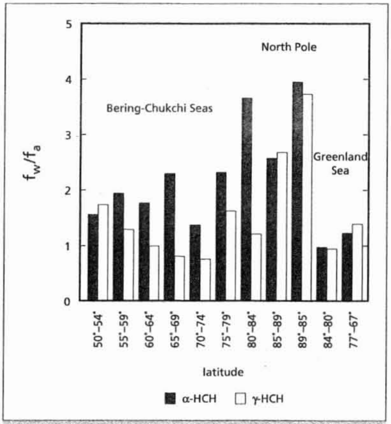


FIGURE 6.4

Water/air fugacity ratios (f_w/f_a) of α-HCH and γ-HCH on a transect from the Bering Sea to the Greenland Sea in July and August 1994. A fugacity ratio of 1 indicates air-water equilibrium. A ratio different than 1 indicates the potential for net deposition (i.e. <1) to or volatization (i.e. >1) from the ocean.

that applications of technical HCH have been curtailed by one or more major users. One of the heaviest users of technical HCH was India, which produced 25 000–47 000 t·a^{-1} throughout the 1980s (Hinckley *et al.* 1991, Iwata *et al.* 1993a). Airborne HCHs measured in 1989 over the Arabian Sea and Bay of Bengal near India were as high as 10 000–32 000 pg·m^{-3} (Iwata *et al.* 1993a). A 1992–93 survey of the same region showed greatly reduced concentrations (Iwata *et al.* 1995). In 1990, India began a program to phase out technical HCH and stopped its application to food crops. In the future India will be placing more emphasis on the use of lindane (γ-HCH; B.S. Parmer, Indian Agricultural Research Institute, letters to T.F. Bidleman). Russia also banned technical HCH in 1990 (Li *et al.* 1996).

Reducing the atmospheric concentration of HCHs over the ocean has the same effect as uncorking a bottle of champagne — the partial pressure of gas above the liquid is lowered and the supersaturated wine releases gas to the atmosphere. The net exchange direction of gaseous HCHs between air and sea is controlled by the fugacity (partial pressure) in surface water relative to air. The water/air fugacity ratio is calculated from (McConnell *et al.* 1993):

$$f_w/f_a = C_w{\cdot}H/C_aRT_a \qquad (6.1)$$

where C_w and C_a are the HCH concentrations in water (dissolved) and air (gas) (ng·m^{-3}), H is the Henry's Law constant at the water temperature (Pa m^3·mol^{-1}), T_a is the air temperature (K) and R = 8.314 Pa m^3·mol^{-1} K.

In the 1980s mean fugacity ratios of α- and γ-HCHs were 0.74 and 0.28 in the Arctic Ocean, and 0.76 and 0.55 in the Bering-Chukchi seas. That is, the surface water was 28–76% saturated with respect to the partial pressure of HCHs in air. In these years, air-to-water deposition of gaseous HCHs was estimated to have contributed 80% of atmospheric loadings, or 63 t·a^{-1}, to the Arctic Ocean (Barrie *et al.* 1992, Cotham and Bidleman 1991). The 1992–94 fugacity ratios measured in the Canadian Archipelago (Falconer *et al.* 1995a), the Bering-Chukchi seas (Jantunen and Bidleman 1995) and on AOS-94 (Jantunen and Bidleman 1996) were >1.0 at most stations (Figure 6.4). This implies that northern waters are now oversaturated and outgassing HCHs to the atmosphere. This tends to offset, but not eliminate, the reduction in HCH air concentrations in the Arctic that are associated with a reduction of global HCH usage (see Section 2.2.2).

Fugacity calculations which show the potential for α-HCH volatilization are supported by the enantiomeric ratios (ERs) of α -HCH in arctic air. Surface waters of the Bering-Chukchi seas are depleted in (-)α-HCH, whereas (+)α-HCH is preferentially degraded in the Canada Basin and the Greenland Sea (Figure 6.3). This leads to ERs which are >1.0 in the Bering-Chukchi seas and <1.0 in the Arctic Ocean. The α-HCH in air samples collected over these seas shows the same order of enantioselectivity as the surface water. The presence of non-racemic α-HCH in the atmospheric boundary layer suggests that a portion is derived from volatilization from the ocean and can be differentiated from the racemic pesticide that has been atmospherically transported from source regions (Jantunen and Bidleman 1996). Although the enantiomeric composition of α-HCH in air at lower latitudes is not well defined, racemic α-HCH was found in air samples collected from a continental area (South Carolina) and the subtropical North Atlantic at Bermuda (Bidleman 1997).

The framework for the atmospheric portion of the budget and descriptions of the process parameters are presented by Barrie *et al.* (1992) and Cotham and Bidleman (1991). Deposition mechanisms include:

a) rain and snow scavenging of particulate HCHs (associated with haze aerosols),
b) dry deposition of particulate HCHs,

TABLE 6.3

Air-Surface Exchange Processes in the Arctic

Process	Flux Term
Rain/snow scavenging of particles	$W_p\phi C_aP$
Rain scavenging of vapours	$W_g(1-\phi)C_aP$
Snow scavenging of vapours	$V_{s,g}(1-\phi)C_a$
Dry deposition of particles	$V_d\phi C_a$
Air-sea gas exchange, deposition	$K_a(1\phi)C_a$
Air-sea gas exchange, volatilization	k_aC_wH/RT_a
Volatilization from the snowpack	Not calculated

c) rain scavenging of vapour-phase HCHs,
d) adsorption of vapours to falling snow, and
e) air-to-sea gas exchange.

Processes that release HCHs to the atmosphere from within the Arctic are:

f) volatilization from ocean surface water, and
g) volatilization from ice and snow.

The equations used to calculate these air-surface fluxes are summarized in Table 6.3. Process parameters are defined and sources for their values are given in Table 6.4.

Uptake and release of vapour-phase HCHs by snow and ice are expected to be significant (Wania and Mackay 1995, Hoff *et al.* 1995). Estimating the contribution of these processes to the HCH budget is, however, highly uncertain because the specific surface area of fresh snow changes by two orders of magnitude as the snow ages (Hoff *et al.* 1995). Using a surface area:volume ratio of snow = 1×10^5 m^{-1}, the amount of HCHs scavenged from the atmosphere by snow at -30° is estimated to be about 10–30 times greater than by rain at 0°C (Figure 4.2a).

The atmospheric budget takes into account monthly variations in the atmospheric concentration of HCHs, haze aerosols, air temperature, precipitation and the area of the Arctic Ocean that is ice-free. The total area used for calculating loadings by wet and dry deposition is 1.03×10^7 km^2. Based on recent surveys of HCHs in surface seawater (Table 6.1, Figure 6.1), the concentrations in the Canada Basin and the Archipelago are about twice those in the Eurasia Basin and the regional seas. As discussed above, we divided the Arctic Ocean into regions of "high" (NAAO = 2.57×10^6 km^2) and "low" (EAO = 7.72×10^6 km^2) HCH concentrations, representing 25% and 75% of the total ocean area. These areas, adjusted for monthly ice cover, were used for gas exchange estimates.

Precipitation and dry deposition fluxes of particulate and vapour-phase species were calculated from the relationships in Table 6.3. The fraction of HCHs on particles was estimated from the Junge-Pankow adsorption model (see Section 2.2.3), using the subcooled liquid vapour pressure as a function of temperature. Details of these calculations are given by Cotham and Bidleman (1991). Because HCHs are relatively volatile, even at arctic temperatures, sorption to haze aerosols does not play a large part in their environmental distribution. Percentages sorbed to particles were highest during March, reaching 2% for α-HCH and 6% for γ-HCH.

Scavenging of vapour-phase HCHs by snow during winter is estimated from the model of Hoff *et al.* (1995), assuming a surface area:volume ratio for snow = 1×10^5 m^{-1} and an average raindrop diameter of 0.24 mm. The calculation is carried out for an eight-month winter at a temperature of -30°. Although this is simplistic, the much larger uncertainty in the specific surface area of snow precludes using a more detailed approach.

Gas exchange fluxes were estimated from the concentration of vapour-phase HCHs in air and dissolved HCHs in surface water. Corrections were made for the small fraction of HCHs bound to atmospheric particles. HCHs are relatively water-soluble and sorption to particles and colloidal organic matter in the water column is expected to be negligible (Cotham and Bidleman 1991). Separate terms were calculated for deposition and volatilization (Table 6.3); the difference is the net flux.

Gas exchange fluxes were converted into the mass of HCHs depositing or volatilizing by assuming

TABLE 6.4

Process Parameters and Environmental Data.

		Frequency	Source
C_a	Concentration in air, $ng \cdot m^{-3}$	Monthly	Fellin *et al.* 1996, Barrie, unpub.
C_w	Concentration in water, $ng \cdot m^{-3}$	Constant	Jantunen and Bidleman 1995, 1996
F	Flux, $ng \cdot m^{-2} \cdot d^{-1}$	Monthly	Calculated
F_{open}	Fraction of open water	Monthly	Gloerson and Campbell 1988, 1991, LeDrew *et al.* 1992
H	Henry's Law constant, Pa $m^3 \cdot mol^{-1}$	Monthly (rain) Constant (seawater)	Kucklick *et al.* 1991
k_a	Mass transfer coefficient for air, 518 $m \cdot d^{-1}$	Constant	Cotham and Bidleman 1991
P	Precipitation rate, $m \cdot d^{-1}$	Monthly	Barrie *et al.* 1992
ϕ	Fraction of HCHs on aerosols	Monthly	Calculated
R	Gas constant, 8.314 Pa $m^3 \cdot mol^{-1}K$	Constant	
T_a	Air temperature, K	Monthly	Barrie *et al.* 1992
T_w	Water temperature, 273 K	Constant	
V_d	Particle dry deposition velocity, 86 $m \cdot d^{-1}$	Constant	Cotham and Bidleman 1991
W_p	Washout ratio for particulate HCHs, $C_{precip}/C_a\phi$		Cotham and Bidleman 1991
	100 000	Jan.–Apr., Sep.–Dec.	
	300 000	May–Aug.	
W_g	Washout ratio for HCH vapours $C_{precip}/C_a(1-\phi) = RT_a/H$	Monthly	Calculated

values for the area of the ocean that is unfrozen and available for gas exchange. The extent of sea ice cover and area of open water within the ice pack has been measured by a multichannel microwave radiometer from the NASA Nimbus-7 satellite operated from 1978–87 (Gloerson and Campbell 1988, 1991). Their estimates of the proportion of the entire Arctic Ocean that is ice-free ranged from 11% in February to 64% in August. These figures are heavily weighted by the large areas of open water in the regional seas, and we used them for the EAO. The ice cover in the Canada Basin is greater. Satellite imagery for 1987–90 in the Beaufort Sea shows ≥90% cover in January–February and 10%–60% in August (LeDrew *et al.* 1992). Satellite data collected in August–September 1994 in connection with the AOS-94 cruise show 10%–50% ice cover between 70°N to 75°N and 90%–100% at higher latitudes. We assumed the open water in the NAAO to be 5% and 25% of the total area in February and August. Linear changes in ice cover were taken for the summer-winter transitions in the EAO and NAAO.

The relative magnitude of the atmosphere-ocean exchange pathways was discussed in Section 2.4.1 (Figure 4.8). Gas exchange is the dominant process, accounting for 89% of the atmospheric budget for α-HCH and 63% for γ-HCH. Loadings of α-HCH to the Arctic Ocean are 13 $t \cdot a^{-1}$ by precipitation (rain and snow scavenging) 0.1 $t \cdot a^{-1}$ by particle dry deposition and 38 $t \cdot a^{-1}$ by air-to-sea gas transfer. These inputs are more than offset by -65 $t \cdot a^{-1}$ volatilization, and the net result today is a loss to the atmosphere of -14 $t \cdot a^{-1}$. About one-third of this loss occurs from the NAAO. The potential for outgassing in the NAAO is actually greater than in the EAO due to the substantially higher α-HCH concentrations in the surface waters of the Canada Basin and the Archipelago (Figure 2.20), but volatilization is suppressed by the ice cover.

Inputs of γ-HCH to the Arctic Ocean by precipitation and dry particle deposition are 8.2 and 0.05 $t \cdot a^{-1}$. Deposition and volatilization of vapour-phase γ-HCH account for 6.2 and -7.5 $t \cdot a^{-1}$. The nearly equal atmospheric gain and loss by gas exchange results in a net loading of 7.0 $t \cdot a^{-1}$, mainly by precipitation. Thus the atmospheric budget for α- and γ-HCH in the early 1990s indicates a net loss of -7 $t \cdot a^{-1}$. By comparison, Barrie *et al.* (1992) estimated that the net loading of α-and γ-HCH in the 1980s was 79 $t \cdot a^{-1}$, 80% of which was by gas-phase deposition.

2.6.1.3 Overall HCH Budget for the Arctic Ocean

The complete budget for HCHs in the Arctic Ocean, representative of the early 1990s, is summarized in Table 6.1. Ocean currents provide 76% of the total HCH input, with atmospheric deposition and rivers contributing 22% and 2%, respectively. Inflow through the Bering Strait is the best-known component because of the detailed measurements of HCHs in the water column of the Bering and Chukchi seas in 1993 (Jantunen and Bidleman 1995). Advection into the Eurasian Basin is less certain because the only available measurements of HCHs in the Norwegian Coastal Current and the Barents Sea are from 1985 (Gaul 1992). Outflow of HCHs occurs mainly through the Canadian Archipelago (46%) and the East Greenland Current (22%). Volatilization and ice export account for 16% and 2% of the loss. The only chemical loss quantified in this exercise was hydrolysis, estimated to account for 14% of the removal of total HCHs. However, the enantiomer data suggest that microbial breakdown may also be an important sink.

The output/input ratio for α-HCH is 1.6, indicating a net removal of this isomer from the Arctic Ocean, whereas the budget is nearly balanced for γ-HCH. The upper 200 m of the Canada Basin contains α-HCH concentrations that are a factor of 2–3 above those in the Eurasia Basin and the regional seas (Figure 6.1). It is likely that loadings of HCHs to the Arctic Ocean from the atmosphere as well as ocean currents were greater in the past and were dominated by technical HCH mixtures that contained a high proportion of the α-isomer. The picture that emerges from this mass balance exercise is of the Arctic Ocean in steady state with γ-HCH and exporting α-HCH. Inputs and outputs of HCHs are dominated by ocean current advection, with about a 20% contribution by atmospheric processes, largely air-sea gas exchange. Microbial breakdown may also account for some loss of HCHs, but at the present time we are not able to quantify rates. In the future, we can expect to see a decline in the large pool of α-HCH in the Canada Basin due to drainage out through the Canadian Archipelago and to a lesser extent by sea-to-air volatilization. Outgassing will also result in a slow decline of both HCH isomers in waters of the Eurasia Basin and the regional seas, assuming that atmospheric concentrations continue their downward trend.

A number of uncertainties remain in the arctic HCH budget, which can only be remedied by further investigations:

a) Much work remains to be done on the physical oceanography of polar waters to establish the residence times of the various water masses and advective flows. Uncertainties in the advective flow estimates are on the order of 30% to 50% (Table 2.1).
b) The fraction of open water, which controls the air-sea gas flux, is a poorly known parameter that is subject to bias when estimated by the conventional technique of satellite imagery.
c) Although the HCH burdens of the Canada Basin and the Bering-Chukchi seas have been relatively well established, the database for HCHs in the water column of the Eurasian Basin and its regional seas is weak, most of the measurements

being from 1985. Better coverage of these regions is needed to establish the burden of HCHs in the water column and concentrations in the major currents feeding and draining the Arctic Ocean.

d) Improved estimates of HCH concentrations in Russian rivers are needed.

e) Microbial breakdown of HCHs in the water column is a potentially significant sink which needs to be quantified, especially considering the long residence times of water masses in the central Arctic Ocean and the extremely low rates of other chemical processes (e.g., hydrolysis). Also, biota may be important in the seasonal cycling of HCHs and other pesticides in shelf waters by controlling rates of sedimentation (Hargrave 1995).

f) Scavenging and release of HCH vapours to snow are estimated to be significant components of the atmospheric budget (see Section 2.4.1 Fig 4.5) , but these processes are poorly quantified. Improvements are needed in the experimental measurement and modeling of scavenging by snow and in the estimation of the specific surface area of snow.

g) Further work needs to be done on the air-sea gas exchange component, especially with regard to improving estimates of mass transfer coefficients under different states of ice cover. Direct measurements of air-sea exchange would lend credence to predictions of the two-film model for estimating gas fluxes.

h) The concentrations of HCHs in ice given in Table 6.1 are based on few measurements and further data are required to better understand the role of ice in transporting HCHs within and out of the Arctic Ocean.

- *What do we know about the pathways of the most abundant OC in the Arctic Ocean? The budget of ΣHCH shows that ocean currents, the atmosphere and rivers contribute 76%, 22% and 2%, respectively to the total annual input to the Arctic Ocean surface layer. The outputs- via ocean currents, loss to the atmosphere, chemical destruction and ice export account for 69%, 16%, 14% and 2% respectively of the total annual loss. Outflow through the Canadian Archipelago accounts for two-thirds of the total marine outflow. Atmosphere-surface exchange is very significant.*

2.6.2 Toxaphene: Long-range transport and Air-Surface Exchange

Toxaphene is an insecticidal mixture consisting largely of chlorinated bornanes with a small proportion of chlorinated camphenes. The theoretical number of components containing 6–9 chlorines is over 16 000 (Vetter 1993), although only a few hundred of these are likely to occur in the environment to a significant extent (Hainzl *et al.* 1994). Because a number of toxaphene-like products are made in different countries, the term "chlorobornanes" (CHBs) is preferred as a generic name for environmental residues (Muir *et al.* 1993). Its use has been reviewed in Barrie *et al.* (1992). Even though CHB mixtures have been banned or severely restricted in many countries, continued input to the environment may occur from regions where use is still permitted and by volatilization from contaminated soils.

Residues of CHBs in arctic fish, beluga and narwhal frequently exceed those of other OCs such as DDTs, chlordanes and PCBs (Muir *et al.* 1996). Selective metabolism leads to profiles of CHB residues which are substantially altered from toxaphene and similar mixtures. This is most evident for narwhal and beluga blubber and human milk, where the bulk of the CHB residues is accounted for by only two compounds, an octa- and nonachlorobornane (Bidleman *et al.* 1993, Muir *et al.* 1992, Muir and deBoer 1995, Stern *et al.* 1992). Patterns of metabolism vary among cetaceans, pinnipeds, benthic invertebrates and sediments, giving rise to different CHB profiles (Hargrave *et al.* 1993, Miskimmin *et al.* 1995). CHB profiles in air and seawater show the results of "physical weathering," being enriched in the more volatile components (Bidleman *et al.* 1995a). The need to quantify these diverse residues led to an international workshop on "The Analytical and Environmental Chemistry of Toxaphene" (Bidleman and Muir 1993), which was followed by a number of advances in analytical methodology. Partial or complete structural identification of many components in the technical toxaphene mixture has been achieved and several CHBs are now commercially available as analytical standards. Muir and deBoer (1995) have reviewed recent developments in these areas. The results of an international round-robin study of CHBs in cod liver oil has been published (Andrews *et al.* 1995).

The widespread and relatively uniform occurrence of CHBs in marine mammals and fish from across the NWT (Muir *et al.* 1996) and in lichens across Ontario (Muir *et al.* 1993) points to atmospheric deposition as the primary source in remote regions. The specifics of food web structure can amplify the effects of atmospheric deposition, leading to elevated levels of CHBs in top predator fish. This was seen in Lake Laberge of the Yukon Territories where a commercial and sport fishery was closed because of CHB contamination (Kidd *et al.* 1995a,b, Muir *et al.* 1996, Schindler *et al.* 1995). This report summarizes the occurrence of CHBs in arctic air and water and estimates atmospheric loadings to the North American Arctic Ocean (NAAO).

2.6.2.1 CHBs in Arctic Air

CHBs were first detected in arctic air at Spitzbergen in 1981 (Oehme and Stray 1983), but no quantitative measurements were made. Concentrations of CHBs in air were determined during August–September 1986 and June 1987 at the Canadian Ice Island (Patton *et al.* 1989), February–April 1988 at Alert (Patton *et al.* 1991) and August–September 1988 over the Bering-Chukchi seas (Hinckley *et al.* 1991). Average CHB levels, quantified as toxaphene by capillary GC with negative ion mass spectrometry (GC-NIMS), were 40 pg·m^{-3} in summer and 17 pg·m^{-3} in winter.

Year-round monitoring of CHBs has been carried out at Alert and Tagish from 1992 to 1994, with analysis by gas chromatograph-electron capture detector (GC-ECD). Twelve of the Tagish samples were also analysed by GC-NIMS and yielded average values that were 2.7 times higher than ECD results (Muir, unpublished). In contrast, CHBs measured at the Ice Island by GC-NIMS were about a factor of two lower than ECD values (Patton *et al.* 1989). Table 6.5 gives the average CHB concentrations on a monthly basis for the combined Alert-Tagish 1992–94 data set as measured by GC-ECD and also recalculated on a GC-NIMS basis (adjusted values), assuming an NIMS/ECD factor of 2.7. Also given in Table 6.5 are the CHB results from summer 1992/93 air sampling at Resolute Bay (Bidleman 1995) and on the BERPAC-93 cruise in the Bering and Chukchi seas (Bidleman *et al.* 1995b). The discrepancy between the Resolute-BERPAC results, which were determined by GC-NIMS, and the adjusted Alert-Tagish values is a factor of between 2 and 3.5. This uncertainty limits our ability to predict current atmospheric loadings of CHBs to the Arctic Ocean and is a matter that needs to be resolved.

Although the number of samples from the 1980s was small, recent atmospheric measurements suggest that a decline in CHBs has taken place. A direct comparison between summertime measurements taken in 1986–88 (Ice Island, Bering-Chukchi seas) and 1992–93 (Resolute Bay, Bering-Chukchi seas) suggests that CHBs dropped by a factor of six over this time period (Table 6.5). In these studies CHBs were sampled by the same method (filter-polyurethane foam), the same air pumps and volume calibration procedures were used, and the analysis of all samples was done by GC-NIMS using the total peak area method of quantification. A second comparison can be made between the 1980s NIMS measurements and the 1992–1994 adjusted values from Tagish and Alert (Table 6.5), which suggests that the recent CHB levels are 1.5 to 2.5 times lower.

Potential sources of CHBs to the Arctic include atmospheric transport from countries where toxaphene-like products are still used, and volatilization of residues from contaminated soils. It is likely that CHBs have reached the Arctic via atmospheric transport from both Eurasia and North America. CHBs measured in the air of southern Sweden during 1984/85 ranged from 5 to 225 pg·m^{-3} and were highest during late spring and summer and when winds blew from the E-SE, suggesting transport from eastern Europe or western Russia (Bidleman *et al.* 1987). Based on limited information Siberia does not appear to be a significant source of toxaphene. Average concentrations of CHBs measured by GC-NIMS at Lake Baikal in June 1991 were 16 pg·m^{-3} (McConnell *et al.* 1993), scarcely higher than summertime concentrations in the Arctic (Table 6.2). Nearly 80% of the toxaphene applications in the US were in the southern states (Voldner and Schroeder 1989), and it is likely that volatilization of residues from agricultural soils in this region are contributing to the background concentrations in North America, including the Arctic. Despite a decade-old ban on toxaphene use in the US, relatively high concentrations of CHBs still occur in the southern US. The mean concentration of CHBs in Columbia, South Carolina during August 1994–January 1995 was 180 pg·m^{-3} (Bidleman 1997).

TABLE 6.5

Mean concentrations of CHBs in air from arctic and sub-arctic regions (pg·m^{-3}).

Month	Alert & Tagish[1] ECD 1992–94	Alert & Tagish[2] ECD Adjusted to NIMS 1992–94	Resolute Bay & Bering-Chukchi[3] NIMS 1992–93	Ice Island, Alert Bering-Chukchi[4] NIMS 1986–88
Jan.	2.9	7.8	—	—
Feb.	2.3	6.2	—	15
Mar.	3.6	9.7	—	19
Apr.	6.3	17	—	11
May	8.5	23	—	—
June	8.1	22	—	36
July	9.9	27	—	49
Aug.	7.4	20	5.5	44
Sept.	4.5	12	4.3	30
Oct.	3.8	10	—	—
Nov.	2.6	7.0	—	—
Dec.	2.2	5.9	—	—

[1] *Fellin* et al. *1996 and Barrie* et al. *(unpublished).*

[2] *Estimated from the comparison of two methods for 12 Tagish samples, for which the mean NIMS/ECD value = 2.7 (Muir, unpublished).*

[3] *Bidleman* et al. *1995a, Jantunen* et al. *(pers. comm.)*

[4] *Patton* et al. *1989, 1991, Hinckley* et al. *1991. The number of samples in each month varied from 1–7.*

2.6.2.2 CHBs in the Arctic Ocean and Regional Seas

CHBs were first identified in water at the Ice Island in 1986/87 and the concentrations in two surface (10 m) and two deep (225–270 m) samples were reported along with measurements of CHBs in air, snow, zooplankton and amphipods (Bidleman *et al.* 1989). On reviewing the reported GC-NIMS data, it was discovered that an error had been made in calculating the CHB concentrations in three of the water and all of the snow samples (other data in the paper are correct). The revised concentrations of CHBs in seawater from the Ice Island study are: surface = 145-175 $pg \cdot L^{-1}$, deep = 32-45 $pg \cdot L^{-1}$. More recent measurements were made at Resolute Bay in 1992–93 (Bidleman *et al.* 1995b, Hargrave *et al.* 1997) and on the BERPAC-93 and Arctic Ocean sections (AOS-94) cruises through the Bering and Chukchi seas and across the polar cap (Bidleman *et al.* 1996).

Figure 6.5 summarizes measurements of CHBs in surface water on the BERPAC-93 and AOS-94 cruises. Concentrations of CHBs increase from 15-30 $pg \cdot L^{-1}$ in the Bering-Chukchi seas to 90 to 120 $pg \cdot L^{-1}$ at higher latitudes. In comparison, CHBs in lakes within the Yukon River basin ranged from 20 to 270 $pg \cdot L^{-1}$. The spatial distributions of CHBs and HCHs in the Arctic Ocean are similar in that both pesticides are elevated in more northerly waters. The range of CHB concentrations from 1992/93 falls below the 1986/87 Ice Island measurements, but considering that only two surface samples were analysed in the latter study it is difficult to determine whether a real decline has occurred. Unlike HCHs, there are no measurements of CHBs in eastern arctic waters, including the Norwegian and Barents seas. Considering that currents in this region supply half of the HCHs to the Arctic Ocean (Table 6.1), a budget for CHBs cannot be attempted without information on these important transport routes.

The following scenario is suggested to account for the peak in CHBs and HCHs in the northern Canada Basin. Atmospheric levels of HCHs in the early to mid-1980s were about 4–8 times higher than present-day values (Bidleman *et al.* 1995a, Jantunen and Bidleman 1995, Figure 5.1), and there is a suggestion that CHBs were also higher in the last decade, perhaps by a factor of 2 to 6 (see above). Thus it is likely that atmospheric loadings of these pesticides were greater in the past, especially to the regional seas that surround the central Arctic Ocean. These seas are largely unfrozen during the summer months and are susceptible to gas exchange, whereas exchange at higher latitudes is inhibited by ice cover and surface stratification. Over time water containing high concentrations of CHBs and HCHs has been transported from the regional seas into the central Arctic Ocean. Today these seas have cleared themselves by outgassing and sedimentation and have become adjusted to reduced atmospheric inputs, but these processes do not operate efficiently in the high Arctic. CHBs and HCHs that are trapped under the polar cap are thus "ghosts of the past" and will be slowly drained, largely through the Canadian Archipelago, on a time scale of decades (see Section 2.2.3).

Outflow of polar water containing high concentrations of CHBs and HCHs will impact biota in the Archipelago and Baffin Bay. Hargrave *et al.* (1997) monitored OC pesticides in the upper 50 m of the water column at Resolute Bay for the year of 1993. Mean concentrations of HCHs (4100 $pg \cdot L^{-1}$) and CHBs (85 $pg \cdot L^{-1}$) were similar to levels found under the polar cap on AOS-95 (Figure 5.1 and 6.5). The mean concentration for August 1993 (58 $pg \cdot L^{-1}$) agreed excellently with 48 $pg \cdot L^{-1}$ found a year earlier (Bidleman *et al.* 1995a). Hargrave *et al.* (1997) found that CHBs in surface water were highest during winter-spring, and decreased in summer-fall. The decline paralleled an increase in phytoplankton productivity in July and elevated concentrations of particulate organic carbon, which persisted in the water column through early fall. This suggests that CHBs are scavenged from the water column by settling particles during and following the productive season. Fugacity calculations show that enhanced air-to-water gas exchange accompanies the decline in CHB surface water concentrations, thus demonstrating the close coupling between the atmospheric and biotic components of the system.

2.6.2.3 Atmospheric Budget of CHBs in the North American Arctic Ocean

Processes of atmospheric deposition and volatilization of CHBs to the North American Arctic Ocean (NAAO) were considered, using the framework developed for HCHs (Section 2.6.1). As discussed in that section, the NAAO accounts for about one-fourth of the Arctic Ocean and comprises the Canada Basin and the Archipelago. The two relevant physicochemical properties for modelling these processes are the liquid-phase vapour pressures (VP, Pa) and Henry's Law constants (H, Pa $m^3 \cdot mol^{-1}$) of CHBs. Vapour pressures have been measured as a function of temperature for a few CHB congeners, which allow estimates to be made of their sorption to haze aerosols (Bidleman 1995, Hinckley *et al.* 1990). However measurements of CHBs in arctic air and water have been made on a "total toxaphene" basis, so average properties for the toxaphene mixture must be used. The vapour pressure and Henry's Law constant of technical toxaphene have been measured at 20°C by Murphy *et al.* (1987). These were extrapolated to arctic temperatures using the equations:

$$\text{Log VP} = 12.25 - 4487/T \tag{6.2}$$

$$\text{Log H} = 11.48 - 3416/T \tag{6.3}$$

The slope of equation 6.2 is the average value for two hepta- and octachlorobornanes (Hinckley *et al.* 1990). Tateya *et al.* (1988) reported -3416 as the temperature slope of the Henry's Law constants for PCBs, and we assumed the same value for CHBs. Equations 6.2 and 6.3 were also used by Hoff *et al.* (1993b) to estimate the atmospheric deposition of CHBs to Lake Superior.

As discussed above, current estimates of CHB concentrations in arctic air differ by about a factor of three. Deposition of CHBs into the NAAO were estimated by assuming two sets of values for airborne CHBs, those determined by ECD ("low" values, Table 6.5, column 1) and adjusted to the NIMS scale ("high" values, Table 6.5, column 2). The contributions of dry particle deposition, precipitation and net gas exchange to the atmospheric budget for CHBs are shown in Figure 6.6 for high and low atmospheric CHBs. Dry deposition is highest during winter haze season when about 90% of the CHBs are predicted to be associated with aerosols according to the Junge-Pankow model. The monthly contribution of precipitation is fairly constant over the year due to particle scavenging during winter and gas scavenging during summer. Low concentrations of CHBs in air during winter leads to fugacity ratios >1.0 and net volatilization. However little outgassing takes place because of the small area of open water available for exchange. During summer the atmospheric concentrations are higher, fugacity ratios reverse to <1.0 (net deposition), and gas exchange maximizes because of the expanded area of open water.

Net atmospheric loadings to the NAAO are estimated to be 1.1 and 0.22 t/y for the high and low estimates of atmospheric CHBs. The five-fold range is greater than the uncertainty in atmospheric concentrations (about a factor of three) because the net input is primarily due to gas exchange, which responds to the water/air fugacity ratio. Other factors, such as limitations in ice cover data (Section 2.6.1), compound the uncertainty in the CHB loadings calculations. At the present time atmospheric loadings of CHBs can be expressed to no better than an order of magnitude. A number of recommendations for improving the state of air-sea exchange of the HCHs were made in Section 2.6.1, and these also apply for CHBs. In addition, reconciling the discrepancies in atmospheric concentration data should be given high priority.

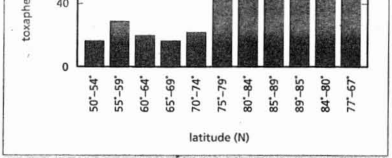


FIGURE 6.5

Measurements of CHBs (pg·L^{-1}) in surface water on the BERPAC-93 (Bering-Chukchi seas) and AOS-94 (Arctic Ocean) cruises.

- *Toxaphene and HCHs in Arctic Ocean water trapped under sea ice will slowly drain through the Canadian Archipelago, on a time scale of decades. Outflow of contaminated water will impact biota in the Archipelago and Baffin Bay.*

2.6.3 PCBs: Local and Long-range Sources

Prior to the mid- to late-1970s, PCBs were a major constituent of dielectric fluids used in transformers, capacitors and other electronic equipment. They were also used in some hydraulic fluids. Therefore, it is to be expected that PCBs were in use in electrical and electronic equipment at arctic radar stations. More importantly, disposal practices during of operation of the stations resulted in PCBs entering the local terrestrial and aquatic environments either through disposal at landfills, down the drain disposal and accidental or intentional spills. As a result, a large, but as yet unquantified portion of the estimated 30 tonnes of PCBs imported to the radar sites may have been released to the environment. Once free in the environment, PCBs are transported regionally and potentially long distances through particulate atmospheric transport and gaseous transport.

A number of radar networks have been constructed in the Canadian north for the purpose of early detection of airborne attacks. Most of these have been US-led or joint activities between the Canadian and US military. Perhaps the best known is the Distant Early Warning (DEW) Line, which consisted of 63 stations across Alaska, Canada and Greenland along the 66th parallel. Over the life of the DEW Line (1955 to 1988), stations were closed and added and communications were improved but radar technologies were not substantially upgraded (Fletcher 1990). The largest change occurred in the Line in 1963 when all 31 gap-filler or "Intermediate" (I) sites were closed, 20 of which were in Canada. Finally, by the 1980s, it was recognized that the radar defenses were no longer adequate for the new series of bombers and cruise missiles and plans were made to replace the DEW Line with the North Warning System (NWS). In 1987, the western half of the DEW Line was closed and replaced by seven new stations at existing DEW Line sites. During

1988, the eastern part of the DEW Line was closed and new stations were constructed at three DEW Line sites and three Gap-Pine sites along the eastern coast of Baffin Island, Quebec and Labrador. A total of 11 new sites were completed in Canada during 1987 and 1988 with eight of these being long-range radar sites. In addition, a large number of un-manned, gap-filler short-range radar stations were constructed between 1990 and 1992 to complete the NWS.

An extensive assessment of the potential for site contamination was undertaken by Environment Canada (Holtz *et al.* 1987). Together, Environment Canada and the Department of National Defense undertook to clean up the sites. This addressed several environmental waste products, including drums of contaminated oils, discarded electronic equipment and contaminated soils and other material. Ongoing environmental assessment and impact studies at the northern radar stations and at arctic reference sites have resulted in the sampling and analysis of over 3500 soil samples (Reimer *et al.* 1991, 1993a, 1993b, 1993c, 1994, Dodd and Reimer 1992, Dushenko and Reimer 1994, Bright *et al.* 1995).

The average total Aroclor PCB concentrations for 525 associated plants and soils collected at various background locations and radar sites indicate that past activities have had a major influence on concentrations of PCBs (see Chapter 3 for more detailed discussion). The highest concentrations are found at radar sites on the east coast where concentrations in soils are the highest reaching 320 000 $\mu g \cdot kg^{-1}$. This is thought to be largely a result of the use of PCB-containing devices at these locations. The absence of proper disposal and maintenance during operation and superficial cleanup at the closure of the site, led to PCB contamination. Thirty years after site closure, concentrations in polluted soils near the "I" sites were as high as 1400 $ng \cdot g^{-1}$ in contrast to 0.90 $ng \cdot g^{-1}$ at remote background sites in the Arctic.

There is ample evidence that localized site contamination has occurred and that contaminated sites require remediation. However, it is important is to assess the relative importance of local sources compares with the deposition of PCBs transported by long-range transport. Gregor and Reimer (1995) compared the PCB profiles for soils, lake sediments and snow. Snow is a major mechanism for the deposition of atmospherically transported contaminants to the Arctic (Gregor and Gummer 1989, Gregor 1991, Gregor *et al.* 1995b) and as such represents a major pathway for PCBs to the terrestrial and aquatic environments. Gregor and Reimer (1995) demonstrated the similarity in the PCB congener pattern of background soil samples and of a Mackenzie Delta lake sediment core sample (Figure 6.7). The background soil congener patterns were similar to those observed in snowpacks at locations about 50 km from DEW Line sites on the Boothia Peninsula. Both were similar to Aroclor 1248. In contrast, the contaminated site samples evaluated from Cambridge Bay and from near Iqaluit are typical of Aroclors 1254 and 1260, respectively. More remote snow samples (Mould Bay and Agassiz Ice Cap) show the effect of preferential transport and scavenging of PCB congeners and are approaching the nature of lower chlorinated Aroclors such as 1242 and 1016 (Figure 6.7). This transition to lower chlorinated PCBs is likely the result of preferential transport over a long distance rather than an indication of the nature of the PCB mixture at the source. The lower abundance of mono- through tri-chlorinated biphenyls in soils is the result of the revolatilization of these compounds from the snow whereas the predominance of tetra- and penta-PCBs is believed to be the result of the preferential adsorption of these compounds to the soil matrix.

The analysis by Gregor and Reimer (1995) suggests that the impact of the DEW Line sites, with respect to the contribution of PCBs to the arctic environment, is limited to the immediate vicinity of the site. This con-

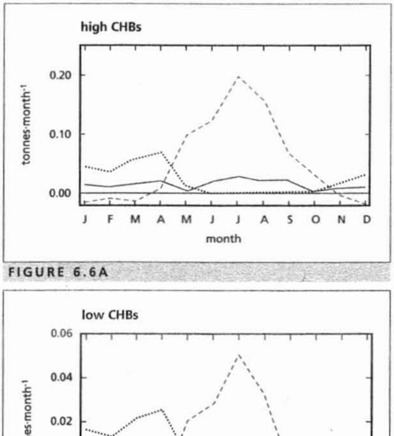


FIGURE 6.6A

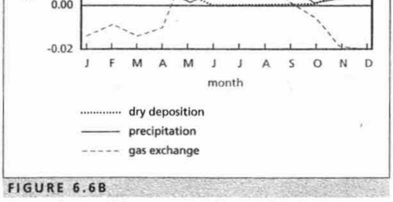


FIGURE 6.6B

The contributions of dry particle deposition, precipitation and net gas exchange to the atmospheric budget for CHBs. Two scenarios are presented that represent the range of analytical certainty of toxaphene measurement: (A) high values adjusted to the NIMS scale, and (B) low values determined by ECD.

clusion is based on a spatial distribution study at Cambridge Bay (Bright *et al.* 1995). The authors concluded that PCBs were found only at a distance of 5 km or less from the site. It was concluded that there is a "halo" of impacted soils around each contaminated site which gradually blends into the background soil PCB signature determined by atmospheric deposition.

The size of the PCB halo around contaminated sites depends on the total mass of PCBs released, the areal extent of contamination, mean particle size of the soil matrix, vegetational cover, exposure to wind erosion and moisture content. Detailed information is not available for each site, but a conservative assumption can be made that the diameter of this halo at any one site is approximately 20 km. This equates to an area of influence of approximately 300 km^2 about each site, or a total land area of about 6300 km^2 for all 21 DEW Line sites in the NWT and the Yukon. This represents less than 0.2% of the total surface area of the Yukon and Northwest Territories.

Based on the snowpack chemistry observations reported in Section 2.3.2 and the snowpack burden estimates in Section 2.4.1 (Figure 4.7), Gregor and Reimer (1995) estimated minimum local PCB deposition of the order of 400 $ng \cdot m^{-2} \cdot a^{-1}$ for areas typical of the DEW Line sites.

Assuming that the ΣPCB concentration for the Cambridge Bay background site at 5 km east of the contaminated site represents the average PCB concentration for an area of 300 km^2 around each DEW Line site, it can be estimated that approximately 18 kg of PCB are contained in the surface 1 cm of soil density of 1 $g \cdot cm^{-3}$. In reality, these would be selectively deposited in areas of fine soils with some organic matter. By comparison, annual long-range transported PCB deposition within this same area would amount to an estimated 0.120 kg. Over the 30 years that PCBs have been deposited in the Arctic from long range atmospheric transport, total deposition can be estimated to be about 3.6 kg or about 20% of the local impact of the DEW Line site contamination. Clearly, the local contamination is considerably more important than atmospheric deposition within the locally impacted area, but the extent of this area is quite limited. The aerial deposition of PCBs on an arctic-wide scale remains dominated by long-range atmospheric deposition.

- *Are the main sources of PCBs in the Arctic local DEW Line dumping or long-range transport? The answer depends on what area is considered. For the areas within a 10 km radius of DEW Line sites, dumped PCBs are the major source (about 85% of total PCB deposition over a 30-year period). In contrast, for the entire Yukon and NWT, dumped PCBs are only 1% of the total deposition over 30 years. The atmosphere is clearly the dominant source.*

2.6.4 Priority Metals — Mercury, Lead and Cadmium: A Problem?

2.6.4.1 Mercury: Natural and Anthropogenic Sources

The human impact on the global Hg cycle is difficult to assess, partly because natural fluxes and inventories are large and variable, and partly because we must rely heavily on deductive methods. Hg, which has both inorganic and organic forms, is biologically significant and geochemically active, making it mobile within environmental reservoirs. Stable isotope patterns, so useful in discriminating Pb sources, provide no help with Hg. Nevertheless, there are several lines of evidence which, taken together, strongly suggest that fluxes of Hg are increasing globally. Cold condensation, biomagnification, and arctic food web structure, the factors that make the Arctic particularly sensitive to globally transported OCs (Chapter 3), also apply in whole or in part to Hg (Macdonald and Brewers 1996).

Atmospheric concentrations of Hg were reported to be increasing in the Northern Hemisphere (at least up until 1990) by 0.6% to 1.5% per year (Langner 1992, Fitzgerald 1995). In total, there has been perhaps a three-fold increase in Hg concentration from pre-industrial levels in air and in the upper ocean (Mason *et al.* 1994, 1995). Unfortunately, there is neither a long enough nor an extensive enough data base to assess recent arctic atmosphere or ocean trends, especially when it is considered that earlier Hg data were compromised by contamination during sampling.

A second line of evidence supporting recent increased Hg fluxes to the Arctic is found in profiles from dated sediment cores (Table 5.2, Lockhart *et al.* 1995). In a study of nine Canadian lakes, plus a site near the eastern shore of Hudson Bay, Hg concentrations were found generally to increase toward the sediment surface. Using the applicable sedimentation rates, modern fluxes were estimated to exceed pre-industrial fluxes by as much as seven-fold (Table 5.2). Although these authors cautioned that the apparent increase in flux for the cores from western lakes may not be significant, (Gobeil and Cossa 1993), they did conclude that eastern lakes in the NWT had been affected by human activities through long-range atmospheric transport and deposition. A similar surface enrichment of Hg has been observed in Alaskan lake cores (Landers *et al.* 1995).

The current global atmospheric burden of Hg, estimated at about 6000 tonnes (Slemr and Langner 1992, Fitzgerald 1989) is about the same as the annual rate of removal (Fitzgerald and Clarkson 1991). Thus, the atmospheric residence time of Hg is about one year. Emissions of Hg to the atmosphere are made up of three roughly equal components: oceanic

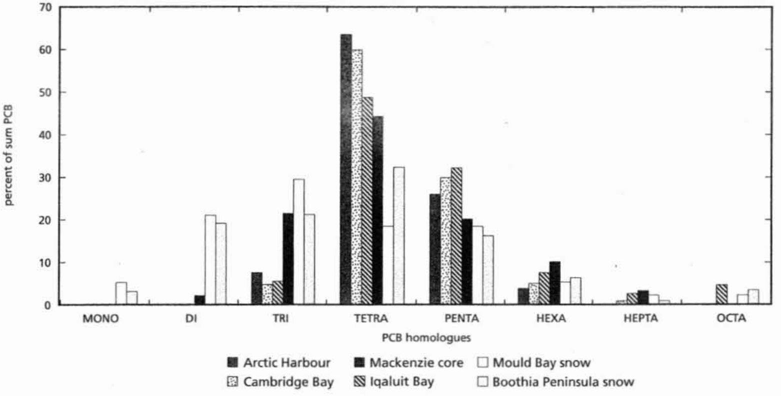


FIGURE 6.7

PCB homologue patterns (expressed as the percent of ΣPCB congeners) for background soil samples from Arctic Harbour, Iqaluit and Cambridge Bay compared with snow samples from Mould Bay (fresh snow) and snowpack samples from the Boothia Peninsula (Lady Melville and Chartrand Lake sites) and to lake sediment cores from the middle Mackenzie Delta (Gregor and Reimer 1995).

degassing, terrestrial natural sources, and anthropogenic releases. It is variously estimated that 50% to 70% of the Hg currently cycling through the atmosphere is due directly or indirectly to human activities (Lindqvist *et al.* 1991, Mason *et al.* 1995). The most significant anthropogenic sources of atmospheric Hg are from the combustion of coal and urban refuse (Nriagu and Pacyna 1988). Despite the uncertainty in the individual components of the global Hg budget, estimated at about a factor of two, (Porcella 1994), it is clear that human activities contribute a significant, if not a dominant, portion of the Hg currently cycling in the global environments.

The evidence for increased fluxes and burdens of Hg in atmosphere and oceans (mixed layer) outlined above has clear implications for both terrestrial and marine biota, since Hg is known to biomagnify (Guthrie *et al.* 1979). Hence, recent data showing an increasing Hg trend in biological samples, (Wagemann *et al.* 1996, Hansen *et al.* 1991) may reflect a coupling between arctic biota and the human impact on the global Hg cycle. Although increasing trends in Hg are clearly a priority concern, the present data base for Hg in the Arctic atmosphere, ocean, ice and sediment is too limited to make a proper assessment.

2.6.4.2 Pb: Evidence of Contaminant Controls that Work

Lead in the environment can originate from natural or anthropogenic sources. It is found in the atmospheric aerosol (Barrie and Barrie 1990) and in glacial ice (Boutron *et al.* 1995) to be highly enriched relative to crustal elements such as Al, Fe or Ti as well as relative to sea salt elements such as Na. There are no known process of fractionation of soil or sea salt elements as they enter the atmosphere that will explain the atmospheric enrichments of Pb. There is no doubt that the mobilization of Pb by anthropogenic activities has changed the atmospheric environment. Recognizing that Pb additives in gasoline constitute a hazard to human health, regulatory agencies around the world enacted changes in the use of Pb in gasoline. In North America, Pb in gasoline was completely removed by the late 1970s and early 1980s and air concentrations have declined quickly in response to these controls.

Eurasian countries have been slower to react to health concerns but nevertheless reductions in lead in gasoline have occurred. These are manifested in a slow but steady decrease in airborne lead in Europe. For instance, measurements in southern Norway indicate that between 1978/79 and 1985/86, Pb concentrations decreased by 30% and 43% in air coming from eastern and western Europe, respectively (Amundsen *et al.* 1992). At Alert, from 1980 to 1993 in the winter months, Pb has decreased by a factor of 2 reflecting the decreased use of lead additives in automotive fuel (Figure 5.2). From chemical transport modelling (Akeredolu *et al.* 1994) and lead isotopic studies, it is known that aerosol Pb at Alert is mainly from Eurasian sources: equally split between eastern Europe, western Europe and the former Soviet Union. In contrast, isotopic studies at in Greenland

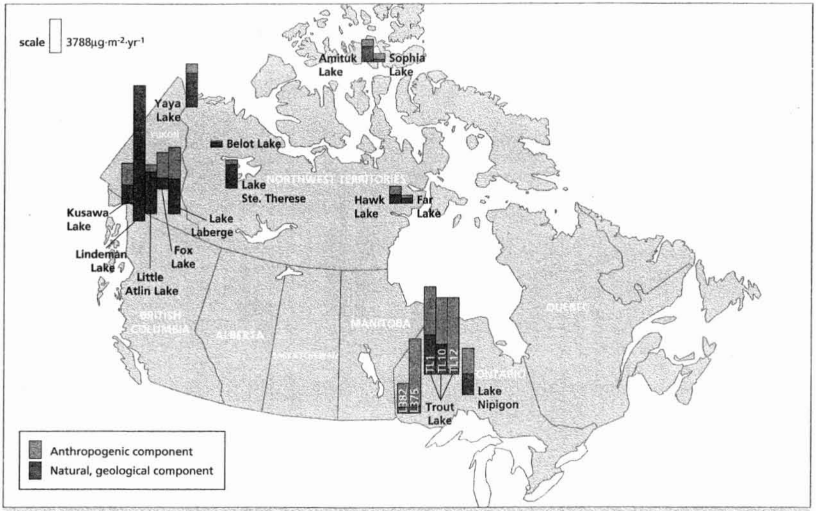


FIGURE 6.8

Current fluxes of lead to some Canadian lakes from natural and anthropogenic sources. (Lockhart et al. *1996).*

(Boutron *et al*. 1991, Boutron *et al*. 1995) clearly showed that prior to Pb emissions control in North America, middle tropospheric air reaching the elevated Greenland plateau (2.3 km high), further south than Alert (i.e., 82.5°N vs. <75°N), received substantial fractions of its Pb from North American sources. There are also trends observed in freshwater lake sediments that, when corrected for a natural geochemical background Pb concentration, confirm that Pb has increased since the pre-industrial era to the present (Figs. 6.8 and 6.9). However, the low temporal resolution of these sediments does not yet show a decrease in anthropogenic lead in the past 25 years.

In summary, trends in Pb concentrations in arctic air, glaciers and lake water sediment reflect trends in decreasing anthropogenic emissions of lead to the atmosphere. Pb emission controls are an example of controls that worked.

2.6.4.3 Cadmium: Is Long-range transport of Cadmium Important?

The organs of marine mammals from the Canadian Arctic are often found to contain high Cd concentrations (see Chapter 3). These high concentrations imply that even with modest portions of these organs in the diet, it is possible to exceed human consumption guidelines (450 mg·week^{-1}, Health Canada). Similarly, Hansen *et al*. (1991) suggest that in sealing districts of Greenland, maximum Cd content in food as recommended by the World Health Organization (WHO) is being exceeded. The detection of high concentrations of Cd in marine mammal organs, however, does not necessarily imply anthropogenic contamination — it may simply reflect a high natural background or geochemical mobility of the element. Regional differences in the Cd burdens in marine mammals observed in the Canadian Arctic (Wagemann *et al*. 1994) implies strongly that metal concentrations in tissues does in some way depend on regional geology or biogeochemistry. The question, therefore, is whether or not anthropogenic loadings of Cd are having a significant impact on Cd burdens in marine mammals.

As discussed in Section 2.3.3, it is difficult to discriminate between contaminant and natural fluxes because Cd is so biologically and geochemically active. Data from marine sediment cores are unlikely to help determine whether fluxes to sediments have recently increased (see Figure 3.18), and oceanic measurements to determine distributions or trends are confounded by bio-uptake/regeneration cycles (de Barr et al. 1994). Stable isotope composition, which has proved so useful for discriminating sources of contaminant Pb (Figure 3.19, Sturges *et al*. 1993, Gobeil *et al*. 1995), provides no help for Cd. Therefore, our only approach is to examine the regional budget for

Cd to estimate at least the scale of the contaminant fluxes compared to the natural ones.

To calculate the sources and sinks for Cd in the Arctic Ocean we start with Table 2.1, the water budget. Table 6.6 presents the results of the calculations discussed below. Cd concentrations in the various water and ice masses have been chosen from the literature (see References column in Table 6.6). In particular, we note a previous budget by Yeats and Westerlund (1991); we have updated this budget using their approach but revising the Cd concentrations and water flows according to recent data. There are three main points of difference between the budget presented here and the earlier budget. First, we have incorporated an atmospheric flux of Cd to the Arctic Ocean (27 $t \cdot a^{-1}$). This flux, which was considered as insignificant in the earlier budget, is an important identifiable pathway for contaminant Cd and, therefore, the focus of this Section. Second, we believe that uptake in authigenic phases in suboxic sediments is an important sink for Cd. We have attempted to estimate this flux using values reported for other coastal areas (5 $ng \cdot cm^{-2} \cdot a^{-1}$, Gobeil *et al.* (1987) and van Geen *et al.* (1995)) together with an assumed area for the Arctic Ocean of 10×10^6 km^2. Third, we have estimated the flux of Cd via ice exported through Fram Strait. The measurement of Cd in ice made by Campbell and Yeats (1982) of 0.3 $mg \cdot L^{-1}$ seems too high (see for example snow values by Mart (1983) and Mart *et al.* (1983)) and would imply a Cd export via ice of 1500 $t \cdot a^{-1}$. Therefore, we have arbitrarily chosen double the average surface ocean concentrations (8.7 $ng \cdot L^{-1}$) given in Mart *et al.* (1983). Clearly, a broader data set for Cd in ice and snow would be of value.

In the budget presented here (Table 6.6), Cd entering the Arctic Ocean via ocean currents has not been differentiated into natural and anthropogenic (contaminant) portions. Estimates put the anthropogenically-derived Cd influx to global aquatic ecosystems at from 2 to 17×10^6 $kg \cdot a^{-1}$ (Simpson 1981, Nriagu and Pacyna 1988, Pacyna *et al.* 1995) with a median value of about 7×10^6 $kg \cdot a^{-1}$ to the ocean. In comparison, the estimated natural flux of Cd to the ocean is $3.4 - 4.3 \times 10^6$ $kg \cdot a^{-1}$ in dissolved plus exchangeable particulate form plus another 1.8×10^6 $kg \cdot a^{-1}$ in detrital particulate form (Yeats and Bewers 1987). Therefore, presently about half the cadmium flux to the ocean could be anthropogenic. Despite the significant increase in Cd emissions during this century (Shen *et al.* 1987), there is as yet no evidence of an altered relationship between Cd and phosphate that would indicate a significant cumulative influence of contaminant Cd to the marine environment. This is partly because cadmium is rapidly recycled in surface ocean waters by biological activity (Boyle *et al.* 1976, 1981, de Baar *et al.* 1994), and partly because contaminant Cd is entering a large natural pool as implied

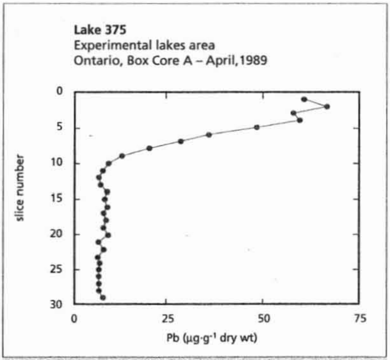


FIGURE 6.9A

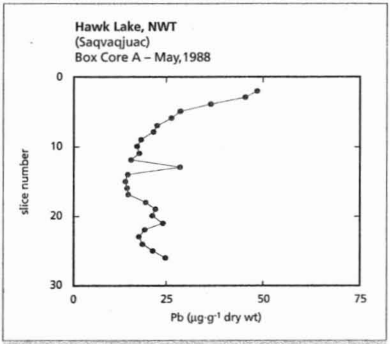


FIGURE 6.9B

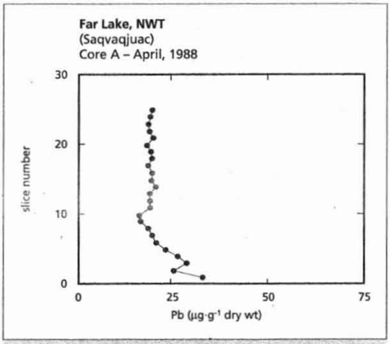


FIGURE 6.9C

Profiles of Pb in freshwater lake sediments measured by Lockhart et al. *(1996). See Figure 5.5 for site locations.*

by the comparatively long residence time of oceanic Cd (ca. 50 000 years, Boyle *et al.* 1976). This view is supported by the lack of an "industrial" signal at remote ocean sites (e.g., corals near the Galapagos, Shen *et al.* 1987) accepting that where direct, long-range atmospheric transport from industrial sources can be inferred, there may be a concurrent regional signal in the surface ocean (e.g., corals near Bermuda, Shen *et al.* 1987). In summary, ocean currents entering the Arctic Ocean probably contain a very small proportion of contaminant Cd, presently obscured by variability driven predominantly by natural cycling. It should be noted that the budget in Table 6.6 is well balanced considering the uncertainties in the calculations. Previous calculations of the atmospheric flux, which we will assume here to be equivalent to the long-range contaminant flux, agree well, and especially so if prorated to a common deposition area (the Arctic Ocean). Furthermore, the relatively low atmospheric flux in Table 6.6 is corroborated by estimates of Cd flux from the Greenland Ice cap (2–5 pg·g^{-1} Cd [Wolff and Peel 1985, Boutron *et al.* 1995] multiplied by 21.5 g·cm^{-2}·a^{-1} snow accumulation [Rosman *et al.* 1994] multiplied by 10 x 106 km^2 gives 4.3–11 t·a^{-1}).

Table 6.6 shows the atmospheric contaminant Cd flux to be less than 1% of either the Cd fluxes into or out of the Arctic Ocean and only about 5% of the estimated flux of Cd to sediments. Clearly, the long-range, large-scale Cd flux through the atmosphere from human activities will be difficult to detect and is unlikely to have much impact on the natural cycles. Problems arising from human inputs of Cd are, therefore, more likely to be due to local inputs (e.g., mining or smelting), not long-range inputs. For Cd, therefore, there is no compelling evidence of serious compromise of arctic marine biological resources by long-range anthropogenic introductions. Despite this conclusion, it must be emphasized that Cd is of concern because it is observed at high concentrations in the organs of some animals (i.e., it bioaccumulates, Ray 1984) and it is easily mobilized from marine sediments through small changes in oxygen content (redox conditions) or acidity (Gobeil *et al.* 1987). Our conclusion that the anthropogenic loading of Cd is not presently of concern rests on the relatively low contaminant versus natural flux and the fact that Cd does not tend to biomagnify (Guthrie *et al.* 1979). However, not enough is known about the transfer of Cd into the food chain to predict with confidence how the system responds to incremental increases in the atmospheric flux resulting from anthropogenic activities.

- *In the Arctic, Pb pollution is very evident in air, glacial ice cores, sediments and ocean water. However, global emissions to the atmosphere have decreased greatly since the 1970s. In glacial ice cores and in the atmosphere at Alert from 1980 to 1995, there is evidence for decreasing atmospheric inputs to the Arctic This is clearly a shining example of pollution controls that work.*
- *Is the source of mercury in the Arctic natural or anthropogenic? There is tentative observational evidence indicating that that there has been a three-fold increase in Hg concentrations in air and in the upper ocean since pre-industrial times. Furthermore, strong evidence of post-industrial increases in Hg flux to the Arctic is found in freshwater lake sediment cores. In a study of nine Canadian lakes plus a site near the eastern shore of Hudson Bay, Hg concentrations were found generally to increase toward the sediment surface. Best global budget estimates of Hg sources to the atmosphere indicate three roughly equal components: oceanic degassing, natural terrestrial sources, and anthropogenic*

TABLE 6.6

Cadmium budget for the Arctic Ocean. It should be emphasized that the uncertainties given in the table are simply best estimates. These have been calculated by choosing Cd concentrations and ± ranges to bracket the available data in the references. The flux was then calculated by multiplying the median Cd concentration by the appropriate volumetric flow (Table 2.1). The uncertainties of the fluxes were estimated from the Cd uncertainty and the volumetric flow uncertainty (Table 2.1) using the propagation of error formula for AxB. The uncertainty of the totals given at the bottom of the table is simply the square root of sum of squares of uncertainties. The Cd flux into sediments is estimated from pore-water data for temperate coastal oceans. References are for studies reporting Cd concentration observations or estimates of Cd fluxes.

Import/Export	In (t·a^{-1})	Out (t·a^{-1})	References
Runoff	23±15	—	Dai and Martin (1995), Martin *et al.* (1993), Rahn (1981), Yeats and Westerlund (1991)
Water through Bering Strait	2300±1000	—	Yeats and Westerlund (1991) and references therein.
Water through the Archipelago	—	2140±1200	Campbell and Yeats (1982), Seakem (1991), Thomas *et al.* (1983)
East Greenland Current (<0°C)	—	1100±600	Moore (1983)
East Greenland Current (AIS)	—	1130±600	Moore (1983)
Norwegian Coastal Current	485±250	—	Danielsson *et al.* (1985)
Barents Sea from Greenland	760±400	—	Danielsson *et al.* (1985)
West Spitzbergen Current	1260±650	—	Moore (1983), Danielsson *et al.* (1985)
Sedimentation	—	500±250	Gobeil *et al.* (1987), Rosenthal *et al.* (1995), van Geen *et al.* (1995)
Atmospheric Transport	27±9	—	Rahn (1981), Jickells *et al.* (1990), Akeredolu *et al.* (1994)
Total	**4855±1300**	**4960±1500**	

releases. The present observational database for Hg in the arctic atmosphere, ocean, ice and sediments is too limited to make a proper assessment of impacts of anthropogenic Hg.

- *Is long-range transport an important source of anthropogenic Cd to the Arctic? The answer differs for the Arctic Ocean and the Arctic terrestrial environment. A Cd budget for the surface Arctic Ocean shows that atmospheric inputs of anthropogenic Cd are less than 1% of the ocean current inputs of natural cadmium. Thus the answer is a resounding "no". In the terrestrial environment, the deposition of anthropogenic atmospheric Cd onto vegetation may be sufficiently isolated from the huge soil reservoir of Cd that it can be selectively grazed and enter the caribou-wolf food chain. However, evidence for this in measurements of Cd in caribou is not strong. Thus, the answer is "maybe".*

2.6.5 Radionuclides: Local and Long-range Sources

Studies have been initiated by Canada (and other countries) to measure the fluxes of radioactive contaminants along oceanic pathways to evaluate the environmental threat to North American arctic seas. Recent reports of the dumping of radioactive wastes by the former Soviet Union in the Barents and Kara Seas (Yablokov *et al.* 1993, OTA 1995) have heightened concern regarding the potential for radioactive contamination of the Arctic Ocean.

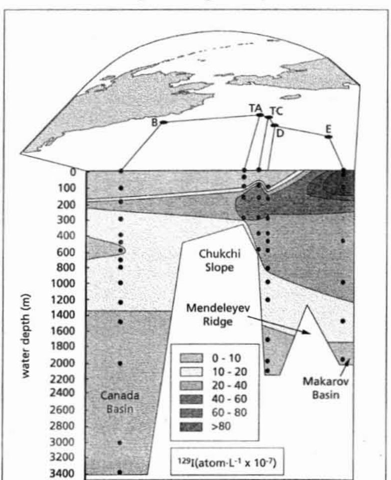


FIGURE 6.10

Section from the CSS Larsen cruise in 1993 (view is southerly towards Alaska and Northern Russia) shows the transport of ^{129}I labelled Atlantic water at depths of 100-200 m from the Makarov Basin (Station E) into the Canada Basin (Station B). ^{129}I concentrations (<10^8 atoms L^{-1}) derive from European reprocessing plants. Note that the bottom contour shows only the disposition of stations along the section and not the bottom topography of the basins and is not meant to imply a sill at stations TC or TA.

2.6.5.1 Long-range Marine Transport to the Arctic not a threat

In addition to Russian disposal, important sources of anthropogenic radionuclides to the Arctic Ocean are the European nuclear fuel re-processing plants, Sellafield, Dounreay, and Cap de la Hague, located in western England, Scotland and northern France. Sellafield is historically and currently the most important among these. Since the 1960s, large quantities of radioactive waste, especially the isotopes ^{137}Cs and ^{129}I, have been released by these plants directly into the ocean (Figure 4.9. Maximum releases from Sellafield occurred in about 1975 when approximately 5 PBq were discharged to the Irish Sea (Gray *et al.* 1995) and, to date, a total of about 10–15 PBq of radiocesium has been discharged to the sea (Aarkrog 1993). Part of this radioactivity is subsequently transported by the Norwegian Coastal Current into the Arctic Ocean, either through Fram Strait (between Greenland and Spitzbergen) or through the Barents Sea within 5 to 7 years from its release (Figure 2.3).

Until recently, observations of the movement of radio-tracers through the Arctic Ocean were limited to a few ice stations such as LOREX (1979), FRAM 3 (1981), CESAR (1983), AIWEX (1985) and the Canadian Ice Island (1985/1989) (Livingston *et al.* 1984, Moore and Smith 1986, Smith *et al.* 1990, Smith and Ellis 1995). However, with the recent initiation of icebreaker and submarine cruises into the central Arctic Ocean it has become possible to produce a synoptic view of the radiotracer field. Samples collected from the CCGS Henry Larsen in 1993 provided evidence of the arrival of radionuclides from European re-processing plants in surface and Atlantic layer water in the Makarov Basin, overlying the Mendeleyev Ridge. The interface between water of Atlantic origin (having elevated ^{137}Cs and ^{129}I concentrations indicative of the re-processing plan source) and Pacific origin (having radionuclide concentrations indicative of a fallout source) was clearly delineated (Figure 6.10). Trace quantities of re-processing radionuclides were evident at depths as great as 2000 m in the Canada Basin.

The Arctic Ocean section in 1994 permitted the sampling and detection of radionuclides on a complete transect of all the major arctic basins (solid line and circles in Figure 6. 11). Between 1979 and 1994, the ^{137}Cs concentration at the North Pole increased by a factor of three due mainly to the advective inputs of re-processing ^{137}Cs (Figure 6.12). From

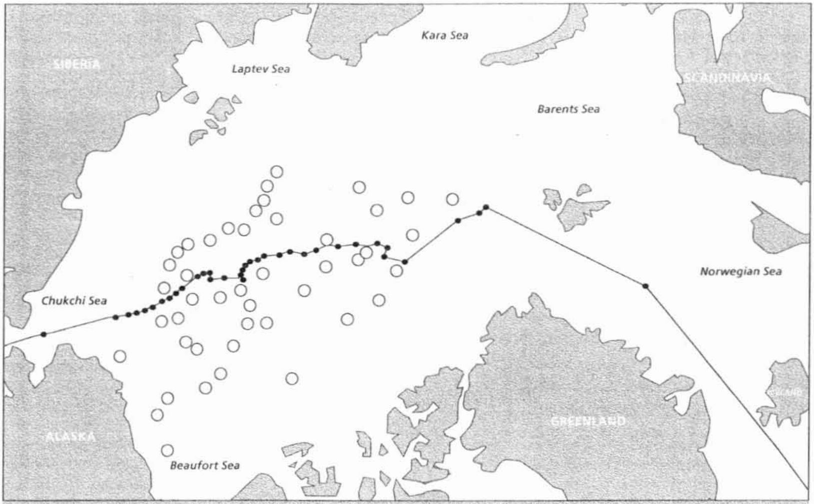


FIGURE 6.11

Trackline and station locations for the Arctic Ocean Section 1994 (solid circles) and stations for the USS Cavalla cruise 1995 (open circles).

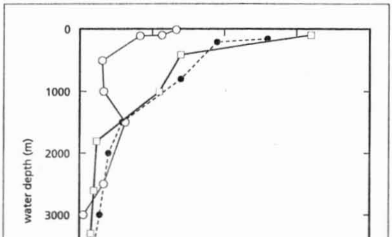


FIGURE 6.12

^{137}Cs water column profiles measured at the North Pole in 1991 and 1994 illustrate the increased inputs of ^{137}Cs (Bq·m^{-3}) to the Arctic Ocean since 1979 from the Sellafield Reprocessing Plant and Chernobyl accident. (LOREX data are from Livingston et al. *1984, Oden data are from Roos and Holm 1993, AOS-94 data are from Smith (unpubl. data).)*

the transect data and the broader spatial coverage now available from submarine cruises (USS Cavalla in 1995) it is becoming clear that a portion of any conservative or quasi-conservative contaminant released to the European or Russian arctic coasts will be transported to the waters of the Canada Basin, Beaufort and Chukchi Seas within as little as a decade (Figure 3.16 for the transport pathways). The pathways that could potentially deliver the Russian radionuclide discharges to Canadian waters have been delineated in considerable detail by the re-processing radionuclides from Europe. There is, as yet, little evidence of radioactivity originating from the Russian shelves entering North American waters or sediments (Baskaran and Naidu 1995).

Despite the ease in detecting the re-processing radionuclide signal, these concentrations are significantly lower than those that would constitute a radiological threat to organisms or humans in North American coastal regimes. Two other factors mitigate the risk from the re-processing radionuclides. First, the release of radionuclides of radiological concern from the European sources has now been reduced by as much as two orders of magnitude since the mid-1970 maximum (Figure 4.9) and, second, a large portion of the signal is contained in deep water within the Atlantic layer (200–800 m), well away from the surface where most of the biota are found.

FIGURE 6.13

Samples collected in Chernaya Bay and at the barge site in the Novaya Zemlya trough during a joint Canadian-Russian expedition in 1992–93. Surface sediments in the Barents and Kara seas have elevated ^{239,234}Pu in the region of Chernaya Bay and elevated ^{137}Cs and ^{60}Co in the Yenesey River estuary.

2.6.5.2 Long-range Atmospheric Transport

The principal atmospheric sources of radioactivity to the Arctic Ocean have been fallout from atmospheric nuclear weapons tests, conducted mainly during the 1950s and 1960s (Figure 4.9) and from the 1986 Chernobyl accident. The total global fallout into the Arctic Ocean up to 1993 has been estimated at 9 PBq ^{90}Sr and 15 PBq ^{137}Cs (Aarkrog 1993). With the Partial Nuclear Test Ban Treaty in 1963, this fallout signal has dropped precipitously (Figure 4.9). An additional regional fallout of 20 PBq ^{90}S and 30 PBq ^{137}Cs is estimated to have been deposited in the Arctic from the 87 test explosions carried out by the Russians at Novaya Zemlya (Aarkrog 1993). All of the Soviet Union's very large explosions took place at Novaya Zemlya. Between September 1955 and October 1990, 132 tests were carried out; 87 in the atmosphere, 3 underwater, and 42 underground. It is estimated that

94% of the total yield of all Soviet nuclear tests was released at Novaya Zemlya. Remnants of the global fallout can be detected throughout the world ocean surface waters; however, as was the case for the reprocessing signal, the fallout concentrations are considerably lower than those that would produce a significant radiological risk. Fallout has now been effectively reduced by about 2 orders of magnitude from its peak in the 1960s, and therefore is not currently of concern.

2.6.5.3 Local Sources

Potential sources of radioactive contamination from the Russian shelves include:

i) liquid and solid wastes dumped in the Barents Sea,
ii) fuelled nuclear reactors, originally used to power submarines or ice-breakers, but subsequently dumped at sea,
iii) local and regional fallout from atmospheric and underground nuclear weapons tests conducted on the island of Novaya Zemlya,
iv) upstream nuclear industry sources transported down the Ob and Yenesey Rivers to the Kara Sea and,
v) wastes discharged into arctic coastal waters from surface sources associated with the Russian civilian and military nuclear operations based in predominantly in Murmansk (Yablokov 1993, Aarkrog 1993).

An additional source of radioactivity is two underwater nuclear weapons tests during the 1950s, which occurred in Chernaya Bay, a small fjord on the southern coast of Novaya Zemlya (Figure 6.13). A joint Canadian-Russian cruise to this fiord in 1993 revealed plutonium concentrations from fallout debris in Chernaya Bay sediments to be as high as 15 000 $Bq \cdot kg^{-1}$, which ranks among the highest concentrations recorded at any contaminated marine location in the world. The transport of plutonium appears to have occurred over distances of the order of 100 km from Chernaya Bay into the Barents Sea.

Movement of radioactivity in dissolved phases from the above sources into the Arctic Ocean can occur by:

i) advection and mixing of surface water across the Russian continental shelves which takes of the order of years (e.g., Schlosser *et al.* 1994),
ii) advection and mixing within the Atlantic water as it crosses the Barents Sea and circulates through the various basins on time scales in the order of 30 years, and
iii) transport into the halocline layer which is partly maintained by the production of dense water by ice formation over the Russian continental shelves (Schlosser *et al.* 1990).

To date, no signal from such sources has been detected in the basin sections.

Particle-associated radionuclides (e.g., plutonium) which become captured in shelf sediments can later be incorporated in ice through suspension freezing or when ice becomes grounded (see Section 2.3.3). Through the melt/freeze cycle, this contaminated sediment will end up within a few years at the surface of the ice, where it could then come in contact with biota. Subsequent melting of the contaminated ice in marginal ice zones would later release the radioactive contaminants into the surface water at locations and times important for biological productivity. The ice drift patterns shown in Figure 3.15 suggest that the most likely pathway for such Russian shelf sediment contaminants is into the Barents Sea or, via Fram Strait, into the Greenland Sea, rather than the North American marginal arctic seas. Nevertheless, elevated concentrations of ^{137}Cs have been detected in particulate material in ice collected in the Canada Basin and slightly enhanced inventories of radionuclides in underlying sediments confirms that some amount of radioactive material is being transported into this region by ice-rafting. Ice back-trajectories suggest that it is improbable that the source of this material is the Russian shelves.

The main risk from radioactive waste disposed on the Russian shelves is from sealed containers that have been dumped or lost in the Kara Sea. The residual radioactivity will inevitably be released as the containment deteriorates, but how long this might take is not known. A sunken vessel reportedly loaded with 0.004 PBq of radioactive wastes (Yablokov *et al.* 1993) was located in the Novaya Zemlya Trough during the joint Canadian-Russian cruise. Measurements of radioactivity in samples of sediment and seawater collected at precise, proximal sites show only minimal, if any, releases of radioactive contaminants up to 1993. It can be deduced from the known inventory of radioactivity in the various contained sources that threats may be posed on local and regional scales, but it is not likely that there will be a significant threat to the general Arctic environment.

In 1968, a B52 aircraft carrying nuclear weapons crashed onto the ice 11 km west of Thule, Greenland. Between 2.8 and 4.2 kg of plutonium escaped onto the wreckage into the surrounding areas, which left an estimated 0.2–0.6 kg lost to the marine environment after recovery operations (Aarkrog 1995). Subsequent surveys show this plutonium distributed in local sediments where it is subject to downward mixing and burial but not further transport (Smith *et al.* 1994).

Lastly, there remain contained sources presently operating within the Arctic including Russian nuclear power plants, re-processing plants, nuclear powered vessels and various nuclear waste storage sites. These sources present a potential, large-scale environmental threat which, in the event of a catastrophic release similar to the Chernobyl nuclear reactor accident, could pose a significant future radiological contamination in the Canadian Arctic.

- *Do local and long-range sources of radionuclides pose a risk to the Arctic? Long-range transport of radionuclides into the Arctic by oceans is not significant. Uranium processing plant effluent that was released into European waters in the past from the UK and France is now radiologically inactive (^{137}Cs, ^{129}I), thereby having more benefit (as an ocean circulation tracer) than risk. Long-range transport of anthropogenic radionuclides through the atmosphere to the Arctic is negligible. The main source of locally dumped radioactive waste is from the Russian continental shelves, particularly in the Kara and Barents seas. Radioactivity will inevitably be released as containment of waste deteriorates, but how long this might take is not known. Studies show that presently there is no indication of contamination. If leakage occurs, threats may be posed on local and regional scales, but it is not likely that there will be a significant threat to the general arctic environment. Ocean and ice currents do not favour the movement of waste to the North American Arctic.*

2.6.6 New Generation Pesticides in the Arctic Environment

Many of the OCs discussed in this report were registered pesticides with widespread, high-volume use in southern regions that have now been greatly curtailed or eliminated in many parts of the world. They have been replaced, in some cases, with other classes of pesticides. Newer pesticides based on formulations of nitrogen and phosphorus are currently used in high volumes. These substituted "agro-chemicals," while generally more readily degradable than the OCs, still have properties that take them to areas remote from their place of usage via mechanisms similar to those operating for OCs. The persistence of new-generation pesticides in the Arctic and en route may be sufficient to cause concern over their exposure to biota but there is little information on their degradation in cold conditions.

Lartiges and Garrigues (1995) observed half-lives for 16 pesticides (13 organophosphorus) in unfiltered river water in France averaging 14 d (±12 d) while for seawater under natural conditions it was 34 d (±44 d) (Table 6.7). With a drop in temperature from 22°C to 6°C, they observed a 20-fold increase in half-life for riverwater and a 6-fold increase for seawater (some of the pesticide half-lives could not be estimated since there was "no observed degradation"). The high usage triazines — atrazine and cyprazine were among the most persistent pesticides tested.

Two studies attempted to measure these compounds using the relatively new solid phase microextraction (SPME) analytical technique. Gregor *et al.* (1995a) on the Agassiz Ice Cap and Boyd-Boland *et al.* (1995) in the Russian Arctic applied SPME to surface snow and snowpack. Their data indicate that a number of organo-phosphorus and nitrogen pesticides may be reaching the Arctic in significant quantities. The SPME is still in the development stage, however, and further work is needed.

Actual measurements of non-chlorinated pesticides in the Canadian Arctic are limited. As part of the Slave River study, selected new-generation pesticides were measured in water to determine if there was significant transport from agricultural use areas upstream (i.e., to the south) in the drainage basin (McCarthy *et al.* 1997). These included the phenoxy herbicides 2,4-dichlorophenoxy acetic acid (2,4-D) 2,4,5-trichlorophenoxy acetic acid (2,4,5-T) and methylchlorophenoxy acetic acid (MCPA). Other herbicides measured in water using SPME were trifluralin (Treflan), a relatively volatile dinitroaniline used in the control of grasses and broadleaf weeds in North America and the chlorothiocarbamates such as triallate, which are applied on field crops such as peas. Carbamate pesticides are extensively used globally because of their pest specificity and low mammalian toxicity. None of these pesticides were identified in water above the reported analytical detection limits. Trifluralin has a high organic carbon-to-water partition coefficient (K_{oc}) which correlates well with the octanol-water partition coefficient (K_{ow}) (Muir 1991) indicating ready adsorption to sediment or partitioning into lipid membranes of aquatic organisms. Neither trifluralin or triallate were measured in the sediment from the Slave River

TABLE 6.7

Half-lives (days) of selected pesticides compared at different temperatures.

Pesticide	High Temperature	Low Temperature
atrazine*	200	NOD[1]
azinphos-methyl*	26	NOD
chlorpyrifos	4	27
cyprazine	184	260
diazinon	50	125
diazinon*	14	45
dimethoate*	36	219
dimethoate	34	142
ethion	34	84
fenitrothion*	34	224
fensulfothion	18	82
isofenfos*	118	224
malathion*	6	41
methadithion	20	139
mevinphos	19	150
ethyl parathion*	44	542
methyl parathion*	30	233
phosmet	2	4
terbufos	2	34

[1] *NOD = no observed degradation after sixty days*

* *Half lives for compounds noted with an asterisk are from Lartiges and Garrigues (1995) and are measured at 22°C and 6°C. Half lives for compounds without an asterisk are from Frank* et al. *(1991) and were measured at 21°C and 4°C.*

above the analytical detection limit that ranged from 5 to 50 µg·kg^{-1}.

In a 1993 (August–September) study of new generation pesticides in the Arctic and near-Arctic region, Chernyak *et al.* (1996) found low but measurable levels in several media. Chlorpyrifos was found in surface water samples (4 of 5) at a mean concentration of 38 pg·L^{-1} (detection limit 0.8 pg·L^{-1}) in the Chukchi Sea/Chirikov Basin up to the ice edge at circa 74°N. Also observed in 2 of the same five samples, was endosulfan I (the α-isomer) at 5 pg·L^{-1} which was only twice its detection level, however. In surface microlayer samples (stainless steel screen), from the same locations, only the sample from the westernmost site (near Wrangel Island) showed any of the target analytes at concentrations above detection; levels (and detection) of chlorothalonil and trifluralin were 220 (0.4) and 1100 (0.5) pg·L^{-1}. Chlorthalonil was also found in all 3 fog samples from the same area (7000 pg·L^{-1} mean) as was chlorpyrofos at an average 2000 pg·L^{-1}. Two of the three fog samples had metaloclor (147 000 and 2000 pg·L^{-1}) and one had terbufos (12 000 pg·L^{-1}). Air samples were taken while the expedition ship was in transit; the only one available in the Chukchi Sea had no detectable levels of any of the analytes except a near-detection limit 0.5 pg·m^{-3} of endosulfan II. Bidleman *et al.* (1995b) found endosulfan I at approximately 4 pg·m^{-3} further east at Resolute Bay, NWT in 1992.

Other samples from the Bering Sea and Gulf of Anadyr (Chernyak *et al.* 1996) had detectable levels of chlorpyrifos, chlorthalonil, endosulfan I, metalochlor, trifluralin and atrazine in one or more of the media at approximately the same frequency as found in the more northerly samples. Analytes sought in both areas but not observed were: fenvalerate (I and II), butylate, vernolate, simazine, diazinon, malathion, alachlor, methyl parathion and cyanazine. The significance of these observations is uncertain. Chernyak *et al.* (1996) mention that chlorpyrofos, chlorthalonil, atrazine and diazinon have some small use in Alaska and endosulfan has been observed elsewhere in the Arctic (Bidleman *et al.* 1995a, Fellin *et al.* 1995, Figures 3.1 and 3.2). The compounds are unlikely to bioaccumulate in any arctic species but the sub-acute effects of continuous exposure to the levels indicated are largely unknown particularly for northern species. What may be of value is the observation that even these "readily biodegradable" compounds can survive long enough to be transported to the Arctic where their chances of persistence are so much greater. A need exists to more accurately define the extent of this possible problem.

2.6.7 PAHs: Anthropogenic and Natural Sources

Because PAHs include such a large number of compounds (Figure 1.3), they are traditionally presented in assessments as the sum of the concentrations of a small, toxicologically significant, subset of 17 "priority" PAHs. This "sum of PAHs" is often used to evaluate the potential for biological effects, but it contains virtually no other information and, indeed, it does not even provide a sufficient basis to evaluate toxicity because it ignores alkyl PAHs (Payne *et al.* 1988). If the goal is to assess the potential for impacts, the PAHs derived from human activities must always be evaluated against a varying natural background (Yunker and Macdonald 1995). Knowing the origin of PAHs is important for another reason: hydrocarbons from petroleum and combustion sources can differ in their resistance to bacterial degradation (Jones *et al.* 1986), likely have different particle size associations (Yunker *et al.* 1994) and may differ in their bioavailability (see Payne *et al.* 1988 for a recent discussion). Fortunately, from the point of view of assigning source, combustion, petroleum, and natural sources have different PAH compositions. However, extracting information on sources from the PAH data is usually not trivial because there is considerable overlap in the PAH composition between the sources.

Within the marine environment of the Canadian Arctic, the Mackenzie shelf of the Beaufort Sea provides the best hydrocarbon data base from which to assess sources of PAHs (see Section 2.3.3). PAH concentrations in Mackenzie shelf sediments are found to be significantly higher than would be normal for a pristine area. The immediate conclusion that contamination has occurred is not supported by a full assessment of the data. Our first clue about PAH sources is that sediment and suspended sediment from this shelf generally contain only low proportions of the linear PAHs like anthracene or PAHs containing five-membered rings (e.g., indeno (1,2,3-cd)pyrene) (Figure 1.3). These less stable (kinetic) isomers, which tend to be prevalent in combustion byproducts, are simply not major constituents in either Mackenzie River suspended particulates or shelf sediments. This rules out significant contributions from combustion PAHs (Laflamme and Hites 1978, Grimmer *et al.* 1983). In suspended sediments from surface water and sediments from locations near the shelf edge, there is some indication of the presence of combustion PAHs (Yunker *et al.* 1993, 1994). Hence, combustion inputs appear to be small everywhere, but are more likely to be detected further offshore, away from the dominating influence of the Mackenzie River.

In the environment, parent PAHs (PAHs with unsubstituted rings) are usually accompanied by a series of alkyl-substituted homologues. For example, alkyl-substituted derivatives of the parent PAH phenanthrene include compounds like pimanthrene (C_2, or two carbon substituents) and retene (C_4) (see Figure 1.3). Our second clue about PAH sources for the Mackenzie Shelf is that the observed alkyl homologue distributions maximize at C_1 or higher for the naphthalenes, phenanthrene/anthracenes and the fluoranthene/pyrenes. Hence a significant portion of the lower molecular weight parent PAH fraction in Mackenzie Shelf sediments is petrogenic material (Shaw *et al.* 1979, Wakeham *et al.* 1980, Sporstøl *et al.* 1983, Steinhauer and Boehm 1992, Yunker *et al.* 1993). A "smoking gun" is provided by the presence of hydrocarbon sources (oil seeps and source rock bitumens) in the lower Mackenzie River watershed which have similar biomarker patterns for the isoprenoid alkanes and hopane triterpanes to those observed in the delta (Snowdon *et al.* 1987, Feinstein *et al.* 1988). Concentrations and ratios of the parent and higher plant PAHs for the river and shelf are very similar, and we therefore conclude that the Mackenzie River is the prime source for both parent and alkyl substituted PAHs in shelf sediments.

The alkyl homologue distributions suggest that the PAHs delivered to the shelf by the Mackenzie River have their most likely origin in petroleum hydrocarbons. Transport from the river to shelf sediments occurs without substantial change in PAH composition suggesting that the majority of the two- to four-ring compounds are encapsulated in eroded bitumen, and hence protected from dissolution or bacterial degradation.

To take full advantage of the information available in the hydrocarbon data and to provide a more robust analysis of sources of PAH requires more advanced techniques than the simple compound by compound comparisons illustrated above. Multivariate analysis using principal component analysis (PCA) is one such technique in which all of the data are applied to the search for patterns to reveal sources. A PCA of the Mackenzie Shelf data set indicates that PAHs in Mackenzie River particulates, shelf sediments and peat samples from eroding coastal areas of the shelf have a composition pattern distinctly different than long-range-transported atmospheric aerosol collected at Alert, NWT by Patton *et al.* 1991. The atmospheric samples from Alert contain high proportions of fluoranthene while, in contrast, samples of suspended particulate, sediment and peat from the Mackenzie Shelf have high proportions of benzo (ghi)perylene, benzo (e)pyrene and benzo (a)pyrene. Apparently, it is the Mackenzie River that supplies most of the shelf's PAH, and not the atmospherically delivered combustion products of fossil fuel.

But what is the source of the higher, non-petroleum PAHs in the Mackenzie River? A comparison of the concentrations and distributions of the high molecular weight parent PAHs to the naturally-derived PAHs that are produced by the diagenesis of conifer resins and other higher plant lipids can help to identify the origin of parent PAH (Wakeham *et al.* 1980, Simoneit and Mazurek 1982, Bouloubassi and Saliot 1993). Data suggest that the naturally derived PAHs are preformed in soils and peat and are subsequently delivered to the river and transported to shelf sediments with little alteration (Bouloubassi and Saliot 1993). Concentrations and ratios of the high molecular weight parent PAHs and the naturally-derived PAHs show little change between the river and the shelf, again suggesting that the Mackenzie River is the prime source for both components. The high molecular weight PAHs are tentatively attributed to combustion relics that have been preserved and concentrated in peat (Yunker *et al.* 1993).

The majority of the high molecular weight PAHs, together with the petrogenic PAHs found in sediments of the Mackenzie Shelf, are delivered "preformed" to the region by the Mackenzie River. Despite the high PAH content of shelf sediments, we consider the area still to be pristine — that is, dominated by natural PAHs. Only at the shelf edge, about 100 km from the mouth of the Mackenzie River, do we see any indication of combustion PAHs, and even there, the signature is weak. One consequence of the naturally high PAH background may be that this region of the Arctic Ocean may be more sensitive to additional PAHs from human activities.

- *The majority of the high molecular weight PAHs and petrogenic PAHs found in sediments of the Mackenzie Shelf, are delivered to the region by the Mackenzie River. These PAHs are predominantly from natural sources. Only at the shelf edge, about 100 km from the mouth of the Mackenzie River, do combustion PAHs become more important as a relative source.*

2.7 Conclusions and Recommendations

Between 1991 and 1996, significant progress has been made by the Canadian Northern Contaminants Program (NCP) in understanding the occurrence, sources, and pathways of contaminants in the Arctic environment north of 60° latitude. There has been a proportional increase in the knowledge of the pathways of OCs, PAHs, metals, and radionculides of 93%, 60%, 32%, and 34%, respectively. However, there are still many unknowns, since due to limited observations and knowledge of processes, a preponderance of pathways have been estimated, rather than accurately described.

2.7.1 Transport and Distribution

Most contaminants in the Canadian Arctic originate from external globally distributed sources. The atmosphere and the oceans are the main transport routes. Rivers play an important role in re-distributing contaminants within the Arctic and possibly as sources of contaminants to the Arctic Ocean.

2.7.2 Sources

Under the NCP, progress has been made in quantifying the geographical distribution and history of the global application of agricultural pesticides. For example, studies clearly indicate that the reductions in the use of HCHs (in China in the 1980s and in India in the 1990s), have been reflected by a 90% decrease in observed arctic atmospheric HCH concentrations. Similar declines in HCH levels, however, have not been observed in the Arctic Ocean. An improved mass balance budget of HCHs in the Arctic Ocean, based on observations collected under the NCP, provides insight for this lack of change in HCH concentration in the Arctic Ocean. The majority of the polar mass of HCH resides in the Arctic Ocean, from which it is slowly lost, over decades, by outflow in marine currents (68% of total mass) out-gassing to the atmosphere (16%), chemical destruction (14%) and ice export (2%). The Arctic Ocean has now become a source, rather than a sink, of HCHs to other parts of the globe. If HCH global usage suddenly stopped, the time required for the Arctic Ocean to return to an HCH-free state (assuming zero emissions), has an upper limit of the residence time of water in the Arctic surface ocean layer, which is 12.5 years. Continued observations of the recovery of the Arctic Ocean to an HCH-free state will provide insight into general oceanic contaminant pathways and exchange.

2.7.3 Phase-Partitioning

The partitioning of semi-volatile contaminants, in particular OCs, PAHs, and Hg, between various environmental media such as air, water, snow, suspended solids, and atmospheric particles, is a key process in understanding their fate in Arctic systems. The degree of phase-partitioning varies among the contaminants of concern, and is dependent on the physicochemical properties of the chemical. Contaminants that are less likely to partition to particles, such as HCHs, tend to remain in air or water rather than sediments. In contrast, contaminants with a high affinity for particulate matter, such as DDT, PCBs, toxaphene, and chlordane, tend to move to such media as soil, suspended solids, sediments, snow, rain and atmospheric particles. Budgets for the concentrations of these compounds in various environmental media have been determined for small freshwater lakes (for example Amituk Lake in the high Arctic and Lake Laberge in the Yukon). In contrast, there are no budget estimates for the partitioning of these compounds in larger aquatic systems, such as the Arctic Ocean, the Mackenzie delta estuary or Great Slave Lake.

2.7.4 Modelling

The environmental modelling of organic contaminants has clearly demonstrated that global dispersion of volatile-substances can be realistically simulated. For example, the global fate of HCH, which partitions strongly into water, and the fate of OCs and Hg in freshwater systems have been successfully modelled. The application of global models, such as that of Wania and Mackay (1993b), to semi-volatile contaminants that are subject to solid-phase partitioning, such as DDT, PCBs, toxaphene, and chlordane, remains to be done. Modelling efforts are limited by a lack of information for the whole globe on the interaction of semi-volatile OCs with continental solid surfaces (soil, vegetation and snow). Unique laboratory experiments conducted at the University of Waterloo have advanced our understanding of these interactions. Such experiments have extrapolated the results that describe soil and snow exchange processes for non-contaminant compounds, that have similar physicochemical properties to the semi-volatile OCs present in the Arctic. Verification of these theoretical predictions for the semi-volatile OCs under Arctic conditions is now required.

2.7.5 Atmosphere

A continuous seasonal record of atmospheric concentrations of OCs, PAHs, and Hg has been obtained in the Arctic for the first time because of the NCP. OCs and PAHs have been measured at three Canadian monitoring sites and one Russian Arctic site. These data provide valuable insight into the transport pathways of contaminants (when combined with global scale models of contaminant transport). In the high arctic, the atmosphere generally contains a higher proportion of lighter PCB congeners (di- and tri-chlorinated PCBs) than do snowfall, snowpack, and sediment. This implies that the heavier PCB congeners are selectively scavenged from the atmosphere and retained by snow at the Earth's surface. The level of PAHs in the atmosphere generally decreases from Russia to Alert (Canadian high Arctic) to Tagish (Yukon). The proportional composition of PAH compounds in environmental samples, including the atmosphere, contains relevant information on the origin of the total PAH mixture. For example, a wood smoke PAH (retene) is much more abundant in the Yukon and Russian air samples than in the high Arctic air samples. This has been observed even during winter, when forest fires are absent, indicating the presence of wood fire pollution. For the first time, arctic atmospheric Hg concentrations have been measured routinely for a year or more under the NCP. The seasonal variation observed in 1992–93 provides comparative baseline values for the future verification of the hypothesized 0.6% to 1% a^{-1} increase in global levels of anthropogenic Hg.

OCs deposited in snowfall have been routinely measured throughout the snow season at 12 Arctic locations. Such data is useful in understanding the atmospheric budget of OCs and for describing post-depositional losses of OCs from the snowpack. Post-depositional losses were estimated by comparing the cumulative seasonal OC concentrations in snowfall to the end-of-season snowpack concentration burden. Significant losses of HCHs, but not of PCBs, were observed. This is consistent with studies of OC retention in Agassiz glacial ice. Snowpack burdens of OCs have been made throughout the Arctic between the spring (April-May) and snowpack melt in June. During this period, dieldrin, DDT, and PCBs are retained by the snowpack, while HCHs, HCB, and chlordane volatilize to the atmosphere. Those substances exhibiting little loss to the atmosphere are available for run-off into streams and lakes. No measurements of snowpack Hg have been made. A carefully designed field experiment is needed to measure the process of contaminant retention by the snowpack as it warms in the spring from -20°C to O°C. Results would contribute to a quantitative model of the atmosphere-snowpack exchange. To complete an understanding of the transport of contaminants via wet deposition requires the measurement of OCs and Hg in rainfall.

2.7.6 Oceans

Recent observations confirm the relative abundance of OCs in arctic seawater as, in decreasing abundance: α-HCH > HCBs > γ-HCH (≈) CHBs (toxaphene) > chlordanes (≈) PCBs > DDTs. However, with the exception of HCHs, there are too few measurements available to assess the spatial or vertical distribution of OCs in the water column. Sea ice generated on continental shelves of the Arctic Ocean has been identified as a potentially important medium for contaminant transport over long distances across the Arctic. In future, measurements of the transport of arctic ice, and its contaminant burden, are needed to better quantify the significance of this transport mechanism. Measurements of OCs in suspended particles, in vertically settling particles and bed sediments would be extremely valuable in producing complete budgets and removal rates of particle-reactive contaminants from the upper ocean. However, there are few measurements presently available. Similarly, there are few data with which to estimate the removal of OCs from the water column by processes such as hydrolysis, photolysis, and metabolism.

Historically, the world's oceans have been contaminated by industrial Pb inputs with the result that there is no baseline against which to measure current inputs from non-natural sources. From work done in other oceans and form stable isotope measurements in arctic media it seems likely that much of the Pb presently cycling in the Arctic Ocean originates from industrial, rather than natural, sources. In contrast to Pb, Cd concentrations in the Arctic Ocean are within the range of concentrations measured in other oceans. Because Cd follows phosphorus in the biological cycle, water column distributions reflect removal at the surface by primary production and addition at the depth where nutrients regenerate. Since Cd is not quickly scavenged in ocean waters, its residence time in the world ocean is very long (order of 10^4 years); thus anthropogenic inputs are negligible when compared with the large natural ocean burden. Pb and Hg do not share this feature of Cd. Their residence times in the oceans are much shorter. Mercury cycling in the Arctic marine environment is not well known due to a paucity of data for all media except sediments. Consequently, an assessment of the inputs of Hg from natural and anthropogenic sources is not presently possible. It has been hypothesized that anthropogenic emissions of Hg in temperate regions over the past century may have resulted in as much as a three-fold increase in global atmospheric and ocean surface-water concentrations. It is estimated that it would require 15 to 20 years to eliminate the anthropogenic Hg load from the world's upper oceans if all anthropogenic emissions of Hg were to be stopped. The rate of decrease of Hg in the Arctic Ocean is anticipated to be governed by the residence time of water in the ocean surface layer which is approximately 12.5 years. Since Hg, in particular methylated Hg, has similar properties to many of the OCs of concern, (i.e., volatile, toxic, and biomagnifies), it should be a priority to determine its fate and distribution within the Arctic Ocean.

2.7.7 Rivers

OCs in Canadian Arctic rivers have not been systematically or thoroughly measured with sufficiently sensitive analytical techniques to yield quantitative concentrations of dissolved and particle-associated fractions. Under the NCP, limited measurements have been made of some OCs: ΣHCH, ΣPCBs, ΣDDT, and chlordane (listed in decreasing concentration). There are no systematic measurements of the heavy metals, Hg, Pb and Cd, in Canadian Arctic Rivers.

PAHs have been measured primarily in the suspended sediments of the Mackenzie River. The Mackenzie River has a higher suspended solid load (127×10^6 $t{\cdot}a^{-1}$) than any other northern river and PAHs tend to sorb to this particulate matter. The suspended sediments and associated PAHs are transported from the Mackenzie River into its delta and the Beaufort Sea. The principal sources of these PAHs are natural seepage from hydrocarbon reservoirs, and natural erosion from tar sands and hydrocarbon-rich shales. An estimated 50% of the sediment load of the Mackenzie River is retained in the delta. Therefore, the predominant source of PAHs to the delta, and to a lesser extent the Beaufort Sea, are natural PAHs delivered by the river. Anthropogenic sources of PAH contamination to this area include oil production, atmospheric long-range transport, and shipping. The anthropogenic load of PAHs is trivial in comparison to the natural PAH load of the river, however, as the direct influence of the Mackenzie River decreases, at the shelf edge and into the Arctic Ocean, the relative importance of atmospheric deposition increases, and finally becomes the dominant source.

2.7.8 Lakes

Most lakes in the Arctic and sub-Arctic receive contaminants by long-range transport. Snow in catchment areas can capture and convey contaminants to lakes. In general, a high Arctic lake retains a small fraction of the total OC input to the catchment basin; the specific fraction is a function of lake hydrology (e.g., the timing of snowmelt versus lake turnover), and productivity (e.g., the fraction of contaminants conveyed to bed sediments in association with suspended particles). In the spring, lakes act as rivers, channelling the year's accumulated contaminant inputs downstream. In northern lakes, low productivity and organic carbon result in minimal retention of hydrophobic OCs. Consequently, unlike temperate lakes, "individual" northern lakes are weak sinks for contaminants. However, "systems" of northern lakes can act cumulatively to be major sinks. In more productive systems in the lower Arctic, a higher proportion of contaminant loadings are retained by the system, which may be available for uptake through the food web.

2.7.9 Exchange Processes

Research conducted under the NCP has advanced the knowledge of the exchange of contaminants between the major environmental abiotic compartments in the Arctic including the atmosphere, oceans, terrestrial systems, and freshwater. Post-depositional losses of OCs to the atmosphere after snowfall have been estimated from comparing OC concentrations in snowfall and snowpack. Application of atmosphere-snow partitioning theory, based on laboratory experiments, indicates that in general, snow is a more effective scavenger of OCs than rain or than dry particle deposition. However, post-depositional losses from the Earth's surface after rainfall may be less than after snowfall, thus reducing the relatively greater scavenging ability of OCs by rain. Atmosphere-soil exchange is presently poorly understood. Gas exchange is the most important mechanism dictating the fate of HCHs in the Arctic. It is relatively less important for OCs, of a lower volatility and solubility than HCHs such as HCBs, dieldrin, chlordane, DDT, and PCBs. For these compounds other exchange mechanisms, such as dry and wet precipitation and particle deposition, are important in dictating their fate and distribution in the Arctic.

Canadian riverine inputs of selected OCs to the Arctic Ocean have been estimated from limited observations. The Mackenzie River is the major contributor of contaminant burdens to the Arctic Ocean, however, considerable uncertainty exists regarding the quantity of inputs from smaller Canadian rivers. There are few measurements of the contribution of Russian rivers to the Arctic Ocean contaminant burden. The information that does exists indicates that Russian rivers are 100 times more important to the overall water budget of the surface Arctic Ocean. Therefore, the contribution of these river systems to the contaminant burden in the Arctic Ocean is highly uncertain but potentially significant. There is a need to measure contaminant concentrations in Russian rivers over a full hydrological cycle.

The Arctic Ocean is unique because it has a large fraction of area covered by shelves (30%) that act as a transition zone between the land and the interior ocean. Deltas and shelves trap riverine inputs of suspended particles and sediments together with the associated particle-reactive contaminants. It is estimated that half the particulate loading from the Mackenzie River remains in the delta, while the majority of the remainder becomes trapped on the shelf. Only a small proportion (10% to 15%) exits the shelf.

Ocean-to-sediment exchange is a potentially important contaminants pathway (sink) in deltas, shelves, and open oceans. Many OCs, PAHs, PCBs, and other contaminants are transported to sediments in association with particulate matter. However, there are few measurements of sediment contaminant concentrations and accumulation rates. Knowledge of contaminant dynamics in temperate aquatic systems can be used to understand the potential fate of contaminants at the sediment-water interface in the Arctic. Thus particle-associated contaminants may be released to pore-waters to diffuse into the overlying water column, or contaminated sediment may be ingested by foraging biota and re-enter the food chain. In addition, sediments may be resuspended during storms, especially on shallow parts of the shelves (<20 m), or incorporated into bottom-fast or frazil ice. While these processes are an important component in ocean-sediment exchange, few measurements are available to quantify their rates or the quantities of contaminants moved by them in the Canadian Arctic Ocean.

2.7.10 Temporal Trends

Contaminant trends over time have been estimated from three sources of information: long term atmospheric concentrations (15 years), glacial ice cores (100 years), and sediment cores (100s of years). Atmospheric observations indicate that since 1980, concentrations of HCHs in the high Arctic have decreased by approximately 90%. This decline is consistent with changes to the global usage of this agricultural pesticide, particularly in China and India. Pb in the atmosphere at Alert, has decreased by approximately 50%, which is consistent with changes in emissions to the atmosphere in Eurasia, but not with emission changes in North America, which were much greater than Eurasia. This information, together with atmospheric chemical transport models, implicates Eurasia, rather than North America or southeast Asia, as the main source of Pb to the Arctic. The concentration of PAHs and PCBs in ice cores from the Agassiz glacier, northern Ellesmere Island, indicate clearly that these compounds have peaked in concentration in the last half of the 20th century. This is consistent with the general global use of these compounds. The PAH compounds (phenanthrene and fluorene) show a marked decrease in levels after a peak in the late 1960s and early 1970s, while PCB concentrations show no consistent trend. There is a need for more ice-core chemistry observations on Ellesmere Island. It would be worthwhile to quantify the highly particle-associated substances, for example B[a]P and heavy PCB congeners, that are not likely to experience post-depositional losses from the snow.

Freshwater sediments are a good source of temporal trend information because sediment layers build-up relatively undisturbed over time. In comparison marine sediments are subject to disruption by ocean currents, ice scouring, and biological mixing. In Arctic lakes, contaminants that are entrained in sediments persist due to a lack of perturbations at the sediment water interface, including: minimal resuspension, low rates of sediment-water diffusion, and minimum biological activity. A lake sediment survey throughout the Canadian Arctic has shown that in high Arctic lakes, PCBs tended to peak in the 1980s and 1990s, lagging the peak in production and use of these compounds in the early 1970s. This is consistent with a predicted lag time of decades, between the release of a semi-volatile compound and its appearance in the Arctic. Further south, in lakes undisturbed by local dumping, there is less of a lag-time between peak PCB concentrations in dated sediments and the time period of actual maximum PCB production. Local dumping of PCBs between 1945 to 1955 was evident from sediment cores from Lake Laberge, near Whitehorse, Yukon. Sediment cores also revealed the occurrence of local dumping of DDT during the war and postwar years. Concentrations of Pb and Hg in lake sediment cores indicate that anthropogenic sources contribute substantially to the observed levels. Unlike synthetic OCs, however, metals have a natural geological source that contributes to the total concentration in lake sediments. The anthropogenic input to freshwater systems has been estimated from the ratio of the current depositional flux to the natural pre-industrial flux of metals. These ratios ranged from 1.1 to 7 for Hg and from 1.1 to 10 for Pb, indicating that anthropogenic sources are on average greater than natural inputs.

Artificial radionuclides have entered the Arctic predominantly from three sources: atmospheric weapons testing, nuclear waste discharges into northern European seas, and disposal of radionuclides and underwater radionuclide testing carried out by the Russians on their Arctic continental shelf. Since the 1960s, the atmospheric source of radionuclides has been reduced by two or more orders of magnitude due to test ban treaties. Although remnants of the testing can still be detected in soils, sediments and in the surface ocean, this source is no longer of significant concern. Similarly, the discharge of radionuclides from re-processing plants into northern Europe seas has been reduced and is strictly regulated at source. Radionuclides from these re-processing plants enter the Canadian Arctic within about 10 to 15 years of discharge and, ironically, have served a beneficial purpose as sensitive tracers revealing the current layered structure of the Arctic Ocean. Discharges of radionuclides directly into the Arctic Ocean by Russian activities have recently caused the greatest concern. There is, at present, little evidence of leaking radioactive material circulating into Canadian or other waters. The detection of the European re-processing signal has shown the likely pathways that radionuclides will follow in the event of any leakage of disposed containers. It is likely that radioactive elements disposed on the Russian Arctic shelves will either remain in the sediments or will be transported around the basin margins at depths below which most biota reside. Perhaps the greatest concern is the potential for ice to transport radionuclide-contaminated bottom sediments to distant locations, with little or no dilution. The predominant transport direction of ice formed on the Russian shelves is, however, into the Barents Sea, through Fram Strait, and into the Greenland Sea. Thus North America is likely to remain relatively immune to such transport.

2.7.11 Marine Pathway Knowledge Gaps

Knowledge gaps related to the pathways of HCH and other OCs in the marine environment are listed below:

a) Much work remains to be done to establish the residence times and volumetric flows of the various water masses into and out of the Arctic Ocean. Currently, uncertainties in the advective flow estimates are in the order of 30% to 50%.
b) The fraction of open water, which controls the air-sea gas flux, is a highly variable and poorly known parameter that is subject to bias when estimated by the conventional technique using satellite imagery.
c) Although the HCH inventories of the Canada Basin and the Bering-Chukchi seas have been relatively well established, the database for HCHs in the water column of the Eurasian Basin and its regional seas is weak, most of the measurements being from 1985. Better coverage of these regions is needed to establish the inventory of HCHs in the water column and concentrations in the major currents feeding and draining the Arctic Ocean.
d) Improved estimates of HCH concentrations in Russian rivers are needed.
e) Microbial breakdown of HCHs in the water column is a potentially significant sink which needs to be quantified, especially considering the long residence times of water masses in the central surface Arctic Ocean (12 years) and the extremely low rates of other chemical processes (e.g., hydrolysis). Also, vertical flux mediated by biological production may be important in the seasonal cycling of HCHs and other pesticides especially in highly productive regions of the marginal seas.
f) Scavenging and release of HCH vapours to snow are significant components of the atmospheric budget.
g) Further work needs to be done on air-sea gas exchange, especially with regard to improving confidence in estimates of mass transfer under different states of ice cover. Direct measurements of air-sea exchange are needed to corroborate present estimates that depend wholly on predictions of the two-film model for estimating gas fluxes.
h) Further measurements of the concentrations of particle active OCs like DDT or PCBs in ice are required to better understand the role of ice in transporting them within and out of the Arctic Ocean.

2.7.12 Toxaphene

Toxaphene is an insecticidal mixture consisting largely of chlorinated bornanes (CHBs) with a small proportion of chlorinated camphenes. Even though CHB mixtures have been banned or severely restricted in many countries, continued input to the environment may occur from regions outside the Arctic where use is still permitted and by volatilization from contaminated soils. CHBs, originating from long-range transport through the atmosphere, have been responsible for the closure of a commercial and sport fishery in Lake Laberge (Yukon). In winter, they are deposited mainly by precipitation and particle dry deposition while in summer gas exchange with open water facilitates their movement. The food chain can amplify the effects of atmospheric deposition, leading to elevated levels of CHBs in top predator fish in some lakes and not in others. One of the great difficulties in making quantitative budget estimates of CHBs for lakes or the Arctic Ocean is the disparity in analytical methods of measuring this complex mixture of compounds. It is recommended that considerable effort be made to quality control and assure a variety of sampling and analytical methodologies used for measuring CHBs. A laboratory inter-comparison of analytical techniques that includes recognized international laboratories is highly desirable.

2.7.13 PCBs: Local and Long-Range Sources

One of the frequently posed questions in the north is: "how much is local dumping at Dew Line sites contributing to PCB levels?" A careful assessment of this issue has been made based on the combined results from the NCP and from a major DEW Line study funded by the Department of National Defence. It is concluded that local contamination is considerably more important than atmospheric deposition within the locally impacted areas but that the extent of these areas is quite limited. PCB levels on an arctic-wide scale remain dominated by long-range atmospheric transport and deposition. Contamination surrounding DEW Line sites (less than 10 km radius) comprises 0.2% of the total area of the Yukon and NWT. A comparison of total DEW Line PCBs remaining in the Arctic and anthropogenic PCBs transported into the region and deposited over a 30 year period was made for two areas: (i) the total area within 10 km of the DEW Line stations and (ii) the area of the Yukon and NWT. For area (i), dumped PCBs are five times greater than long-range transport inputs while, for area (ii), they are only 1% of the long-range transported PCBs.

2.7.14 Mercury

Natural mercury cycling has been globally augmented by a multitude of anthropogenic activities, such as smelting, mining, or combustion, that mobilize this element. Evidence from a variety of sources, including atmospheric monitoring, lake and marine cores, marine mammal samples and human hair samples, suggest that Hg concentrations have been increasing in the environment. This increase is consistent with global environmental budgets of mercury that suggest post-industrial acceleration of Hg cycling. However, the budgets and trends are very uncertain, as is the role of the Arctic in the global Hg cycle. Because Hg shares some of the organochlorine properties (i.e. it is volatile and biomagnifies), it is likely that polar regions are more sensitive to increases in Hg cycling than other parts of the globe. More research is needed to reduce uncertainties in these budgets and to determine Hg trends in polar regions.

2.7.15 Cadmium

Cadmium is listed as a metal of concern in the international Arctic Monitoring and Assessment Program (AMAP). Cd is easily mobilized from marine sediments and bioaccumulates to high concentrations in the organs of some marine mammals. However, there is no compelling evidence to indicate that anthropogenic loading of Cd is presently of concern in the arctic marine environment. This conclusion is based on very low anthropogenic atmospheric inputs of Cd compared to very high natural marine cycling, and the fact that Cd does not tend to biomagnify in the food chain (biomagnification in the food chain is different than bioaccumulation in particular organs of a species mentioned above). However, not enough is known about the transfer of Cd into the food chain to predict with confidence how the system would respond to small incremental increases in the atmospheric flux resulting from anthropogenic activities. Also the effect of anthropogenic Cd deposited on lichen and available to caribou and wolf needs further consideration.

2.7.16 New Generation Pesticides

New generation pesticides are appearing in the Arctic, despite the fact that they are classed as readily biodegradable. Evidence indicates that rates of degradation of these compounds in water slow rapidly as temperature drops to Arctic Ocean temperatures. Research is needed to understand potential threats to the arctic environment from these compounds and their degradation products.

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Chapter 3: Ecosystem Uptake and Effects

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CHAPTER 3 Ecosystem Uptake and Effects

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3.1 Introduction

3.1.1 Objectives

This chapter reviews the pertinent information available on the spatial and temporal trends of contaminants in biota in the Canadian Arctic and, where possible, discusses biological effects. Most of the data are drawn from the Ecosystem Uptake and Effects subprogram of the Arctic Environmental Strategy Northern Contaminants Program (AES–NCP) (1991–1997). The design of this subprogram evolved from the policy objectives of providing timely advice to northerners regarding consumption of country foods and of protecting the health of northern ecosystems. Specific research objectives were to document spatial and temporal trends in biota in order to establish if contaminant levels were increasing or decreasing in space and time and to quantify the effects of contaminants on the Arctic ecosystem using biochemical indicators of exposure of wildlife to toxic chemicals (DIAND 1991). The overall objective was to integrate this information into a comprehensive assessment of the risks to the ecosystem.

A six-year subprogram was formulated which, at its maximum size in fiscal year 1994/95, included 28 projects investigating the presence of persistent organics, heavy metals and radionuclides in marine, freshwater and terrestrial biota and biological effects on fish and marine mammals.

3.1.2 Background

Information on the presence of persistent organic pollutants (POPs) and heavy metals in Arctic ecosystems is not new. The first report on organochlorine (OC) contamination of Arctic marine mammals in the Canadian Arctic was by Holden (1970) who detected the chlorinated pesticides dieldrin and DDT, as well as PCBs, in blubber of ringed seals (*Phoca hispida*) from Baffin Island. More detailed reports on DDT-related compounds and PCBs in ringed seal and beluga whales (*Delphinapterus leucas*) were published by Addison and Smith (1974) and Addison and Brodie (1973). Bowes and Jonkel (1975) were the first to report OCs in polar bears. OC contaminants were also determined in Arctic freshwater and anadromous fish in the early 1970s (Reinke *et al.* 1972, Bowes and Jonkel 1975).

Studies of heavy metal contaminants in Arctic marine mammals and fish also began in the early 1970s with reports of mercury in beluga, ringed seals and arctic char (*Salvelinus alpinus*) (Bligh and Armstrong 1971, Smith and Armstrong 1975) and in lake trout (*Salvelinus namaycush*) (Reinke *et al.* 1972). Measurements of metals were stimulated by mining developments such as the Nanisivik mine near Strathcona Sound on Baffin Island where large numbers of samples of benthic fish and invertebrates were analysed prior to and post development (Fallis 1982). Unlike the persistent OCs, which are anthropogenic (man-made), the presence of heavy metals in arctic biota is due to both natural and anthropogenic sources. It is virtually impossible to determine the proportion due to each source from measurements in biota. However, some insight can be gained by studies of atmospheric transport, and dated sediment or snow cores (see Chapter 2), and by analysis of fossils such as shells of mussels. Similar anthropogenic/natural source issues confound discussion of contamination of biota by hydrocarbons and some radionuclides.

Measurements of polyaromatic hydrocarbons (PAHs) and aliphatic hydrocarbons in Arctic marine biota began in the early 1970s as part of background studies carried out with petroleum exploration (Wong *et al.* 1976, Cretney *et al.* 1987). A wide range in concentrations of aliphatic hydrocarbons were observed with pristane and squalene being the dominant components in fish liver and muscle of arctic species (Johansen *et al.* 1977). The low bioaccumulation potential of most petroleum hydrocarbon components is well known (Malins and Hodgins 1981). Aromatic hydrocarbons are rapidly taken up by fish, but are readily transformed by monooxygenase enzyme systems to polar products such as phenols or methanol derivatives, as well as to various conjugates, and eliminated. As a result the Northern Contaminants Program focused relatively little attention on PAHs and aliphatic hydrocarbons. Nevertheless, contamination by hydrocarbons as a result of oil spills, oil field development, or natural hydrocarbon seeps is an ongoing pollution issue in the Arctic and measurements in biota will be briefly reviewed and assessed in this chapter.

Although the presence of toxic chemicals in Arctic ecosystems was well known among scientists interested in the global distribution of these chemicals, information on spatial trends in contaminants was very limited until the early 1990s. Most studies had regarded the Arctic as a relatively pristine site for comparison with more contaminated southern locations. Several developments in the late 1980s helped to accelerate the assessment of contaminants in arctic food webs. The results of studies on these contaminants in the human diet (Kinloch *et al.* 1992) and mothers' milk (Dewailly *et al.* 1989) demonstrated the transfer of bioaccumulating contaminants to humans and created a demand from Aboriginal communities for more information on contaminants in "country foods." Improvements in analytical chemistry techniques in the 1980s also permitted a more comprehensive search for individual OC and hydrocarbon contaminants, and more rapid processing of samples for mercury and other heavy metals. Development of biochemical effects tests such as cytochrome P450 mixed function oxidase (MFO) enzyme activity, DNA oxidative damage and immunocompetence in the 1980s also provided tools for assessing biological effects of the contaminants in biota.

3.2 Pathways and Processes of Delivery of Contaminants to Marine, Freshwater and Terrestrial Ecosystems

3.2.1 Pathways of Transport of Contaminants to Arctic Ecosystems

Pathways of transport of the OCs, heavy metals, radionuclides, and hydrocarbon contaminants to the Arctic include transport in the troposphere in gas phase and on particles, as well as via ocean currents (Barrie *et al.* 1992). Evidence for this is reviewed in Chapter 2. The focus here is on the processes that deliver the contaminants to marine, freshwater, and terrestrial environments where they can become available for uptake by biota.

Airborne contaminants are removed from the atmosphere by absorption, precipitation, and dry deposition (Figure 3.2.1). Many chlorinated organics are present as gases even at low temperatures (see Chapter 2) and are absorbed from the gas phase by water, snow and plant surfaces. Precipitation scavenging of gases and particles from the air also deposits particle-associated OCs and metals in snow and rain. Dry deposition is a third pathway of input of aerosol-bound contaminants to terrestrial and aquatic ecosystems.

In the water column, the highly chlorinated OCs such as DDT and PCBs (Cl_4–Cl_{10}) are associated with particles while others which are more water soluble (HCH and toxaphene) are mainly in the dissolved phase. Recent measurements of OCs in small high arctic lakes show ΣHCH concentrations similar to sea water but much higher concentrations of PCBs (0.3–0.9 $ng{\cdot}L^{-1}$ in filtered lake surface waters versus 0.045 $ng{\cdot}L^{-1}$ in Cambridge Bay). The higher levels in remote arctic lakes than in sea water probably reflect substantial contributions from snow melt runoff, as well as higher dissolved and particulate organic carbon in the freshwater environment (Reimer *et al* 1993a).

Water-borne contaminants also enter Arctic ecosystems from northward flowing rivers such as the Athabasca/Peace/Slave River system which feeds into Great Slave Lake, the Nelson River/Lake Winnipeg drainage, and other major rivers flowing into Hudson Bay.

3.2.2 Bioavailability

Hydrophobic organics and heavy metals such as cadmium and lead are readily adsorbed by living and dead organic matter such as particulate organic carbon, waxy plant surfaces, animal membranes, and fats. Once adsorbed the bioavailability of these chemicals to aquatic and terrestrial animals will

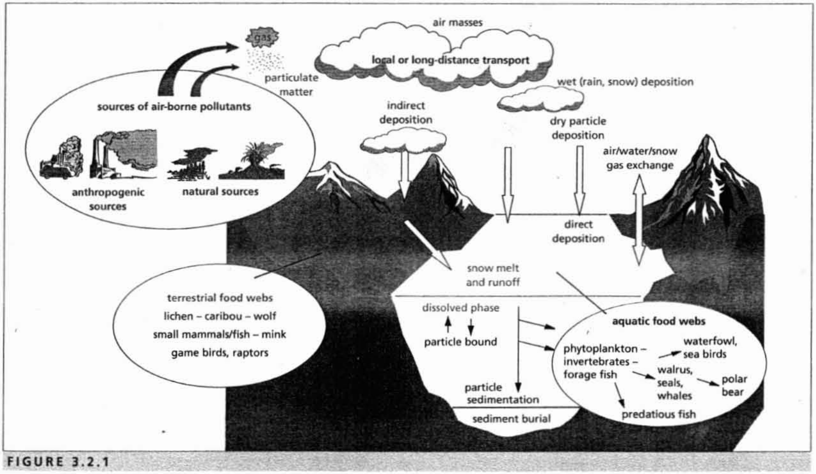


FIGURE 3.2.1

Pathways of transport and accumulation of persistent organics and metals to Arctic freshwater and marine ecosystems.

depend on the properties of the chemical and on the physical, chemical and biological environment into which it is released. Bioavailability refers to the extent to which pollutants associated with soils, plants, sediments or suspended or dissolved organic carbon in the water column are available for uptake by biota (Dickson *et al.* 1994). Many factors such as the organic carbon content of soils and sediments, pH, and kinetic limitations influence the release of adsorbed contaminants and therefore the environmental bioavailability; for example, from food particles in the gut or from sediment into surrounding pore-waters. Despite being tightly bound to particles, membranes and fat globules or proteins, most OC and metal contaminants of concern in the Arctic have been shown (in laboratory studies with invertebrates, fish, mammals and birds) to be readily assimilated from the diet, and when present in the dissolved phase in water, to be rapidly accumulated from water.

3.2.3 Bioaccumulation Processes

Bioaccumulation is the transfer of a chemical from water and/or diet into an organism. For persistent OCs, such as PCBs, properties of low water and high lipid solubility and the relative biological inertness of these compounds account, in part, for bioaccumulation. For metals, differences in uptake due to speciation of the element (which may be influenced by water hardness, alkalinity, redox conditions in sediment, pH and temperature), as well as metabolic rate of the organism, affect transfer across biological membranes (Heath 1987). The protein metallothionein is important in regulating the accumulation of metals in the liver of mammals and fish and the elimination of metal into the bile following uptake.

In the aquatic environment, chemicals are accumulated by fish and invertebrates via three major routes of exposure: gills, dermis (skin) and diet. Dissolved chemicals, both organics and metals (in the ionic form), enter organisms by diffusion across the gill membrane (or other respiratory surfaces) into the blood (Connell 1988, Heath 1987). A steady state is established between the concentration of OCs in the water and in the lipid phase of the organism. For metals a steady state is established between dissolved forms in water and dissolved or protein-bound forms in blood and other tissues. In small organisms, direct absorption of these chemicals through the integument is a major pathway of uptake because of their high surface area to volume ratio (Sheldon *et al.* 1972). OCs and heavy metals which are adsorbed onto particle surfaces are taken up as food by grazing animals (e.g. zooplankton).

Food ingestion (coupled with metabolism and excretion) is the dominant pathway for bioaccumulation of OCs and heavy metals in larger marine and freshwater organisms, and in terrestrial organisms. For most of the OCs of concern in the Arctic, as well as methylmercury, accumulation via food predominates over uptake from water. This is due to relatively low concentrations in water (typically low or sub-ng·L) and low rates of elimination of these chemicals by the organism. Absorption from the diet involves diffusive transfer across the gut membrane for both organic contaminants and metals. In the case of organochlorines, diffusive transfer across gill, gut or internal membranes is influenced by the lipophilicity and polarity of the molecule as well as by its molecular size and shape and biodegradability. Hydrophobic organics, which are not metabolized and which have a molecular weight of less than about 600 daltons, are highly bioaccumulative. This group includes PCBs, and OC pesticides such as DDT, chlordane and toxaphene.

Transfer within the food web leads to biomagnification (the successive increase in concentration of a chemical with increasing trophic levels). The food requirements of an organism are controlled by its metabolic rate and production (e.g. growth, lipid deposition, reproduction). Thus metabolic rate is linked to the rate of uptake of a contaminant. Homeotherms (mammals, birds) maintain their body temperature and therefore have greater energy requirements for the same body weight than poikilotherms (e.g. fish). Thus, their metabolic rate and caloric requirements are higher. As a result, the largest biomagnification factors (i.e. the concentration of contaminants in predator divided by concentration in prey) for the higher chlorinated PCBs, OC pesticides and mercury, are usually observed between fish and marine mammals or sea birds in arctic food webs (Muir *et al.* 1992a).

3.2.4 The Food Webs

3.2.4.1 General Biological Characteristics

Characteristics of arctic food webs relating to the extent and manner in which contaminants accumulate are described below.

1. In the Arctic, food consumed by humans is often higher in the food chain than in southern latitudes. This is related, in part, to anthropogenic influences in southern ecosystems such as the dominance of agriculture. It is also related to cultural differences.

   High level carnivores in temperate ecosystems (e.g. seals and other marine mammals, birds of prey, wolves, wild cats), seldom constitute major food items for humans. In contrast, in the Arctic it is not uncommon to have several levels of carnivory, with humans harvesting animals at all levels. Also, much energy in the arctic food chain is transferred in the form of lipids, and thus associated contaminants.

2. Productivity is low in the Arctic. This is primarily due to low levels of nutrient input, rather than the cold climate or short growing season (Dunbar 1986). Atmospheric deposition of nutrients and organic matter, the upwelling of nutrient-rich water from southern water masses, and riverine inputs of nutrients are all important to the productivity of Arctic systems. All inputs tend to carry organic matter, and thus associated contaminants.

   The low productivity in the Arctic results in slower-growing, and thus longer-lived poikilotherms and vegetation than in more temperate climes. Slow-growing organisms can be exposed to bioaccumulating contaminants for a long period of time before being consumed at the next level in the food chain. For example, the slow growth of lichen may account for the high levels of metals and radionuclides in caribou tissues (Thomas *et al.* 1992). Homeotherms such as mammals grow at rates similar to temperate animals of the same size. However, large animals, some marine mammals and caribou for example, take a long time to reach maturity. These animals will also be exposed to bioaccumulating contaminants for a long period of time before being consumed.

3. Productivity in some biological communities in the Arctic is highly cyclic. This is due to extreme fluctuations in light levels, nutrient inputs, and temperature. Plants and animals are adapted to these fluctuations in a variety of ways. Many animals migrate or disperse to more desirable overwintering, feeding, or breeding habitats. This means that contaminant levels in some prey species and in their predators may not relate to contaminant deposition in the Canadian Arctic. Many arctic animals undergo periods of starvation in the winter. Female polar bears, for example, fast when overwintering. These periods of starvation may affect contaminant intake since animals metabolize body fats, and therefore, fat soluble contaminants. Research indicates that existing body burdens of OCs may be sequestered effectively when blubber reserves are high, but may become significant during a poor feeding season.

   Another consequence of this cyclicity is that organic carbon and associated contaminants can move through several levels of a food chain very rapidly. Nutrients and contaminants deposited on snow, ice, soil, and plants in the arctic winter are mobilized and assimilated very quickly in the spring. In the Arctic marine environment, a burst of primary productivity occurs under the ice when light levels become high in the spring. In freshwater systems, the spring melt carries nutrients and contaminants into streams, ponds, and lakes. Relatively little is left in the terrestrial environment, especially in the high Arctic.

3.2.4.2 Terrestrial Food Webs

Arctic terrestrial food webs are generally simple, often consisting of plants or lichens at the primary producer level, a few herbivores, and one or two main predators. The primary Canadian Arctic herbivores include caribou (*Rangifer tarandus*), muskoxen (*Ovibos moschatus*), ptarmigan (*Lagopus* spp.), and arctic hares (*Lepus arcticus*). Numerous small mammals also occur in the Arctic including arctic ground squirrels (*Spermophilus parryii*), voles (*Clethrionomys rutilus* and *Microtus* spp.), lemmings (*Lemmus sibiricus* and *Dicrostonyx torquatus*), and shrews (*Sorex* spp.) (Sage 1986). These herbivores are consumed by weasels (*Mustela erminea*), red foxes (*Vulpes vulpes*), arctic foxes (*Alopex lagopus*), barren-ground grizzly bears (*Ursus arctos*), wolverines (*Gulo gulo*), wolves (*Canis lupus*) and humans. The air→ plant→animal contamination pathway represented by the lichen-caribou food web is of particular interest because of the importance of caribou as a food source to many Northerners.

Shorebirds (Charadriidae and Scolopacidae), although first and second level carnivores in freshwater food webs, are preyed upon by many of the mammalian predators listed above and by birds of prey including owls (*Nyctea scandiaca* and *Asio flammeus*), eagles (*Aquila chrysaetos*), hawks (*Buteo lagopus*), and falcons (*Falco* spp.). When interpreting contaminant levels in birds, the extent of migration and feeding behaviour in the more southern overwintering habitats must also be considered.

3.2.4.3 Freshwater Food Webs

Arctic lake food webs usually include four well-defined trophic levels, with phytoplankton at the base, herbivores that include zooplankton feeding on these, detrital feeders such as benthic insect larvae and crustacea, and carnivores feeding on benthic organisms. In the NWT and northern Québec lakes, arctic char and lake whitefish (*Coregonus clupeaformis*) are the most common first-order carnivores. Several other species of whitefish (*Coregonus* spp.), lake trout, several species of salmon (*Salmo* spp.) and grayling (*Thymallus arcticus*) replace char at lower latitudes or less common habitats. Arctic predatory fish include inconnu (*Coregonus leucichthys*), lake trout, and burbot (*Lota lota*). In the high Arctic, some arctic char are cannabalistic, feeding on smaller char (Welch 1991). All of these species are important dietary sources for Northerners and are therefore of interest for monitoring of contaminant levels.

Tundra ponds that freeze to the bottom in winter do not contain fish. However the biological productivity in summer months can be high and does transfer to several food chains. Numerous herbivorous and some carnivorous invertebrates live in such ponds. The third trophic level usually consists of migratory birds.

Riverine food webs are usually similar to lake food webs in habitats of the same dimensions, but detritivorous micro-organisms and invertebrate species are more common in flowing water. In general, the same carnivorous fish species inhabit both rivers and lakes. Burbot is of particular interest because their fatty liver has proven useful for determining OCs and their relatively sedentary behavior makes them useful indicators of spatial trends. They are readily available along the Mackenzie and Yukon Rivers.

Rivers are the migration routes for many species of anadromous fish such as arctic char and coregonids, which are extensively fished during the fall migrations by local people. Also, seals and whales will move into inland lakes and rivers during the summer months.

3.2.4.4 Marine Food Webs

The inter-relationships amongst water, land, and ice result in diverse habitats for marine arctic flora and fauna. These habitats include open seas, channels, estuaries, shallow bays, deep fiords, lakes, rivers, fast ice, pack ice, ice edges, polynias and shore leads (Clarke 1993). A diagram of energy and nutrient flow, which includes all major species or taxa of interest, is shown in Figure 3.2.2. Different parts of this chain are more important in different marine areas and at different parts of the year.

Production from the epontic community is available earlier in the year than production from the phytoplankton community (Bradstreet and Cross 1982). In ice-free offshore waters, the phytoplankton-supported part of the food web dominates. The benthic component of the food web is most important in nearshore areas beyond the intertidal and ice scour zone (Clarke 1993).

Estuaries such as Hudson Bay and the Mackenzie River delta are moderately productive and support coregonids and major concentrations of beluga. The marine sub-arctic consist of seas and basins in which arctic waters mix with other Atlantic and Pacific waters or freshwaters. Because these areas are vertically unstable, nutrient upwelling from deep water occurs, and these areas can be very productive. Some important fisheries, including Atlantic cod, salmon, redfish, and capelin, occur in these areas. Areas where water masses meet or upwelling occurs, such as Lancaster Sound in the high Arctic, have relatively high productivity. Polynias, leads, and ice edges, are areas where marine mammals and sea birds concentrate (Stirling and Cleator 1981, Bradstreet and Cross 1982).

The most commonly studied component of the Arctic marine food chain is represented by the middle portion of Figure 3.2.2, leading from primary producers to top level carnivores such as sea birds,

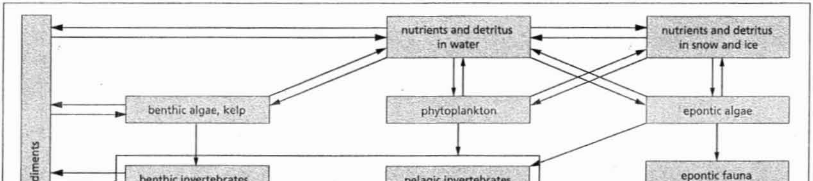


FIGURE 3.2.2

Schematic of the marine food web in the Canadian Arctic.

polar bears, and humans. Most work on contaminant bioaccumulation has focused on the invertebrate→ arctic cod (*Boreogadus saida*)→ringed seal→polar bear food chain. Detrital food webs, or webs that precede primary productivity in water, are not easily accommodated into such diagrams, but may be important in the transfer of persistent organic pollutants and heavy metals. Scientists are only beginning to recognize the potential importance of detritus and free organic carbon (Hargrave 1993).

The Lancaster Sound food web has received detailed study (Welch *et al.* 1992). Lancaster Sound, an unusually fertile part of the Canadian high Arctic, is driven by nutrient upwelling under the polar ice pack and near the ice edge. The area supports many species of birds and mammals and is utilized by four Inuit communities. The food web links through the centre of Figure 3.2.2, namely phytoplankton, pelagic invertebrates (mainly copepods), arctic cod, murres to gulls and humans, and ringed seal, narwhal, beluga, polar bear, harp seal, and walrus to humans, dominate this food web. Marine food webs in other regions of the Canadian Arctic are similar, although not as thoroughly studied as in Lancaster Sound.

Melnikov (1980) describes webs of transfer of organic material within the snow, meltwater, and ice. Detritus, organic matter from precipitation, and dead organic material are acted on by micro-organisms. The resultant nutrients released contribute to primary production. Although this route is not important in terms of the total mass of carbon involved, it may be important in the transfer of contaminants into the food chain.

At ice edges, high levels of primary production that are concentrated near the surface of landfast ice are transferred to sea birds and marine mammals via a few key taxa. This leads to relatively high concentrations of sea birds and mammals at ice edges. The ice edge food chains at Barrow Strait and Pond Inlet in the Canadian high Arctic were examined by Bradstreet and Cross (1982). Sea birds (mainly fulmars, black-legged kittiwakes *Rissa tridactyla*, and black guillemots *Cephus grylle*) and marine mammals (narwhal and ringed seals) eat arctic cod and to a lesser extent, zooplankton (calanoid copepods and *Parathemisto*), and ice-associated amphipods. Cod eat zooplankton and other ice-associated taxa, (amphipods, harpacticoid and cyclopoid copepods). Calanoid copepods and ice-associated amphipods eat primarily diatom ice algae.

As the top predators in the Arctic marine food web, marine mammals are of special interest as monitors of trends and biological effects of contaminants. The ringed seal is by far the most abundant and widely distributed resident arctic pinniped. It is a good species for contaminant monitoring because samples are relatively easily obtained with the cooperation of Inuit hunters and there is much scientific and traditional knowledge of them. Ringed seals have a preference for annual, landfast ice, but are also found in multi-year ice (Kingsley *et al.* 1985). Although ringed seals are generally territorial and philopatric, they may leave their wintering areas as ice decreases in the summer (Smith and Hammill 1981). Fish, mainly arctic cod, and crustaceans (amphipods, mysids and euphausids) are important in the diet; the proportion may vary seasonally (Bradstreet and Cross 1982, Welch *et al.* 1992). A summer diet of about 54% cod has been estimated for ringed seal in Barrow Strait using ^{15}N isotope analysis (Hobson and Welch 1992). The use of ^{15}N to characterize trophic levels is discussed in more detail in Section 3.3.1.2.

The ecology of the polar bear is closely tied to that of the ringed seals. Polar bears preferentially consume ringed seal blubber and skin. In addition to ringed seal, polar bears may eat lesser amounts of bearded seals (Stirling and Archibald 1977) and occasionally prey on beluga and walrus (Lowry *et al.* 1987, Calvert and Stirling 1990). In late spring polar bears become highly active and hyperphagic in response to readily available young and molting ringed seals. The bears acquire most of the nutrient reserves required to meet their growth and annual maintenance needs during this period (Ramsay and Stirling 1988). Using ^{13}C stable isotope analysis it has been shown that polar bears do not eat significant amounts of terrestrial food even when forced onto land by poor ice conditions (Ramsay and Hobson 1991).

3.3 Spatial Trends of Contaminants in Biota

3.3.1 Freshwater Fish and Food Webs

3.3.1.1 Spatial Trends of Contaminants in Fish in the NWT, the Yukon and Northern Québec

Reviews of contaminant data in freshwater fish from arctic and subarctic Canada available to 1991 (Lockhart *et al.* 1992, Muir *et al.* 1990a) indicated that information on the levels and geographic variation of OCs, PAHs and heavy metals was limited while data on temporal trends were nonexistent. Since then a considerable effort has been made to determine contaminant levels in fish from a broad range of lakes and rivers in the NWT, the Yukon and northern Québec. Most of the emphasis has been placed on OC chemicals such as PCBs and toxaphene as well as mercury, rather than on PAHs, because OCs and mercury are known to biomagnify in aquatic food chains. The main focus has been on piscivorous fish such as burbot, lake trout, northern pike and arctic char, because of their importance in the traditional subsistence fishery and because of interest in biomagnification in top predators. But fish feeding at lower trophic levels, such as whitefish, that are also of dietary importance to Aboriginal people in the NWT, northern Québec, and the Yukon have also been analysed.

Study design

Sampling locations for major species are shown in Fig. 3.3.1 and are listed in Tables 3.3.1, 3.3.2 and 3.3.3. Most sampling sites for fish, except a few in the high Arctic islands, were located near Arctic communities. For OCs, samples of dorsal muscle and skin were analysed with the exception of burbot, for which liver was analysed. For metals, muscle (fillet) was most frequently analysed. In the NWT and in studies carried out in northern Québec by Hydro-Québec, individual fish were analysed so that variation of contaminant levels with age, size, sex and fat content could be assessed. The sampling strategy for most lakes in the Yukon was to collect sufficient fish for six pooled samples of each of whitefish and lake trout.

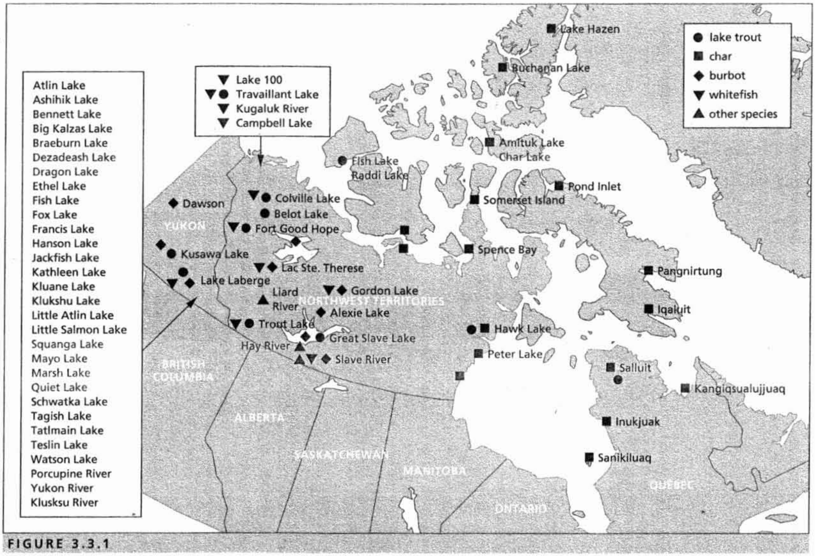


FIGURE 3.3.1

Sampling locations for freshwater and anadromous fish in the NWT, the Yukon and northern Québec (1990-95).

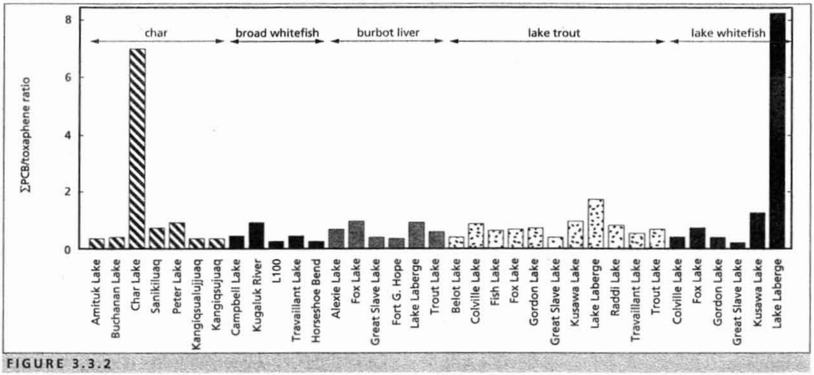


FIGURE 3.3.2

ΣPCB/toxaphene ratios in arctic char, burbot, broad and lake whitefish and lake trout in the NWT and the Yukon. Ratios > 1 are found only at sites where local contamination by PCBs is suspected.

Pooled sampling was used to increase the number of fish on which the average OC level for each location was based. In both the NWT and the Yukon, as well as in the northern Québec studies (Hydro-Québec 1993) samples from each fish were archived for future analysis. Further details on sample collection, quality assurance and analysis can be found in the annual synopsis reports of Indian and Northern Affairs Canada's Northern Contaminants Program (Muir and Lockhart 1992, 1993, 1994, 1996, Palmer 1992, 1993, 1994). Results from northern Québec are taken mainly from the large study by Hydro-Québec (1993) conducted between 1989 and 1991 in the Great Whale study area (Langlois and Langis 1995).

Organochlorines

Mean concentrations of the major OCs total PCB congeners (ΣPCB), chlordane (ΣCHLOR), total-DDT related compounds (ΣDDT), hexachlorocyclohexanes (ΣHCH) and toxaphene are listed in Tables 3.3.1 (the NWT and N. Québec) and 3.3.2 (YK) by species, region and collection site (lake/river). Toxaphene was the major OC contaminant in all fish. There are only limited results from N. Québec for toxaphene because it was not determined in the Hydro-Québec study. The prominence of toxaphene is illustrated in Fig. 3.3.2 by use of a ΣPCB/toxaphene ratio. These ratios are almost always <1 (for example, ranging from 0.12 to 0.98 in lake trout from Peter Lake (NWT) and Kusawa Lake (YK), respectively) except at locations where local PCB contamination is likely, such as Char Lake (near the Resolute airport) and in Lake Laberge.

PCBs (sum of 80–90 congeners in the results reported in Tables 3.3.1 and 3.3.2) were generally present at 1.5- to 2-fold higher levels than DDT- or chlordane-related compounds. Hexachlorocyclohexanes (sum of α-, β- and γ-HCH isomers) and chlorobenzenes were present at much lower concentrations than the four major OCs. The remainder of this section will therefore focus on PCB and toxaphene levels.

Salvelinus species (lake trout and char) are the most widely distributed species that have been analysed for OCs. A wide variation in toxaphene and PCBs is evident in lake trout in the NWT and Yukon lakes and in sea-run and landlocked char (Table 3.3.1 and 3.3.2). Highest concentrations observed so far are found in lake trout from Lake Laberge. Converting the results to a lipid weight basis to eliminate the effect of different fat content of samples from different locations does not remove this variation. Indeed, landlocked char from high arctic lakes, with very low lipid (1%–2%) content have relatively high PCB and toxaphene levels on a lipid basis. The three species of salmon sampled in the Yukon, had similar levels of OCs, with toxaphene being dominant, despite different life histories and different marine feeding areas (Table 3.3.2). Even in lakes of similar size at the same latitude (Peter, Belot and Colville; Travaillant, Raddi, Fish) there were large differences in concentrations of toxaphene and to a lesser extent of PCBs. Within lakes, age and fish weight were significantly correlated with toxaphene levels in lake trout at several locations. This is illustrated in Figure 3.3.3 in the case of lake trout from Peter Lake. The two largest trout (11.5 and 13 kg, respectively), which had high toxaphene and ΣPCB levels, were also much older (40 and 48 yrs, respectively) than all other trout analysed. The toxaphene concentrations were significantly related ($r^2 = 0.696$, $P<0.001$) to weight of lake trout. ΣPCB concentrations were also significantly correlated ($r^2 = 0.942$, $P<0.001$) with lake trout weight. Thus comparisons between lakes need to be made

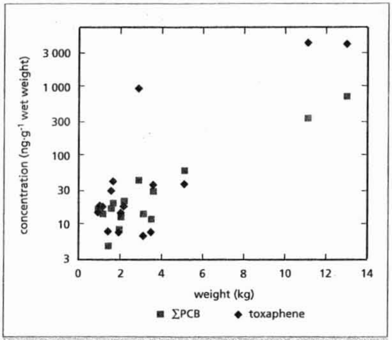


FIGURE 3.3.3

Variation of toxaphene and ΣPCB concentrations with weight of lake trout from Peter Lake (near Rankin Inlet, NWT).

with fish of similar weight. But even where size/age differences were accounted for, for example, between Colville and Belot lakes (Table 3.3.1) there were still significant differences in mean concentrations of all major OCs, which are probably due to the trophic position of the lake trout. Trout can be strictly piscivorous in some lakes and insectivorous in others (see sections 3.3.1.2 and 3.3.1.3).

Whitefish (*Coregonus* sp.) also show considerable variability even within a relatively small geographic area. For example, concentrations of PCBs and toxaphene in broad whitefish from lakes in the Inuvialuit Settlement Region were approximately 2 to 10 times higher than concentrations in samples from Horseshoe Bend (a nearby site along the Mackenzie River) (Lockhart *et al.* 1993) (Table 3.3.1). It is possible that the whitefish collected at Horseshoe Bend represent a migrating population which has been exposed to a different dietary source than the fish from the other locations. While lipid content appears to be an important variable in explaining some of the observed differences, age does not seem to be important. Fish from Horseshoe Bend were of similar mean age as those from all other locations except Lake 100. The variability of OC levels observed in whitefish muscle of similar aged fish was also observed in results for landlocked arctic char. Char from Amituk Lake on Cornwallis Island, Lake Hazen and Buchanan Lake (Axel Heiberg Island) show eleven-to-twelve-fold differences in mean concentrations of PCBs and toxaphene even though sources of contamination are probably atmospheric in all cases and thus expected to be quite similar (Muir and Lockhart 1993).

The levels of PCBs and toxaphene in broad whitefish from the Western Arctic are lower than observed for lake whitefish from Great Slave Lake and Gordon Lake in southwestern NWT (Table 3.3.1). This could be due to dietary differences, especially for Great Slave Lake whitefish, which have much higher lipid content than other Coregonids analysed. But the north-south trend is consistent with trends in OC contaminants previously observed in burbot liver collected from a series of lakes and riverine sites between northwestern Ontario and Fort McPherson (Muir *et al.* 1990a). Concentrations of PCBs (but not toxaphene or more volatile OCs such as HCH) declined significantly with increasing north latitude over the transect.

Whitefish from lakes in the Grand Baleine region also had low levels of ΣPCBs in flesh (skinless fillets), similar to those in Great Slave and Gordon Lakes (fillet + skin) when compared on a lipid basis (Table 3.3.1 and Langlois and Langis 1995). Lake trout had the highest concentrations of PCBs and other OCs of all fish sampled in the Grand Baleine basin (Table 3.3.1). Mean concentrations in lake trout flesh from Lac Bienville of 47 ng·g^{-1} wet wt. (or 3300 ng·g^{-1} lipid wt.) were similar to those in Great Slave lake trout and higher than those found in lake trout from small western NWT and Yukon lakes (Tables 3.3.1 and 3.3.2).

Liver of burbot had relatively high concentrations of PCBs and toxaphene compared with lake trout, char and whitefish muscle. This reflects the high lipid content of burbot liver. Muscle of burbot had much lower OC levels similar to those in muscle of all the other fish species examined (Table 3.3.1). The highest concentrations of PCB, toxaphene, and especially DDT-related compounds, were found in burbot from Lake Laberge. By comparison, levels of these same compounds were relatively low in other lakes on the Yukon river system. Clearly, DDT is higher at Lake Laberge and Watson Lake where extensive spraying of DDT for mosquito control was carried out from the 1940s to the 1960s. PCBs also appear to be higher in Lake Laberge than at other locations. This is most probably related to past use or disposal of PCBs in the Whitehorse area. There is some evidence for higher toxaphene levels in fish from large, glacial-fed, mountain lakes (e.g. Atlin) upstream of Lake Laberge (Table 3.3.2). Samples from the Old Crow and Porcupine rivers, in the northern Yukon, were all very low in OCs, possibly reflecting a lower rate of contaminant deposition in this part of the Yukon. Burbot from the east and west arms of Great Slave Lake had higher OC levels, especially of toxaphene, than populations in smaller lakes such as Alexie and Trout. Figure 3.3.4 summarizes results for toxaphene and PCBs in burbot liver from the northern Peace, Slave, and Mackenzie Rivers as well as from major Yukon Lakes. Concentrations of PCBs and toxaphene in burbot analysed for the Slave River study (Peddle *et al.* 1995) are similar to those reported for Great Slave Lake but much lower than levels in Lake Laberge burbot.

TABLE 3.3.1

Mean concentrations ($ng \cdot g^{-1}$ ±SD wet wt.) of major organochlorines in freshwater and anadromous fish from the NWT and northern Québec (1989-94).

Species	Location	Region	Year	Tissue[1]	N	% lipid	ΣHCH	ΣCHLOR	ΣDDT	ΣPCB	Toxaphene[2]	Reference
Arctic char (landlocked and searun)	Amituk Lake	Cornwallis Island	1991	M	12	4.4±2.1	2.05±1.12	47.3±23.5	32.2±15.2	72.5±40.4	203±118	Muir & Lockhart 1993
	Buchanan Lake	Alex Heiberg	1992	M	10	7.2±3.1	2.93±1.54	9.79±3.22	3.65±1.60	6.82±2.74	17.3±7.01	Muir & Lockhart 1993
	Char Lake	Cornwallis Island	1993	M	5	4.2±2.1	2.64±1.40	17.8±7.85	114±52.9	290±118	41.3±21.1	Muir & Lockhart 1994
	Horton Lake	Mackenzie Delta	1993	M	33	4.7±2.5	2.54±5.70	2.17±2.43	1.51±1.82	5.20±5.59	17.1±16.5	Dushenko, Unpublished 1995
	Lake Hazen	Ellesmere Island	1992	M	6	4.6±4.1	2.7±2.4	29.8±6.0	11.9±2.8	39.6±17.4	165±61.3	Muir & Lockhart 1993
	Sanikiluaq	S. Hudson Bay	1992	M	8	4.9±3.2	2.06±0.88	7.79±4.39	7.62±4.39	23.6±17.0	33.7±29.3	Muir & Lockhart 1995
	Sanikiluaq	S. Hudson Bay	1989	M	6	1.8	<0.1	1.00±0.10	0.60±0.20	1.1±0.0	—	Cameron & Weis 1993
	Peter Lake	W. Hudson Bay	1994	M	6	3.2±1.5	2.0±0.90	3.89±2.02	4.81±2.13	11.3±3.51	12.9±3.52	Muir *et al.* 1995b
	Kangiqsualujjuaq	Ungava Bay	1990	M	4	10.2±6.1	6.10±5.27	22.7±6.82	11.8±4.0	53.8±31.6	155±59.1	Muir & Lockhart 1993
	Kangiqsujuaq	Hudson Strait	1990	M	9	8.6±2.0	2.03±0.95	9.85±4.93	5.01±3.0	24.0±9.63	76.5±34.6	Muir & Lockhart 1993
Broad whitefish	Campbell Lake	Mackenzie Delta	1992	M	4	3.2±3.0	1.97±2.67	1.62±1.79	1.01±1.31	2.17±1.37	5.03±2.72	Lockhart *et al.* 1993
	Kugaluk River	Mackenzie Delta	1992	M	9	3.8±1.6	1.09±0.56	1.69±0.68	0.67±0.30	6.19±3.99	6.85±4.91	Lockhart *et al.* 1993
	Lake 100	Mackenzie Delta	1992	M	4	10.8±2.4	3.52±2.25	8.90±2.47	4.76±2.18	8.73±5.11	37.6±15.3	Lockhart *et al.* 1993
	Travaillant Lake	Mackenzie Delta	1992	M	4	2.6±0.5	0.46±0.11	0.67±0.11	0.31±0.05	1.86±0.69	4.30±0.97	Lockhart *et al.* 1993
	Horseshoe Bend	Mackenzie River	1992	M	9	3.1±0.9	0.18±0.11	1.02±0.60	0.15±0.11	0.82±0.42	3.21±1.62	Lockhart *et al.* 1993
Burbot	Alexie Lake	Yellowknife	1993	L	5	26.3±12.9	4.89±1.62	14.7±5.88	13.6±7.88	26.9±18.0	40.5±26.6	Muir & Lockhart 1994
	Great Slave Lake	East & west arm	1993-94	L	29	27.0±8.3	6.37±3.84	62.8±25.5	38.2±25.1	106±44.9	367±231	Muir & Lockhart 1994
	Great Slave Lake	Ft. Resolution	1994	M	5	5.0±1.0	0.12±0.07	0.58±0.38	0.40±0.17	1.94±0.68	2.15±1.89	Evans 1994
	Fort Good Hope	Mackenzie River	1994	L	11	31.0±13.7	9.07±3.3	24.5±6.0	39.6±19.6	56.6±18.1	169±60.5	Muir & Lockhart 1996, Evans 1994
	Trout Lake	Fort Simpson	1990	L	6	40.3±12.1	13.7±4.06	33.3±12.2	19.8±7.20	51.6±8.65	93.2±43.6	Muir & Lockhart 1994
Lake Trout	Belot Lake	Colville	1992	M	11	4.2±3.3	3.17±2.45	17.4±12.9	20.0±14.7	44.1±37.2	115±90.0	Muir & Lockhart 1994
	Colville Lake	Colville	1992	M	7	5.2±2.6	0.89±0.54	3.27±1.91	2.46±1.73	8.92±6.67	10.2±5.57	Muir & Lockhart 1994
	Fish Lake	Banks Island	1993	M	8	4.2±1.9	1.56±0.4 1	3.5±6.3	14.3±3.9	31.9±15.5	50.6±27.8	Muir *et al.* 1995a
	Gordon Lake	Yellowknife	1990	M	6	3.0±1.3	0.68±0.34	7.92±6.34	5.35±5.02	16.3±12.6	23.1±24.1	Muir & Lockhart 1994
	Great Slave Lake	East & west sites	1993-94	M	26	10.5±3.8	2.93±1.2	14.4±7.1	7.69±4.67	23.9±14.4	94.4±69.9	Muir & Lockhart 1996, Evans 1994
	Peter Lake	Rankin Inlet	1993/94	M	12	1.2±0.7	0.62±0.30	5.77±2.58	3.33±2.74	19.0±14.1	15.0±15.7	Muir *et al.* 1995b
	Raddi Lake	Banks Island	1993	M	6	4.0±2.9	1.55±0.60	2.0±4.42	17.1±7.05	33.5±9.91	42.8±18.2	Muir *et al.* 1995a
	Travaillant Lake	Mackenzie Delta	1993	M	5	3.1±0.9	1.15±0.54	4.3±1.78	2.03±0.95	8.79±3.70	17.2±21.8	Muir & Lockhart 1994
	Trout Lake	Fort Simpson	1990	M	9	4.2±2.1	1.60±1.87	7.18±7.28	3.83±3.39	11.2±6.94	17.2±21.8	Muir & Lockhart 1994
	Lac Bienville	Grande-Baleine	1989	M	6	1.5	<5.0	<5.0	6.68	47.1	nd	Hydro-Québec 1993
	Lac Bienville	Grande-Baleine	1989	L	2	6.0	4.00	<5.0	174	898	nd	
	Lac Raraire	Grande-Baleine	1989	M	2	0.7	<5.0	<5.0	<5.0	<15.0	nd	
	Lac Morpain	Grande-Baleine	1989	M	2	2.0	<5.0	<5.0	6.69	34.7	nd	
	Lac des Loups Marins	Grande-Baleine	1989	M	3	1.0	<5.0	<5.0	5.60	28.9	nd	
	Lac Amichinatwayach	Grande-Baleine	1989	M	2	1.1	<5.0	<5.0	<5.0	<15.0	nd	
Lake whitefish	Colville Lake	Colville	1992	M	9	2.6±0.8	1.05±0.71	1.85±0.83	1.71±1.39	4.44±2.10	11.0±13.4	Muir & Lockhart 1994
	Gordon Lake	Yellowknife	1990	M	5	2.74±2.45	0.92±0.91	5.54±3.67	3.72±2.77	7.94±4.35	13.9±10.5	Muir & Lockhart 1994
	Great Slave Lake	East & west sites	1993-94	M	4	18.8±3.45	6.05±2.02	25.9±3.98	8.31±1.73	22.2±2.90	104±8.65	Muir & Lockhart 1996, Evans 1994
	Grande-Baleine lakes[3]	Grande-Baleine	1989	M	3	1.0	<5.0	<5.0	<5.0	<15.0	nd	Hydro-Québec 1993
Northern pike	Lac Bienville	Grande-Baleine	1989	M	1	0.5	<5.0	<5.0	<5.0	<15.0	nd	Hydro-Québec 1993
	Lac Morpain	Grande-Baleine	1989	M	1	0.7	<5.0	<5.0	<5.0	<15.0	nd	
Cisco	Great Slave Lake	Hay River	1994	M	4	5.9±1.6	1.72±0.68	12.6±1.67	4.11±1.13	12.9±2.72	77.4±12.5	Evans 1994
Walleye	Hay River	Hay River	1994	M	3	1.19±0.49	0.16±0.08	0.65±0.20	0.61±0.26	1.4±0.32	0.40±0.19	Muir & Lockhart 1996

[1] *M = muscle + skin analysed; L = liver*

[2] *Toxaphene quantified with a single response factor*

[3] *Mean concentrations for single samples from Lac Bienville, Lac des Loups Marins and Lac Wapask*

TABLE 3.3.2

Mean concentrations (or range of means) of organochlorines in fish from Yukon lakes, 1990–94 (Palmer 1992, 1993, 1994, 1996)[1].

Species/ Tissue type	Location (number of lakes/rivers)	Sample sizes[2]	Lipid[3] %	Toxaphene $ng \cdot g^{-1}$	ΣPCB $ng \cdot g^{-1}$	ΣDDT $ng \cdot g^{-1}$	Chlordane $ng \cdot g^{-1}$	Notes
Burbot liver	Laberge Atlin other (13)	35 6 2-8	44 33 19-47	2301 1533 54-945	1267 136 50-579	3433 105 21-272	217 138 10-183	Burbot liver was consistently the highest in OCs of any fish tissue sampled. By contrast, burbot muscle, sampled for Lake Laberge, was extremely low in OC's.
Lake trout muscle	Laberge Watson other (24)	27 P 2 P 2-15 P	6.1 5.2 0.8-7.1	344 <12.9 0.9-296	448 38 3.5-128	458 3427 2.6-403	30 2.2 1.1-21	Watson Lake trout were elevated in DDT only, while Lake Laberge trout were high in all OCs.
Whitefish muscle	Laberge Watson other (17)	3 P 2 P 2-5 P	3 2.2 0.3-3.6	33.8 <11.7 0.2-52	61 6.9 0.1-8.3	212 464 0.1- 6.6	8.5 2.7 <0.1-5.1	While DDT and PCBs stand out as high in Laberge, Ethel Lake whitefish had higher toxaphene levels. Watson Lake whitefish were elevated in DDT only. Old Crow whitefish had the highest lipid levels and yet had the lowest overall OC levels.
Chinook salmon	Whitehorse, Klukshu	2-6	0.9-1.0	35-43	7.1-14	9.0-13	2.0-3.9	All samples reported here are of muscle tissue. Egg samples had slightly higher lipid and proportionately higher OCs. The three species of salmon spend most of their lives in the Pacific Ocean or Bering Strait. Toxaphene was the dominant OC.
Chum salmon	Porcupine	3	1.2	21	2.3	1.8	0.9	All samples reported here are of muscle tissue. Egg samples had slightly higher lipid and proportionately higher OCs. The three species of salmon spend most of their lives in the Pacific Ocean or Bering Strait. Toxaphene was the dominant OC.
Sockeye salmon	Klukshu	3	1	9.3	3.1	3.1	0.9	All samples reported here are of muscle tissue. Egg samples had slightly higher lipid and proportionately higher OCs. The three species of salmon spend most of their lives in the Pacific Ocean or Bering Strait. Toxaphene was the dominant OC.
Arctic Grayling muscle	Laberge other (3)	6 3-6	1.7 0.7-1.7	25 <0.2-2.7	21 <0.5-1.1	22 <0.3-0.7	1.8 <0.2-0.4	All samples had very low or non-detectable OC levels. Lake Laberge samples were about an order of magnitude higher in OC levels.
northern pike muscle	Laberge other (3)	5 P 3-6 P	1.8 0.2-0.6	48 <0.1-1.2	90 <0.1-1.1	247 <0.1- 2.5	14 <0.1-0.5	All samples except Lake Laberge were below detection or very low in organochlorines, including Hansen Lake, which was treated with toxaphene as a piscicide 30 years ago.

[1] *Results are from the database maintained by the Yukon Contaminants Committee. Study design is described in the reports by Palmer (1992-96).*

[2] *Sample sets marked with a "P" are all or mainly pooled samples, usually with about 6 fish per sample. Other samples are mainly of individual fish.*

[3] *Values of lipids and organochlorines are ranges of mean values for each location. All values are wet wt.s, i.e. the proportion of the whole wet sample that is lipid or a certain class of organochlorines.*

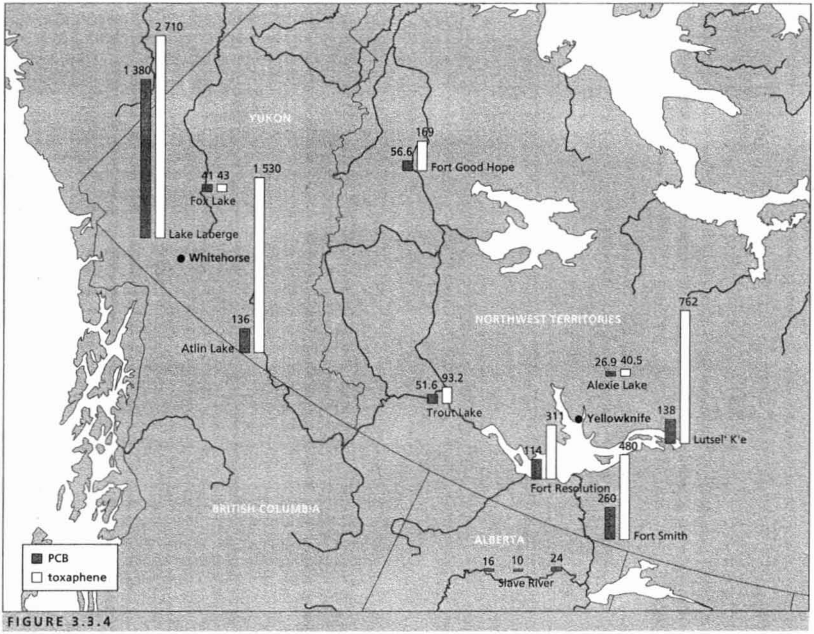


FIGURE 3.3.4

PCBs and toxaphene (ng·g^{-1} wet wt.) in burbot liver from the Peace, Slave, Mackenzie River system and from other lakes in the NWT and the Yukon. Results from the Peace River and the Slave River are from the Northern River Basin Study (Pastershank and Muir 1995) and the Slave River Study (Peddle et al. 1995).

Northern pike, like burbot, have low concentrations of most OCs in muscle (Tables 3.3.1 and 3.3.2). Hydro-Québec (1993) found much higher levels of ΣPCB and other OCs in pike liver than in muscle, in samples from the Nottoway-Broadback-Rupert rivers, which flow into James Bay. Pike liver averaged 340 ng·g^{-1} wet wt. ΣPCB versus 4.5 ng·g^{-1} in flesh.

Chlorinated dioxins and furans have been measured in small numbers of pooled samples of arctic char, lake trout, whitefish and burbot from the NWT and the Yukon (Table 3.3.3). The Slave River study has also conducted a large number of analyses of burbot liver for polychlorinated dibenzodioxins and-furans (PCDD/Fs) (Peddle *et al.* 1995). The PCDD/Fs are among the most toxic contaminants found in the environment along with the 3,4,3',4'-chlorine substituted, non-ortho substituted polychlorinated biphenyls (nPCBs). The mechanism of toxicity of this latter class of compounds is similar to that of 2,3,7,8-TCDD, but at varying potency, thus they may act in an additive fashion to exacerbate their impact on wildlife and humans. The additive effect is usually expressed as Toxic Equivalent (TEQ) concentrations of TCDD, which is the sum of the product of relative potency to TCDD times the concentration of each compound (Safe 1990, Ahlborg *et al.* 1994).

The most prominent PCDD/F congener detected in burbot and lake trout from both Lake Laberge and Great Slave Lake was 2,3,7,8-TCDF. Concentrations of 2,3,7,8-TCDD, the most toxic PCDD/F congener, were below detection limits in Lake Laberge fish and at low levels in fish from Great Slave Lake (Table 3.3.3). Another prominent congener in Yukon fish was octachlorodibenzo-*p*-dioxin (OCDD) which was present at 7.5 pg·g^{-1} (wet wt.) in lake trout muscle from Lake Laberge and 13 pg·g^{-1} in Kusawa Lake. No tetra-, penta-, or hexa-CDDs were detected in lake trout from these two lakes (detection limit <0.5 pg·g^{-1}), but burbot liver from Lake Laberge contained low pg·g^{-1} levels of hexachloro-CDDs, heptachloro-CDD and OCDD. Arctic char from locations in the central Arctic archipelago also had levels of 2,3,7,8-TCDD and-TCDF near or at detection limits (≤1 pg·g^{-1}).

TABLE 3.3.3

Concentrations of non-ortho PCBs and PCDD/Fs + S.D. in fish from the Canadian Arctic (pg·g⁻¹ wet wt.).

Location	Species	Tissues[1]	N	% lipid	CB81	CB77	CB126	CB169	2378-TCDD	2378-TCDF
Amituk Lake	Arctic char	M	12	4.4 ± 2.1	0.61 ± 0.50	7.07 ± 3.55	9.34 ± 5.61	1.14 ± 1.22	nd	nd
Broughton Island	Arctic char	W	8	11.0 ± 1.5	7.07 ± 4.97	63.1 ± 35.5	10.2 ± 5.1	0.49 ± 1.37	nd	nd
Buchanan Lake	Arctic char	M	10	7.2 ± 3.1	6.2 ± 9.0	1.92 ± 4.94	7.31 ± 7.84	0.83 ± 1.43	nd	nd
Charr Lake	Arctic char	M	5	4.3 ± 1.9	2.31 ± 0.87	22.0 ± 8.66	73.4 ± 31.1	3.38 ± 1.23	nd	nd
Kangiqsualujjuaq	Arctic char	W	6	7.8 ± 5.5	4.03 ± 3.15	23.5 ± 16.0	24.8 ± 38.5	0.35 ± 0.29	nd	nd
Lake Hazen	Arctic char	M	7	5.0 ± 2.7	<0.5	<0.5	<0.5	14.4 ± 4.1	nd	nd
Pond Inlet	Arctic char	W	10	11.7 ± 2.5	1.00 ± 0.68	6.90 ± 4.08	1.41 ± 0.80	1.52 ± 3.77	1	1
Salluit	Arctic char	W	2	8.0 ± 1.0	<0.5	24.0 ± 2.44	8.38 ± 5.75	<0.5	nd	nd
Somerset Island	Arctic char	W	8	6.5 ± 2.8	1.92 ± 1.13	10.6 ± 6.65	2.85 ± 13.9	1.73 ± 0.99	2	1
Talurjuaq (Spence Bay)	Arctic char	W	10	13.1 ± 2.7	1.71 ± 2.82	10.7 ± 13.1	1.55 ± 2.07	1.14 ± 1.04	<1	<1
Resolute Bay	Arctic cod	W	3	7.3 ± 0.5	0.31 ± 0.20	5.52 ± 1.44	0.55 ± 0.77	0.46 ± 0.53	nd	nd
Lac Laberge	Burbot	L	7	36.6 ± 11.3	54.9 ± 18.4	396 ± 97	1510 ± 888	1411 ± 906	<0.1	28
Fox Lake	Burbot	L	4	29.9 ± 11.1	2.89 ± 2.72	5.67 ± 3.74	23.8 ± 19.7	11.5 ± 3.06	nd	nd
Great Slave Lake	Burbot	L	7	35.5 ± 7.5	0.66 ± 1.39	4.97 ± 2.60	6.74 ± 2.09	10.1 ± 5.13	<0.1	2.7
Kusawa Lake	Lake trout	M	5	1.2 ± 0.5	1.01 ± 1.30	8.10 ± 1.66	18.6 ± 10.2	48.1 ± 18.9	<2	<2
Lac Laberge	Lake trout	M	6	8.4 ± 3.9	6.09 ± 6.93	60.2 ± 39.6	144 ± 183	61.8 ± 42.7	<2	3
Great Slave Lake	Lake trout	M	P	nd	nd	nd	nd	nd	0.2	1
Great Slave Lake	Whitefish	M	4	18.8 ± 3.4	0.09 ± 0.01	3.02 ± 0.57	2.57 ± 1.28	1.94 ± 1.63	0.1	0.7

[1] *M=muscle+skin; L=liver; W=whole fish. PCDD/Fs were determined in pooled samples from the same group analysed for non-ortho PCBs with the exception of Great Slave Lake trout, burbot and whitefish in which different fish from the same location (Ft. Resolution) were analysed.*

Non-ortho PCBs (nPCBs) were detectable at low concentrations in char, lake trout and burbot liver in fish from the NWT, northern Québec and the Yukon (Table 3.3.3). The highest levels were found in burbot liver and lake trout muscle from Lake Laberge. CB126 (3,4,5,3',4'-pentachlorobiphenyl) was generally the most prominent nPCB in fish tissues. The nPCB data can be converted to TEQs of 2,3,7,8-TCDD by multiplying the detected concentrations by the toxic equivalent factor (TEFs) for each congener. CB126 has the highest TEF of the nPCBs relative to 2,3,7,8-TCDD (0.1 or 1/10th the potency) (Ahlborg *et al.* 1994) and accounts for most of the TEQs in arctic fish.

Fish from the southwestern NWT regions have also been analysed for aromatic hydrocarbons as part of the Slave River study and related work (Table 3.3.4). Low concentrations of 3- and 4-ring PAHs (naphthalene, fluorene, phenanthrene, fluoranthene) were found in muscle of all species analysed. Higher molecular weight PAHs such as benzoanthracene, benzofluoranthene or benzo(a)pyrene, were not detected (<0.01 ng·g⁻¹) in muscle of any of the nine species. No spatial trends were observed in total PAH concentrations, for example, between fish from the Slave River and from surrounding lakes. Previous studies of burbot liver and muscle from the Mackenzie River also showed low ng·g⁻¹ concentrations of most 2- or 3-ring PAH and non-detectable concentrations of the higher molecular weight compounds (Lockhart *et al.* 1989). Low ng·g⁻¹ of PAHs are also found in Great Lakes fish (Lawrence and Weber 1984). Bile from long nose sucker, mountain whitefish and northern pike had much higher total PAH levels (PAH metabolites were not analysed) than muscle. This reflects the ability of fish to rapidly excrete aromatic hydrocarbons (for example, as glucuronide conjugates in the bile) and helps to explain the low levels in muscle.

Metals

Metals in fish from the Canadian Arctic have received relatively little attention, with the exception of mercury. Extensive surveys have been conducted for a few metals in fish populations near specific sources such as smelters and mines (e.g. Bohn and Fallis 1978, Moore and Sutherland 1981, Bodaly *et al.* 1984, Johansen *et al.* 1991, Harrison and Klaverkamp 1990). Recent reviews of the data available up to the late 1980s found that information on metal concentrations in subsistence fisheries was limited (Muir *et al.* 1986, Lockhart *et al.* 1992). Since 1970 substantial effort has been made to analyse mercury in fish of commercial species and size ranges. The sale of these fish is restricted in Canada if levels exceed 0.5 µg·g⁻¹. The recommended maximum is even lower (0.2 µg·g⁻¹) for people who engage in subsistence fisheries and consume larger quantities of fish (Health and Welfare Canada 1978, 1984). The Inspection Service of the

TABLE 3.3.4

Concentrations of polyaromatic hydrocarbons (ng·g^{-1} wet wt. ± S.D.) in freshwater fish from the NWT (1990-94).

Species	Tissue[1]	Location	N	Naphthalene	ΣME-naphthalene[2]	Biphenyl	Dibenzoforan	Acenaphthylene	Fluorene	Phenanthrene	Anthracene	Fluoranthene	Pyrene	Total PAH[3]
Arctic grayling	M	Ron Lake	6	2.47±1.20	2.76±1.28	0.44±0.17	0.13±0.04	0.09±0.17	0.09±0.08	0.25±0.07	0.04±0.05	0.06±0.05	0.07±0.05	5.83±2.86
	M	Carn Lake	8	2.44±0.72	3.28±1.32	0.48±0.18	0.17±0.03	0.05±0.05	0.04±0.07	0.28±0.05	0.07±0.05	0.02±0.03	0.02±0.04	6.19±2.03
Burbot	M	Slave River	10	2.13±0.89	2.72±1.52	0.45±0.19	0.15±0.06	0.01±0.02	0.01±0.03	0.21±0.09	0.05±0.05	0.03±0.05	0.06±0.05	5.21±2.53
	M	Liard River	11	1.81±0.24	2.31±0.85	0.41±0.08	0.13±0.05	<0.01	<0.01	0.16±0.04	0.04±0.03	0.01±0.03	0.05±0.06	4.37±1.07
Longnose sucker	M	Liard River	9	3.31±0.84	0.36±0.12	0.37±0.14	0.27±0.16	0.01±0.04	0.05±0.08	0.34±0.07	0.06±0.03	0.02±0.04	0.06±0.08	4.21±1.12
	B	Liard River	4	104±99.6	130±119	20.8±3.39	6.85±9.59	<0.01	<0.01	12.4±10.9	3.64±6.78	<0.01	<0.01	250±234
Lake whitefish	M	Leland Lake	10	2.66±1.19	2.72±1.48	0.16±0.07	0.30±0.10	<0.01	0.06±0.14	0.39±0.09	0.05±0.05	0.03±0.06	0.01±0.04	5.79±2.96
	M	Fort Smith	20	8.33±33.9	1.48±1.19	0.51±0.21	0.43±0.25	0.02±0.08	0.01±0.04	0.62±0.30	0.05±0.08	0.25±0.32	0.33±0.22	11.1±4.22
Lake trout	M	Lac de Gras	18	4.69±2.34	1.91±2.60	0.45±0.20	0.21±0.11	0.02±0.05	0.09±0.29	0.36±0.24	0.03±0.04	0.07±0.06	0.09±0.07	7.26±3.46
	M	Ron Lake	11	1.89±0.37	2.39±0.50	0.55±0.19	0.16±0.04	<0.01±0.00	0.04±0.07	0.25±0.05	0.04±0.04	0.02±0.04	0.06±0.07	4.70±0.91
	M	Carn Lake	11	2.58±0.80	3.92±1.47	0.54±0.13	0.17±0.03	0.07±0.07	0.17±0.08	0.27±0.06	0.03±0.04	0.02±0.03	0.02±0.03	7.08±2.24
Mountain whitefish	WF	Liard River	10	3.18±0.90	3.60±4.46	2.64±3.97	1.05±1.11	0.08±0.12	0.51±0.59	1.71±2.03	0.09±0.10	0.14±0.11	0.25±0.33	9.68±7.46
	B	Liard River	2	24.5	30.7	7.3	1.3	<0.01	3.02	10.3	<0.01	<0.01	1.38	70.0
Northern pike	M	Liard River	10	2.24±0.45	2.87±0.95	0.55±0.13	0.25±0.05	0.01±0.03	0.03±0.09	0.27±0.06	0.05±0.04	0.09±0.05	0.10±0.06	5.67±1.52
	B	Liard River	2	26.1	35.3	7.24	2.69	<0.01	<0.01	3.57	<0.01	<0.01	<0.01	65.0
	M	Fort Smith	10	2.24±0.53	0.43±0.60	0.11±0.11	0.09±0.07	<0.01	<0.01	0.23±0.04	0.03±0.06	0.02±0.04	0.04±0.10	2.99±0.81
	M	Slave River	9	2.82±0.77	3.29±2.04	0.18±0.05	0.11±0.08	<0.01	<0.01	0.24±0.04	0.11±0.03	0.02±0.07	0.07±0.11	6.55±2.80
	M	Hay River	5	2.24±1.24	0.27±0.20	0.20±0.07	0.21±0.09	0.01±0.03	<0.01	0.33±0.99	0.04±0.04	0.14±0.02	0.12±0.06	3.27±1.63
	B	Hay River	3	39.4±8.14	39.5±11.9	5.87±10.2	3.57±3.13	<0.01	<0.01	5.35±3.05	<0.01	<0.01	<0.01	84.2±22.7
Round whitefish	M	Lac de Gras	21	3.55±1.68	2.91±3.29	0.52±0.34	0.30±0.13	0.03±0.04	0.03±0.06	0.30±0.12	0.03±0.03	0.07±0.05	0.14±0.08	7.34±4.42
	M	Cam Lake	10	2.72±0.92	2.30±0.67	0.59±0.20	0.18±0.03	0.02±0.04	0.06±0.07	0.22±0.04	0.03±0.05	0.01±0.02	0.03±0.05	7.27±2.30
	M	Ron Lake	10	2.14±0.69	4.19±1.40	0.38±0.09	0.17±0.06	0.07±0.08	0.10±0.12	0.19±0.03	0.04±0.03	0.05±0.05	0.08±0.05	4.97±1.45
Walleye	M	Liard River	14	2.11±0.53	2.96±0.48	0.55±0.17	0.21±0.08	<0.01	<0.01	0.25±0.06	0.02±0.04	0.03±0.06	0.07±0.07	5.44±1.06
	B	Liard River	4	175± 251	216±327	41.0±62.5	8.59±13.12	<0.01	<0.01	16.5±21.4	<0.01±0.00	<0.01	<0.01	408±599
	M	Fort Smith	19	5.57±2.86	0.84±0.65	0.23±0.18	0.22±0.13	0.01±0.06	<0.01	0.36±0.19	<0.01	0.06±0.12	0.06±0.12	6.91±3.72
	M	Leland Lake	7	1.93±0.48	1.86±0.40	0.16±0.11	0.19±0.08	<0.01	<0.01	0.25±0.07	0.06±0.08	0.05±0.13	0.05±0.12	4.20±1.07
	B	Hay River	3	80.8±57.8	78.7±54.0	25.1±25.1	8.40±6.97	<0.01	4.00±6.93	10.7±8.23	<0.01	<0.01	1.94±1.84	176±119

[1] *M=Muscle; B=bile; WF=whole fish*

[2] *Sum of 1- and 2-methylnaphthalene*

[3] *Total of 25 PAHs (not including retene and perylene)*

TABLE 3.3.5

Mean concentration of metals in muscle of freshwater and anadromous fish (µg·g^{-1} wet wt.) in the NWT, Yukon and northern Québec .

Species	Location	Region	Year	N[1]	Cd µg·g^{-1}	Cu µg·g^{-1}	Zn µg·g^{-1}	Hg[2] µg·g^{-1}	Se µg·g^{-1}	Reference
Arctic char (anadromous)	Queen Maud Gulf Area	Queen Maud Gulf	1993	20	—	—	—	0.062	—	DFO 1994
	Dease Strait Area	Dease Strait	1993	10	—	—	—	0.027	—	" "
	Hall Beach/Gore Bay	Foxe Basin	1992	10	—	—	—	0.038	—	" "
	Pangnirtung Fiord	Cumberland Sound	1992	10	—	—	—	0.035		" "
	Sylvia Grinnell River	Frobisher Bay	1991	5	—	—	—	0.082		" "
	Rankin Inlet-Arviat area	W. Hudson Bay	1993	25	—	—	—	0.064		" "
	Eastern Hudson Bay	Eastern Hudson Bay	1994	9	0.003			0.057	0.48	" "
Arctic char (landlocked)	NN Lake	Cornwallis Is.	1992	3	—	—	—	0.26	—	Lockhart 1994
	Char Lake	Cornwallis Is.	1992	7	—	—	—	0.26	—	" "
	Resolute Lake	Cornwallis Is.	1992	7	—	—	—	0.20	—	" "
	Small lake	Cornwallis Is.	1992	2	—	—	—	0.05	—	" "
Brook trout	Grand Baleine	N. Québec	1989-90	19	0.01	0.31	6.25	0.16	0.63	Hydro Quebec 1993
Lake Trout	Colville Lake	NW NWT	1993	11	0.0009	1.21	3.08	0.28	0.17	Muir & Lockhart 1994
	Grand Baleine River	N Québec	1989-90	106	0.01	0.35	3.28	0.71	0.79	Hydro-Quebec 1993
	Hawk Lake	W. Hudson Bay	1991	9	—	—	—	0.24	0.27	Muir & Lockhart 1993
	Lac Belot	NW NWT	1993	23	0.0013	1.43	3.07	0.13	0.10	Muir & Lockhart 1994
	Lac Ste-Thérèse	NW NWT	1992	11	—	—	—	0.95	—	DIAND 1994
	Peter Lake	W. Hudson Bay	1994	18	—	—	—	0.67	—	Muir *et al.* 1995a
	Fox Lake	S. Yukon	1992	17	—	—	—	0.36		Lockhart & Kidd 1994
	Kusawa Lake	S. Yukon	1992	11	—	—	—	0.36		" "
	Lake Laberge	S. Yukon	1992	8	—	—	—	0.30		" "
Lake Trout	Aishihik Lake	SW Yukon	1992	4	—	1.05	11.7	0.09	—	Yukon DRR, 1994
	Canyon Lake	SW Yukon	1992	4	—	0.60	11.7	0.16	—	" "
	Kloo Lake	SW Yukon	1992	4	—	0.52	8.0	0.53	—	" "
	Mayo Lake	Central Yukon	1992	5	—	0.20	11.7	0.11	—	" "
	Sekulmun Lake	SW Yukon	1992	4	—	1.04	12.5	0.46	—	" "
Lake Whitefish	Colville Lake	NW NWT	1992	23	0.001	1.07	3.07	0.02	0.14	Muir & Lockhart 1994
	Hay River	S. NWT	1988-90	40	—	—	—	0.07	—	Grey *et al.* 1995
	Lac Ste-Thérèse	NW NWT	1992	11	—	—	—	0.82	—	DIAND 1994
	Slave River	S. NWT	1988-90	30	—	—	—	0.08	—	Grey *et al.* 1995
	Grand Baleine River	N. Québec	1989-90	41	<0.02	0.38	4.0	0.14	0.66	Hydro-Quebec 1993
	Fox Lake	S. Yukon	1992	10	—	—	—	0.23	—	Lockhart & Kidd 1994
	Kusawa Lake	S. Yukon	1992	14	—	—	—	0.10	—	" "
	Lake Laberge	S. Yukon	1992	32	—	—	—	0.09	—	" "
	Aishihik Lake	SW Yukon	1992	5	—	0.61	7.30	0.04	—	Yukon DRR 1994
	Kloo Lake	SW Yukon	1992	5	—	0.26	17.8	0.11	—	" "
	Mayo Lake	Central Yukon	1992	5	—	0.14	16.0	0.06	—	" "
	Sekulmun Lake	SW Yukon	1992	4	—	14.0	14.2	0.09	—	" "
Northern Pike	Grand Baleine River	N. Québec	1989-90	6	<0.05	0.27	3.80	0.63	0.58	Hydro-Quebec 1993
	Hay River	S. NWT	1988-90	21	—	—	—	0.32	—	Grey *et al.* 1995
	Slave River	S. NWT	1988-90	63	—	—	—	0.34	—	Grey *et al.* 1995
	Lac à Jacques	NW NWT	1994	22	—	—	—	0.44	—	Lockhart 1994
	Fox Lake	S. Yukon	1992	6	—	—	—	0.50	—	Lockhart & Kidd 1994
	Aishihik Lake	SW Yukon	1992	5	—	0.59	10.8	0.13	—	Yukon DRR, 1994
	Kloo Lake	SW Yukon	1992	5	—	0.28	12.7	0.20	—	" "
	Mayo Lake	Central Yukon	1992	2	—	0.25	6.10	0.11	—	" "
	Sekulmun Lake	SW Yukon	1992	4	—	0.50	10.8	0.19	—	" "
Walleye	Lac à Jacques	NW NWT	1994	11	—	—	—	0.98	—	Lockhart 1994
	Lac Ste-Thérèse	NW NWT	1992	30	—	—	—	1.34	—	DIAND 1994
	Hay River	S. NWT	1988-90	35	—	—	—	0.22	—	Grey *et al.* 1995
	Slave River	S. NWT	1988-90	99	—	—	—	0.34	—	Grey *et al.* 1995

[1] *Number of fish analysed for mercury. Generally every fifth sample was analysed for other metals.*

[2] *Total Hg*

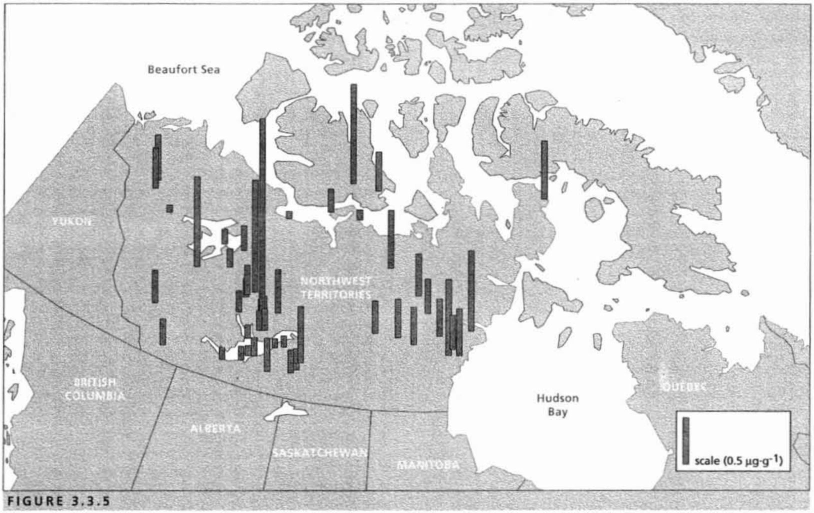


FIGURE 3.3.5

Mercury levels (µg·g^{-1}) in lake trout muscle samples from NWT lakes (from DFO Inspection Service data, DFO 1994). The bar heights indicate the mean total mercury levels relative to the Health Canada Criterion of 0.5 µg·g^{-1} and the base of each bar shows the approximate location.

Department of Fisheries and Oceans (DFO 1994) has analysed samples of northern commercial fish for mercury for the past 25 years. Their results indicate that a number of lakes in the NWT have fish populations with levels exceeding the guidelines. Figure 3.3.5 displays the results from Fish Inspection surveys of lake trout over the period between 1971 and 1992.

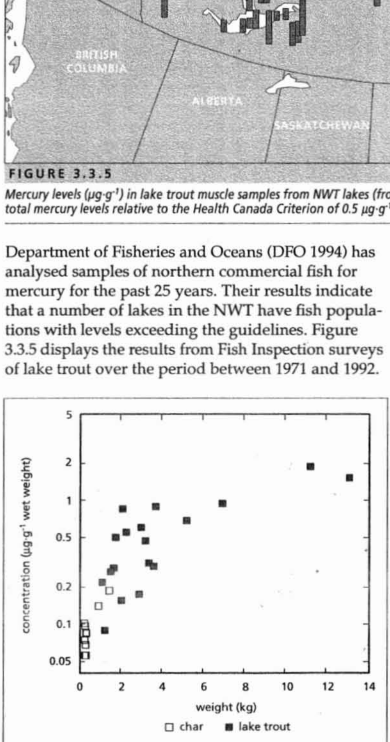


FIGURE 3.3.6

Variation of total mercury (µg·g^{-1} wet wt.) in muscle with size (kg) of arctic char and lake trout from Peter Lake (near Rankin Inlet, NWT).

Mercury levels are high in lake trout throughout the NWT and northern Québec, that is, levels frequently exceed 0.5 µg·g^{-1}, the guideline limit for commercial fish and 0.2 µg·g^{-1} the guideline for subsistence fish consumption. There is substantial lake-to-lake variation in the same species. For example, in the western NWT lake whitefish from Lac Ste-Therese have about 40 times higher levels than those from Colville Lake (Table 3.3.5).

Walleye, lake trout and pike generally have the highest mercury levels. By contrast uniformly low levels of mercury are found in sea run arctic char (Table 3.3.5). Some landlocked char can have elevated mercury. A survey of mercury levels in arctic char from lakes on Cornwallis Island was carried out following the observation of high mercury in char from Amituk Lake (Muir and Lockhart 1993). Mean concentrations in char muscle from two lakes exceeded the 0.2 µg·g^{-1} guideline for subsistence fisheries but were well below the 0.5 µg·g^{-1} level (Table 3.3.5). High mercury levels in lake trout are typically associated with larger, older fish. In Peter Lake (near Rankin Inlet) mercury concentrations in lake trout and char were significantly related to fish weight (Figure 3.3.6) but were only weakly correlated with age of the fish (Muir *et al.* 1995a).

A large number of fish from northern Québec have been analysed for mercury and other metals as part of environmental studies for the Grand Baleine project (Hydro-Québec 1993, Langlois and Langis 1995). The highest concentrations of mercury were found in lake trout and northern pike flesh and the lowest in lake whitefish (Table 3.3.5). Lake trout and lake whitefish in the Grand Baleine region had concentrations of mercury and other metals similar to those in trout and whitefish from large NWT lakes (Belot, Peter, Colville) (Table 3.3.5). Mercury concentrations in northern pike muscle from the Grand Baleine region were higher than in pike from most NWT or Yukon locations. Selenium was present at similar concentrations to mercury in lake trout and pike while selenium/mercury ratios were >1 in whitefish and brook trout (Table 3.3.5).

Relatively few measurements have been made on the forms of mercury in arctic fish. In their survey of mercury in southwestern NWT fish, Grey *et al.* (1995) found that methylmercury concentrations were 88% of total mercury. Whitefish had lower methylmercury/total mercury ratios than walleye or northern pike.

3.3.1.2 Biomagnification of Organochlorines in Lake Laberge

Unusually high concentrations of toxaphene in fish from Lake Laberge (YK) resulted in the closure of an important commercial, sport and native subsistence fishery in 1991. Levels of toxaphene and other OCs such as DDT and PCBs in Laberge fish were up to 125 times higher than those found in the same species from other regional lakes (Kidd *et al.* 1993, Kidd and Schindler 1994, Muir and Lockhart 1992, 1993).

The fish community structure in Lake Laberge is known to be different from other Yukon lakes, with few lake trout and lake whitefish, and many burbot and long nose sucker. The lake trout are also high in lipid and are solely piscivorous, unlike trout populations from other regional lakes. Rasmussen *et al.* (1990) found that lake trout from lakes with longer food webs had higher concentrations of persistent contaminants. It was hypothesized that the different food web structure of Lake Laberge resulted in elevated concentrations of atmospherically deposited contaminants in the fish.

To investigate this hypothesis, Kidd *et al.* (1995a, b) characterized the food web relationships of Laberge, and two reference lakes, Fox and Kusawa, using stable nitrogen isotope ratios ($\delta^{15}N$) and fish stomach content analyses. The heavier isotope of nitrogen (^{15}N) is enriched 3 to 5 parts per thousand (‰) in an organism compared with its diet (see review by Peterson and Fry 1987), and is used as continuous measure of trophic level. Several species of fish, and benthic and planktonic invertebrates were collected between 1992 and 1994 and analysed for $\delta^{15}N$ and OCs using the methods outlined in Kidd *et al.* (1995a, b).

Lake trout and burbot were the top predators in each of the lakes, with mean $\delta^{15}N$ ranging from 8 to 12 ‰ between sites (see Figure 3.3.7). $\delta^{15}N$ indicated that the lake trout and burbot from Lake Laberge feed at a higher trophic position than do the same species from Fox and Kusawa Lakes. Mean $\delta^{15}N$ of lake and round whitefish, cisco and long nose sucker ranged between 6.5 and 8 ‰, a trophic level lower than the top predators in each lake. Results obtained from the stable nitrogen isotope measurements were consistent with dietary information acquired from stomach content analyses of the fish.

Concentrations of toxaphene (ng·g^{-1} wet wt.) were found to correlate significantly with the $\delta^{15}N$ of organisms through the food webs of Laberge, Fox and Kusawa Lakes (Figure 3.3.7). The regression equations are as follows:

Laberge: Toxaphene $= 0.24(\pm 0.03)\ \delta^{15}N - 0.44\ (\pm 0.26)$;
$r^2 = 0.62, n = 40, p = 0.03$

Fox: Toxaphene $= 0.19(\pm 0.08)\ \delta^{15}N - 0.70(\pm 0.63)$;
$r^2 = 0.25, n = 18, p < 0.01$

Kusawa: Toxaphene $= 0.24(\pm 0.04)\ \delta^{15}N - 0.55(\pm 0.31)$;
$r^2 = 0.67, n = 24, p < 0.01$

The slopes of these regressions are not significantly different (ANCOVA, $p<0.05$). Similar relationships between other OCs (i.e., DDT, PCBs, HCH) and $\delta^{15}N$ were found in all three lakes with the slopes of the regressions increasing with the apparent biomagnification potential of the compounds (Kidd *et al.* 1995a). The slope of the $\delta^{15}N$ to organochlorine relationship provides a measurement of biomagnification.

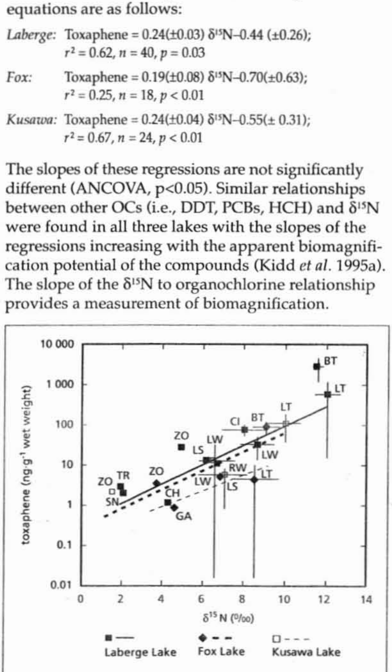


FIGURE 3.3.7

Mean (±SD) of $\delta^{15}N$ versus toxaphene (ng·g^{-1} wet wt., n = 1 to 14) for whole invertebrates and fish muscle (burbot liver used for toxaphene analyses) from Laberge (— solid line), Fox (--- heavy dashed line), and Kusawa (- - - fine dashed line) lakes, Yukon Territory.
Organisms are identified as follows: LT - lake trout, BT - burbot, LW - lake whitefish, RW - round whitefish, CI-cisco, LS - long nose sucker, ZO - zooplankton, GA - gammarus, CH - chironomid, SN - snail, and TR - tricopteran. Reprinted from Kidd et al. *1995a.*

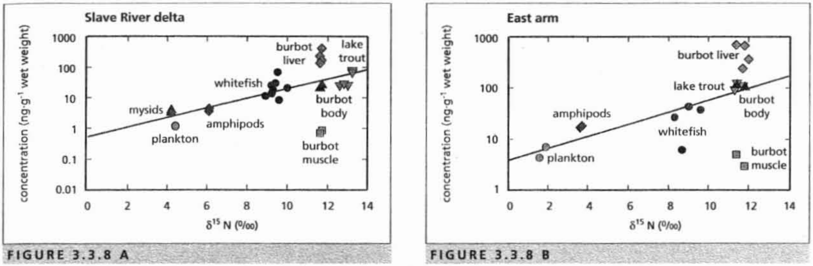


FIGURE 3.3.8 A

FIGURE 3.3.8 B

Bioaccumulation of toxaphene in the Great Slave Lake food web (▲ - mysids, ● - plankton, ◆ - amphipods, ● - whitefish, ◆ - burbot liver, ▼ - lake trout, ▲ - burbot body, ■ - burbot muscle) with increasing trophic level indicated by $\delta^{15}N$.

Results indicated that there are differences in the food web structures of Laberge, Fox and Kusawa Lakes. The top predators in Laberge feed at a higher trophic level than those from the reference lakes, a factor that has been previously implicated in higher contaminant concentrations in lake trout (Rasmussen *et al.* 1990). This trophic difference between lakes provides an explanation for the higher concentrations of toxaphene, DDT and PCBs in the fish from Lake Laberge. $\delta^{15}N$ appears to have significant potential to be used as a predictor of OC concentrations in freshwater biota.

3.3.1.3 Biomagnification of Organochlorines in Great Slave Lake

Great Slave Lake is unique in that, unlike most other large lakes in the central subarctic it receives significant riverine input via the Slave River. The Slave River, in turn, is formed by the confluence of the Peace and Athabasca rivers, which have extensive watersheds and anthropogenic activities in their drainage basins. The commercial, sports and subsistence fisheries on Great Slave Lake are very important and the majority of NWT residents live in communities on or near the lake. Yet contaminant measurements of Great Slave Lake fish, other than for the commercial export fishery, were very limited prior to 1991. Recent studies have determined OC in plankton, amphipods, mysids, white fish (muscle), lake trout (muscle), and burbot (liver, muscle), in two regions of the lake — the Slave River Delta region (West Basin) and the East Arm, a region isolated from the Slave River influence (Evans 1994). The contaminants data are combined with measurements of stable isotopes of N (see Sect. 3.1.2) to examine the biomagnification of OCs in the food web. Studies have also been made of heavy metal contaminants in the Fort Resolution area of Great Slave Lake (Peddle and Lafontaine 1995).

In general, OC concentrations in fish (Table 3.3.1) were within the range of concentrations found in other regions of the subarctic and Arctic. ΣPCB and ΣDDT concentrations did not attain the high values observed in lake trout, burbot, and whitefish from Lake Laberge in the Yukon. Nevertheless, toxaphene concentrations in whitefish were within the range reported for Lake Laberge and higher than reported for Fox and Kusawa lakes. Toxaphene concentrations in burbot liver were lower than in Lake Laberge but higher than in Trout and Alexie lakes. Toxaphene concentrations in lake trout muscle were lower than in Peter Lake and Lake Laberge, but higher than in Trout and Colville lakes.

Comparisons with invertebrates are more problematic because fewer determinations have been made. In general, PCB, toxaphene, and DDT concentrations in plankton collected from Great Slave Lake (Evans 1994) were similar to values observed in Fox and Kusawa lakes in the Yukon but were not as high as in Lake Laberge (Kidd *et al.* 1995a,b). PCB and DDT concentrations in Great Slave Lake amphipods were similar to concentrations observed in Fox Lake, while toxaphene concentrations were higher. Toxaphene concentrations also were higher in amphipods from Great Slave Lake than benthos from Lake Laberge.

Toxaphene, PCB, and DDT concentrations tended to be higher in whitefish (muscle), lake trout (muscle) and burbot (muscle and liver) collected from the East Arm than those collected in the West Basin. Regional differences may be associated with regional differences in the age and lipid concentration of fish. These hypotheses are under investigation. Contaminant concentrations also tended to be higher in invertebrates collected from the East Arm than those from the West Basin. Higher concentrations in invertebrates in the East Arm are possibly due to the fact that the Slave River, because of its heavy suspended sediment load, may rapidly scavenge aerially transported contaminants from the water column and dilute sedimentary concentrations. It also suggests that the Slave River does not contribute significantly to the organic contaminant body burden of West Basin biota.

There was remarkably little variation in carbon isotopes for the food web investigated in the Slave River delta region: this suggests that phytoplankton production was the primary carbon source for the West Basin — that is, there was no obvious river input of terrestrial carbon. The carbon isotope data were substantially more variable in the East Arm (Lutsel K'e), suggesting that there were a variety of carbon sources. Such sources probably included leaf litter and macrophytes (*Chara*) in addition to phytoplankton production. In addition, lake trout, burbot, and whitefish appeared to be feeding on substantially different carbon sources in the East Arm than in the Slave River Delta region.

Nitrogen was used to infer trophic relations in the two regions of Great Slave Lake (Figure 3.3.8). Plankton and mysids had similar ^{15}N values in the Slave River Delta region. Moreover, there was a progressive enrichment in ^{15}N from amphipods to whitefish to lake trout, reflecting trophic feeding. Burbot muscle had slightly lower ^{15}N values than lake trout, possibly reflecting different food sources for the two species of fish inhabiting the West Basin. Similar trends were observed in the East Arm, although ^{15}N values were lower in plankton and amphipods than in the Slave River Delta region. The reasons for these differences are under investigation.

OC concentrations were strongly associated with trophic level as inferred from the ^{15}N and toxaphene data (Figure 3.3.8). Moreover, the regressions for toxaphene concentration versus ^{15}N differed for the two regions of the lake. The two regressions were as follows:

Slave River Delta Log Toxaphene $= 0.16\ ^{15}N - 0.281$ $(r^2 = 0.37)$

East Arm Log Toxaphene $= 0.12\ ^{15}N + 0.572$ $(r^2 = 0.33)$

Slopes for the two regressions were similar although a little less steep than the 0.19–0.24 slope observed for lakes Laberge, Fox, and Kusawa (Kidd *et al.* 1995 a, b). Thus, organic contaminants may not biomagnify as strongly in Great Slave Lake food webs as in Yukon lake food webs.

The intercepts of the regression lines were substantially different for the two regions of Great Slave Lake, primarily because toxaphene concentrations tended to be higher in the plankton and amphipods collected from the East Arm than in the Slave River delta region. In addition, the regression was affected by the fact that ^{15}N values tended to be lower for amphipods and plankton collected from the East Arm than from the Slave River delta. The reasons for this are uncertain. Kidd *et al.* (1995b) observed intercepts of –0.44 to –0.70 for the three study lakes. Reasons for differences in the intercepts in Yukon lakes and Great Slave Lake are under investigation. Differences may, in part, be related to the fact that toxaphene concentrations were higher in Great Slave Lake amphipods than the benthic invertebrates examined by Kidd *et al.* (1995b).

TABLE 3.3.6

Range in heavy metal and organochlorine contaminant concentrations[2] in fish from the Slave River at Fort Smith, 1990–93 (Peddle *et al.* 1995).

Species/Chemical	Collection Years	Tissue[1]	N	Non-Detects[3]	Range
Walleye – Slave River at Fort Smith					
Arsenic	90, 91, 92, 93	M	52	33	<0.01–0.09
Cadmium	90, 91, 92, 93	M	52	51	<0.01–0.02
Copper	90, 91, 92, 93	M	52	1	<0.01–.99
Lead	90, 91, 92, 93	M	52	39	<0.01–0.13
Mercury	90, 91, 92, 93	M	52	0	0.05–0.6
Nickel	90, 91, 92, 93	M	52	40	<0.05–0.85
Zinc	90, 91, 92, 93	M	52	0	2.9–8
ΣDDT	90, 91, 92, 93	W	40	29	<0.002–0.021
Toxaphene	90, 91, 92, 93	W	40	24	<0.002–0.137
2,3,7,8-TCDD	90, 91, 92, 93	W	40	30	<0.063–2.56
2,3,7,8-TCDF	90, 91, 92, 93	W	40	17	<0.013–2.34
ΣPCB	90, 91, 92, 93	W	40	0	0.013–0.118
Burbot liver – Slave River at Fort Smith					
Arsenic	91, 92, 93	L	31	11	<0.025–1.76
Cadmium	91, 92, 93	L	31	5	<0.03–0.33
Copper	91, 92, 93	L	31	0	3.86–16.5
Lead	91, 92, 93	L	31	24	<0.025–0.09
Mercury	91, 92, 93	L	31	2	<0.0025–.0.05
Nickel	91, 92, 93	L	31	30	<0.125–0.38
Zinc	91, 92, 93	L	31	0	12.3–28.5
ΣDDT	90, 91, 92, 93	L	41	0	0.005–0.286
Toxaphene	90, 91, 92, 93	L	41	0	0.061–1.89
2,3,7,8-TCDD	90, 91, 92, 93	L	41	16	<0.06–16
2,3,7,8-TCDF	90, 91, 92, 93	L	41	1	<1.3–45
ΣPCB	90, 91, 92, 93	L	41	0	0.013–0.99

[1] *M=muscle; W=whole fish; L=liver*

[2] *Concentrations in* $\mu g \cdot g^{-1}$ *wet wt., except for TCDD and TCDF in* $pg \cdot g^{-1}$ *wet wt.*

[3] *Number of samples below detection limit.*

3.3.1.4 Slave River Study

The Slave River is a transboundary river that drains an area of approximately 600 000 km^2, nearly all of which is located in the provinces of Alberta, British Columbia and Saskatchewan. The NWT portion of the river basin is located downstream of all three jurisdictions. This portion of the Slave Basin is important as a direct source of drinking water. In addition, the aquatic environment is important to several aspects of a northern subsistence lifestyle as it provides habitat for wildlife species, including fish and is culturally and spiritually significant (Peddle *et al.* 1995). The Slave River Environmental Quality Monitoring Program conducted between 1990 and 1995 was a multimedia transboundary sampling program designed to characterize the baseline aquatic ecosystem conditions in the Slave River at Fort Smith, NWT (Peddle *et al.* 1995).

Water, suspended sediment and fish samples were collected for organic and inorganic contaminant analyses including PCDD/Fs, PCBs and persistent OC pesticides such as toxaphene. Water, sediment and fish were collected from the Slave River at Fort Smith, NWT below the Rapids of the Drowned. Two control sites were chosen: Leland Lake for walleye (*Stizostedion vitreum*), pike (*Esox lucius*) and whitefish (*Coregonus clupeaformis*) and Chitty Lake for burbot (*Lota lota*). Muscle, bile, liver and/or whole fish samples were collected from each species. Further details on sampling and analysis in the Slave River Study are given by Peddle *et al.* (1995).

In walleye (whole fish) and burbot liver, 2,3,7,8-TCDD and 2,3,7,8-TCDF isomers generally made up 100% of the total TCDD and TCDF detected (Table 3.3.6). TCDF levels were approximately five times higher than TCDD levels in burbot liver from the Slave River. This TCDF/TCDD ratio suggested that the major source of these contaminants was from bleached kraft mill discharges. The 2,3,7,8-TCDD and -TCDF levels in walleye whole fish were similar for both the Slave River and the control lake. In burbot liver, however, the levels were higher in the Slave River than in the control lake.

The detection of 2,3,7,8-TCDD and -TCDF isomers in samples indicate that these compounds are present in the environment of the Slave River and biologically available for accumulation. Levels in burbot liver (wet wt. basis) are higher than those in walleye whole fish samples. This is likely due to the much greater lipid content of burbot liver (typically 30%–50%) compared to walleye whole fish (1%–10%).

Toxaphene and PCBs were the predominant OC contaminants in walleye and burbot liver from the Slave River and the control lakes. A wide range of toxaphene concentrations were found in burbot liver (Table 3.3.6) but, in general, concentrations were similar to levels observed in burbot liver from Great Slave Lake (Table 3.3.1).

3.3.1.5 Geographical Trends of PCBs in Freshwater Zooplankton

Net plankton have proven to be useful biomonitors of PCBs and OC pesticides in lakes (Taylor *et al.* 1991). A study by Koenig (1996) examined geographical and temporal variation of PCBs and OC pesticides in a series of 19 arctic lakes ranging from 63° to 79°N (Figure 3.3.9). Samples from Amituk Lake were also collected four times during the open water season in 1994 to examine temporal trends. All samples were collected during July and August 1993 using a 150 µm mesh net. Samples were freeze-dried and analysed for PCB congeners and other OCs. The highest ΣPCB concentrations were observed in the northern-most lakes on Cornwallis Island and Eastern Ellesmere (Figure 3.3.9). The lowest ΣPCB levels were found in samples from the Wager Bay area of northwestern Hudson Bay and in samples from Great Slave Lake and Yukon lakes. These regional differences could be related in part to variations of zooplankton biomass and of lipid content between lakes. Taylor *et al.* (1991) found an inverse relationship between OC concentrations and plankton biomass in 33 Ontario lakes.

3.3.1.6 Overall Assessment of Contaminants in Freshwater Environments

This overview of contamination of fish in the NWT, Yukon and Northern Québec has shown that mercury is the one contaminant consistently exceeding guideline limits for subsistence consumption or commercial sale. Lake trout and northern pike in the Canadian Shield lakes of the NWT and Northern Québec generally have the most elevated levels. Levels of other heavy metals are generally not elevated in fish. The occurrence of elevated mercury levels has been known since the 1970s. However, recent studies have increased our understanding considerably. The sources of mercury, and other metals, to arctic fish are both natural and anthropogenic. Evidence for a natural source of mercury for fish comes from the large lake-to-lake variation within the same species and from the fact that selenium is generally present at similar molar concentrations as mercury. The presence of selenium at equimolar concentrations, may render mercury less bioavailable through formation of insoluble mercury selenides and offer protection against mercury poisoning. If the atmosphere was the only source, as it generally is for OCs, less lake-to-lake variation might be expected. Mercury is a natural constituent of rocks and soils in lake watersheds and is present in arctic lake sediments (Lockhart *et al.* 1995). On the other hand, there is likely an anthropogenic component to the mercury contamination in fish because concentrations in lake sediments, especially in the Keewatin district and the central Arctic archipelago, have increased steadily

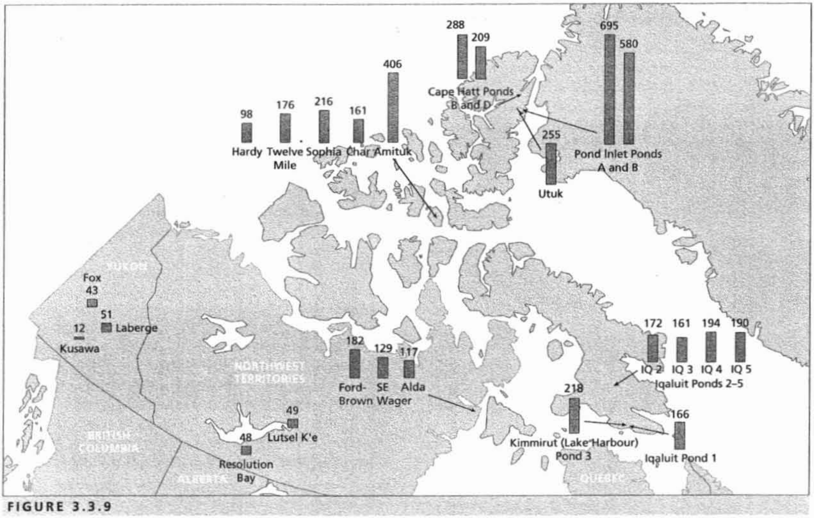


FIGURE 3.3.9

Geographic trends in mean concentrations of ΣPCBs in zooplankton (>100 μm) in the NWT and Yukon lakes. Data for Great Slave Lake from Evans (1994); Yukon lakes from Kidd and Schindler (1994) and all other lakes from Koenig (1996). Concentrations (ng·g^{-1} dry wt.) are indicated above the bars.

this century due to atmospheric input (Lockhart *et al.* 1995).

Toxaphene is the major OC contaminant in all fish analysed. The analysis of toxaphene is less accurate than for other OCs because of a lack of analytical standards for individual hepta- to nonachlorobornanes, the major components of toxaphene. Nevertheless there has been sufficient interlab comparison within the Northern Contaminants Program (see Muir and Lockhart 1992) and by Health Canada (Andrews 1994) to suggest that toxaphene concentrations reported here are relatively accurate. With the exception of Hansen Lake in the Yukon, which was treated with toxaphene in the 1950s to eradicate "coarse" fish, there are no known point sources of toxaphene in the Canadian Arctic. The source is therefore likely atmospheric and, as a result relatively similar levels might be expected in fish over a wide geographic area. However, this is not the case. Highest toxaphene levels are generally seen in fish that are strictly piscivorous such as lake trout and burbot in Lake Laberge and burbot in the east arm of Great Slave Lake. Toxaphene, as well as mercury and PCBs, also show significant correlation with fish size. Elevated toxaphene and mercury co-occur in lake trout in Peter Lake, but in general the correlation of mercury with toxaphene or other OCs is not strong.

The reason for the high concentrations of toxaphene observed in burbot liver may be the fact that the liver is very large, accounting for some 15% of the body weight versus some 4% in lake trout. Thus, proportionately more of the contaminant body burden may be stored in burbot liver than in fish such as lake trout. A whole body toxaphene burden in burbot can be estimated assuming that concentrations in the muscle were representative of all tissues except the liver and that the liver accounted for 15% of the wet wt. In Great Slave Lake this brings the whole body concentrations for burbot from Fort Resolution (23 ng·g^{-1}) in line with concentrations in lake trout muscle (49 ng·g^{-1}) for the West Basin (and presumably close to the whole body concentration for lake trout).

There now exists a relatively large dataset on chlorinated dioxins and furans in fish from the Slave River and more limited measurements for fish in Great Slave Lake and Yukon lakes. TCDD levels are low (typically <1 pg·g^{-1}) in comparison with levels in fish sampled near bleached kraft mills (Servos *et al.* 1994) or to species in the lower Great Lakes (DeVault *et al.* 1989, Whittle *et al.* 1995). Fewer measurements have been made of the toxic non-ortho substituted PCBs in arctic fish, but in general concentrations are an order of magnitude higher than those of PCDD/Fs. Where non-ortho PCBs have been measured along with PCDD/Fs, calculation of TCDD TEQs shows that non-ortho PCBs, especially CB126 account for most of the TEQs. Future measurements of toxic planar OCs can therefore be limited to non-ortho PCBs unless specific sources of PCDD/Fs (such as waste PCB oils or pentachlorophenol use) are suspected. It should be noted that no measurements have been made of many other planar OCs such as chlorinated naphthalenes, chlorinated diphenyl ethers or their brominated analogs in arctic fish.

PAHs such as naphthalene and phenanthrene were detectable in muscle of fish from southwestern NWT but only at low $ng \cdot g^{-1}$ concentrations. There were no discernable differences in mean concentrations between fish from the Slave, Liard or Hay Rivers, which drain hydrocarbon-bearing oil sands areas to the south, and fish from isolated lakes that do not access these rivers. Therefore the low molecular weight PAHs are probably of biogenic origin or the result of atmospheric transport and deposition of these relatively volatile compounds. Concentrations of total PAHs in fish from this region were similar to those in the muscle of Great Lakes fish. Much higher levels of PAHs in bile of fish indicates that they are exposed to hydrocarbon sources but are excreting the hydrocarbons either as untransformed compound or metabolites.

Differences in levels of bioaccumulating contaminants, such as PCBs, in lake trout in Ontario lakes have been attributed to food chain length and trophic status of lakes (Rasmussen *et al.* 1990). Analyses of muscle ^{15}N in lake trout from Peter Lake have shown that they occupy a higher trophic level (as defined by their nitrogen isotope ratios) than lake trout from Yukon lakes except Lake Laberge (Kidd and Schindler 1994). Toxaphene concentrations within the food webs of all lakes examined so far (Fox, Kusawa, Laberge and Great Slave lakes), correlate significantly with ^{15}N.

^{15}N values were similar in burbot from Great Slave Lake and Lake Laberge suggesting that burbot feed at similar positions in the food webs in both lakes. Nevertheless, toxaphene concentrations were substantially lower in Great Slave Lake burbot liver, averaging 244 $ng \cdot g^{-1}$ for the Slave River Delta and 762 $ng \cdot g^{-1}$ for East Arm burbot, compared with 2,820 $ng \cdot g^{-1}$ observed for Lake Laberge. This suggests that trophic level alone cannot account for the high toxaphene concentrations in Lake Laberge burbot liver. There were also differences in concentrations of toxaphene in invertebrates between Laberge and Great Slave. It is difficult to assess whether these inter-lake differences in contaminant concentration reflect true differences between the systems or are more a reflection of the inherent variability in such measurements. The resolution of this is important because the concentrations of organic contaminants in fish appear to be a function not only of trophic level but of other aspects of the lake ecosystem. Thus, while there is some evidence of similarities in the slope of contaminant concentration as a function of trophic level (^{15}N) for a variety of lakes, the intercepts differ (see below). Variations in the intercept will affect variations in the contaminant concentrations in fish.

Although a north-south trend in PCB concentrations (but not for toxaphene or more volatile OCs such as HCH) was previously observed in burbot liver collected from a series of lakes and riverine sites between northwestern Ontario and Fort McPherson (Muir *et al.* 1990a), no geographic trends can be discerned within other species from within NWT and the Yukon. This is probably due to the confounding effects of fish size and food chain length.

The geographical coverage of contaminant measurements is very good in the Yukon, where most major subsistence fisheries have been surveyed, but is much less detailed in the NWT and northern Québec. In the NWT, most measurements have been done in fish from Great Slave Lake and nearby smaller lakes, as well as in the Mackenzie River and delta. Contaminant analysis of freshwater fish in the Nunavut region of NWT and the Nunavik area of northern Québec is limited mainly to arctic char and is insufficient to assess spatial trends. Further sampling of other subsistence fish in Nunavut and Nunavik should be a priority in the future.

- *Mercury is the one contaminant consistently exceeding guideline limits for subsistence consumption or commercial sale. Lake trout and northern pike in the Canadian Shield lakes of the NWT and northern Québec generally have the most elevated levels.*

- *Toxaphene is the major OC contaminant in all fish analysed. Highest toxaphene levels are generally seen in fish that are strictly piscivorous such as lake trout and burbot in Lake Laberge and burbot in the east arm of Great Slave Lake. Toxaphene, as well as mercury and PCB concentrations also tend to increase with increasing fish size.*

- *Chlorinated dioxins and -furans have been determined in fishes from the Slave River, and in Great Slave Lake and some Yukon lakes. Levels of 2,3,7,8-TCDD are low (typically <1 $pg \cdot g^{-1}$) in comparison with levels in fish sampled near bleached kraft mills or to species in the lower Great Lakes.*

- *Toxic non-ortho substituted PCBs are generally present at 10x higher levels than PCDD/Fs. Where non-ortho PCBs have been measured along with PCDD/Fs, calculation of TCDD TEQs shows that non-ortho PCBs, especially CB126, account for most of the TEQs. Future measurements of toxic planar OCs can therefore be limited to non-ortho PCBs unless specific sources of PCDD/Fs (such as waste PCB oils or pentachlorophenol use) are suspected.*

- *The geographical coverage of contaminant measurements is very good in the Yukon, where most major subsistence fisheries have been surveyed, but is much less detailed in NWT and northern Québec. Contaminant analysis of freshwater fish in the Nunavut region of NWT and the Nunavik area of northern Québec is limited mainly to arctic char and is insufficient to assess spatial trends. Further sampling of other subsistence fishes in Nunavut and Nunavik should be a priority in the future.*

3.3.2 Terrestrial Mammals and Birds

Information on levels of organochlorine, heavy metal and radionuclide contaminants in terrestrial wildlife species in the Canadian Arctic and subarctic regions was very limited prior to 1991, and data on temporal trends were virtually nonexistent (Wong 1985, Thomas *et al.* 1992). The limited data indicated that a variety of contaminants were present in terrestrial species but it was too limited to derive any conclusions about spatial and temporal trends, or degree of contamination of most species. Although the levels of the contaminants detected were substantially lower in terrestrial species than in marine mammals, the importance of species such as caribou (*Rangifer tarandus*) as a major country food species in communities across the Canadian north makes the determination of baseline residue data particularly important. It has been shown that long-lived ungulates such as moose (*Alces alces*) and caribou can accumulate significant levels of some heavy metals such as cadmium (Froslie *et al.* 1986, Scanlon *et al.* 1986). Caribou have a winter diet consisting primarily of lichen, which accumulate nutrients and contaminants from the air. Caribou are a staple source of food for people and predators, such as wolves, in the north and their potential level of contamination is therefore of great concern. Waterfowl, terrestrial game birds, shorebirds, sea birds and their eggs are also harvested for consumption in the north (Coad 1994), and a survey of contaminants in harvested avian species in the Canadian Arctic has been a part of a larger national survey since 1988. Other terrestrial species such as mink and wolves are top trophic species that may serve as sensitive indicator species to assess trends in terrestrial environment contamination and ecosystem health.

3.3.2.1 Spatial Trends in Barren-ground and Woodland Caribou

Caribou in the NWT and the Yukon are important sources of food for northern people as well as a key component of the terrestrial food web. Both the barren-ground caribou (*R.t. groenlandicus*) of the NWT and woodland caribou (*R.t. caribou*) in the Yukon have a winter diet consisting primarily of lichens (Kelsall 1968, Parker 1978). Lichens accumulate contaminants more readily than other plants because of their large surface area, longevity and ability to bind heavy metals. Lichens are a slow-growing plant and accumulate atmospheric contaminants in a non-selective manner, resulting in a contaminant load similar to atmospheric input through long-range atmospheric transport (Thomas *et al.* 1992). The defined ranges and pan-arctic distribution of caribou make it a good species for examining geographical variations in terrestrial contaminant deposition and for monitoring spatial and temporal trends in arctic terrestrial ecosystem contamination.

Study design

Baseline levels and spatial trends of OC, heavy metal and radionuclide contaminants were assessed in 10 major barren-ground caribou herds across the NWT (Elkin and Bethke 1995). Three woodland caribou herds in the Yukon were assessed for heavy metals and other elements, as well as OC contaminants (Gamberg and Mychasiw 1994a). Chlorinated dioxins and -furans were also determined in seven of 13 herds (Hebert *et al.* 1995). Barren-ground caribou collections were conducted in cooperation with local Hunters and Trappers Organizations at Arviat, Cambridge Bay, Cape Dorset, Fort Smith, Inuvik, Kimmirut (formerly known as Lake Harbour), Pond Inlet, Southampton Island, Taloyoak, Dettah and Ndilo. In the Yukon female woodland caribou, from each of the Finlayson, Tay and Bonnet Plume herds, were sampled in cooperation with local First Nations. Tissue samples from the NWT herds were analysed for 63 organochlorine residues (43 PCB congeners and 20 pesticides) in fat, liver and muscle, 10 heavy metals in liver and kidney, and 7 radionuclides in muscle. In the Yukon samples, liver, kidney, bone and muscle samples were taken for inorganic analysis, which included 23 elements plus three heavy metals (cadmium, lead, mercury). Fat and muscle from the Finlayson herd were also analysed for 23 organochlorines and Cesium-137 (^{137}Cs), respectively. Further details on sample collection and analysis can be found in Elkin and Bethke (1995), Elkin (1994), Gamberg (1993), Gamberg and Mychasiw (1994a).

Organochlorines

A wide range of OC contaminants were detected in barren-ground caribou. Most compounds were found at very low levels and with the more volatile and less persistent compounds, HCB and HCH, predominating (Figure 3.3.10), as seen in other terrestrial herbivores in the Canadian north (MacNeil *et al.* 1987, Thomas *et al.* 1992, Salisbury *et al.* 1992). Total HCH (ΣHCH) ranged from 3.3 ng·g^{-1} (lipid corrected) in fat from Inuvik caribou to 40 ng·g^{-1} at Kimmirut, and consisted almost entirely of α-HCH. HCB residues

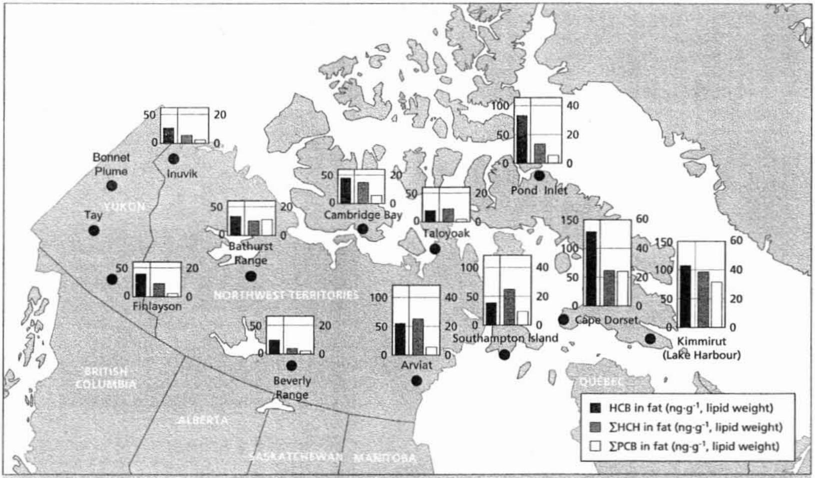


FIGURE 3.3.10

Geographic trends in concentrations (ng·g^{-1} lipid wt.) of HCB, ΣHCH, ΣPCBs in caribou from NWT locations. Scale on right side of graphs applies to ΣHCH and ΣPCB; scale on left to HCB only.

ranged from a mean of 20 ng·g^{-1} in fat of Taloyoak caribou to 129 g·g^{-1} in Kimmirut animals. HCB and α-HCH were detected in fat samples from all caribou at all sites. HCB and HCH have been shown to be the predominant OC contaminants in arctic air (Hargrave *et al.* 1988, Patton *et al.* 1989, Gregor and Gummer 1988), which suggests a direct air-plant-animal pathway into the terrestrial food chain (Thomas *et al.* 1992). Woodland caribou (Finlayson herd) have similar ΣHCH and HCB levels to the western NWT herds (Figure 3.3.10).

Oxychlordane, a major metabolite of several compounds in the pesticide technical chlordane, was the major chlordane-related compound detected in caribou liver. Total chlordane levels ranged from 0.11 ng·g^{-1} in Inuvik caribou to 5.0 ng·g^{-1} at Cape Dorset, with oxychlordane and to a lesser degree heptachlor epoxide predominating. The extent of transformation of chlordane in caribou is evident by the fact that *cis-* and *trans-*chlordane were only minor contaminants in all fat samples, although they form a major proportion of technical chlordane. Total DDT, comprised largely of *p,p'*-DDE, was below detection limits in Inuvik caribou, and ranged from 0.11 ng·g^{-1} in Beverly caribou to 2.6 ng·g^{-1} at Cape Dorset. The levels found in this study were consistent with those found in Baffin Island caribou by Muir *et al.* (1988a). Total toxaphene levels found in 12 pooled fat samples from four sites ranged from non-detectable to 23 ng·g^{-1}.

ΣPCB residues (sum of 43 congeners) ranged from a mean of 1.0 ng·g^{-1} (lipid corrected) in fat of Inuvik caribou to 32 ng·g^{-1} at Cape Dorset (Figure 3.3.10). The CB congener patterns were similar at all sites, with greater accumulation of more highly chlorinated congeners. CB-153 was bioaccumulated to the greatest extent at all sites. The results were consistent with Muir *et al.* (1988a) who found that caribou muscle had low levels of a broad spectrum of congeners that resembled PCB mixtures more closely, while six or seven congeners made up the major portion of total PCB in marine mammals. Higher levels of trichlorobiphenyl and tetrachlorobiphenyl compounds found in the caribou, relative to other terrestrial mammals and birds, may be a result of the direct air-lichen-caribou food chain. These findings suggest that the source of contamination in terrestrial herbivores is airborne PCBs which are absorbed or deposited on plant surfaces. Regional differences in PCB, HCH and DDT distribution in caribou herds appear to be more marked than for the other OC compounds, possibly reflecting a North American origin (Thomas *et al.* 1992).

Heavy metals

Barren-ground caribou had moderately elevated levels of cadmium in both kidney and liver tissue. Levels in kidney tissue ranged from 7.49 µg·g^{-1} dry weight in Taloyoak caribou to 42.7 µg·g^{-1} at Inuvik (Figure 3.3.11), and levels in liver ranged from 0.98 µg·g^{-1} at Pond Inlet to 5.83 µg·g^{-1} at Inuvik. Cadmium

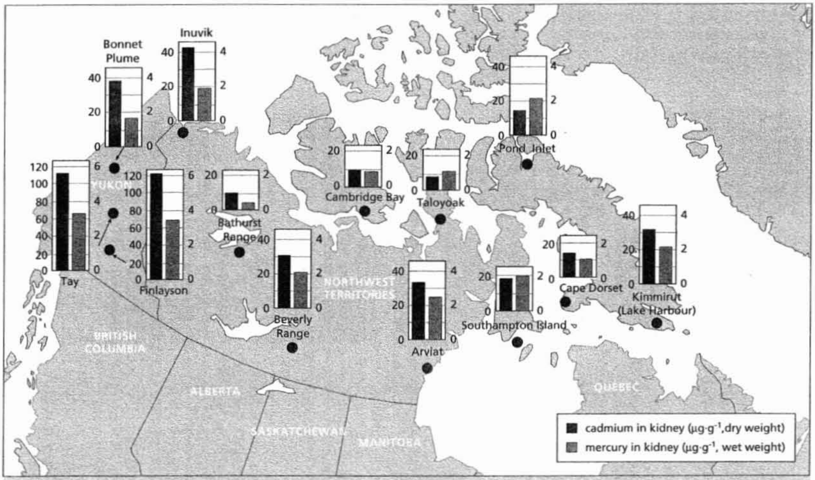


FIGURE 3.3.11

Geographic trends in concentrations of cadmium and mercury in caribou from the NWT.

levels found in livers and kidneys of the Bonnet Plume woodland caribou herd were similar to levels seen in the barren-ground caribou herds in the NWT. The Finlayson and Tay caribou herds, however, show markedly higher cadmium levels than those other herds (Figure 3.3.11). Cadmium concentrations increased with age in the Tay herd, but showed little variation with age in the Finlayson and Bonnet Plume herds (Figure 3.3.12).

The cadmium levels detected in the NWT herds, and in the Bonnet Plume herd from the Yukon, are comparable to those found in caribou from northern Québec and reindeer in Norway (Froslie *et al.* 1986, Shaw and Gunn 1981, Scanlon *et al.* 1988, Crête *et al.* 1987, 1989) and moose and whitetailed deer (*Odocoileus virginianus*) from Ontario and Newfoundland (Glooschenko *et al.* 1988, Brazil and Ferguson 1989). These cadmium levels are considered normal because of the relatively high levels on a circumpolar basis, however, they exceed Canadian environmental quality guidelines (30 $\mu g \cdot g^{-1}$) for wildlife (Environment Canada, 1996).

Although cadmium levels in the Tay and Finlayson herds were higher than those in the more northerly Yukon herd (Bonnet Plume) and in the NWT and Québec herds, lichens from the Bonnet Plume, Tay and Finlayson ranges did not differ in their cadmium content. All of the lichens sampled in the Yukon had cadmium concentrations lower than those measured in Québec, where the caribou are far less contaminated (Crête *et al.* 1989). Levels in Yukon lichens averaged 0.04 $\mu g \cdot g^{-1}$ cadmium whereas lichens from Québec averaged 0.17 $\mu g \cdot g^{-1}$. Additional lichen samples taken from areas frequented by caribou within the Finlayson range showed the same uniformly low cadmium levels. This spatial trend in lichen contaminant levels indicates that the source of high cadmium in the caribou is probably not via

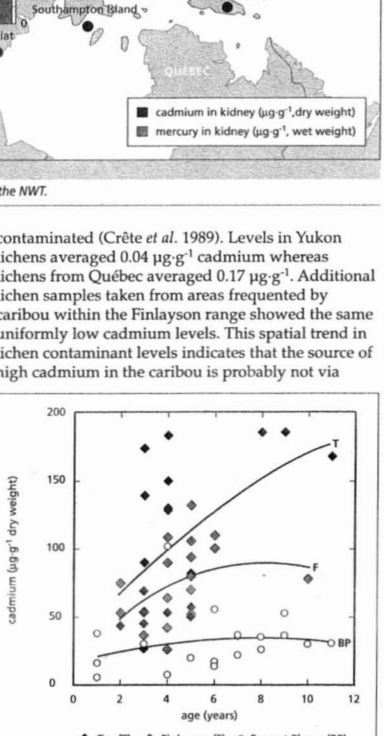


FIGURE 3.3.12

Relationship of cadmium levels in kidneys to age of the animal for three Yukon woodland caribou herds.

ingestion of lichen. Another potential source of cadmium could be the water the caribou drink. It is a commonly held local belief in the Yukon that caribou seek out overflow (slush or water) on frozen lakes for the minerals. However, overflow sampled on five Yukon lakes that showed recent caribou use in the Finlayson range had cadmium levels below the detection limits of 0.01 $mg \cdot L^{-1}$. Surface soil sampled from the Bonnet Plume and Tay ranges was uniformly low in cadmium (0.33–0.60 $\mu g \cdot g^{-1}$). Only soil from the Finlayson range showed elevated levels of cadmium, and even here, the concentrations tended to differ markedly among sites (1.07–3.82 $\mu g \cdot g^{-1}$ cadmium). This suggests not only high levels of natural cadmium in the area, but also that concentrations show great local variation depending on the site. Considering the high levels of cadmium found in stream sediments throughout this whole area (Friske and Hornbrook 1988) including both the Finlayson and Tay home ranges, it is quite reasonable to suspect that soil in the Tay range is similar to the Finlayson in its variability in cadmium concentrations, but that sample numbers (N=3) were not high enough to reflect this variation.

The source of high cadmium in the Finlayson and Tay caribou has not yet been identified, but other work in southeast Yukon has shown cadmium levels up to 22 $\mu g \cdot g^{-1}$ (dry wt.) in winter twigs of willow (*Salix* sp.) and 13 $\mu g \cdot g^{-1}$ in balsam poplar (*Populus balsamifera*). These contrast with a maximum of 1 $\mu g \cdot g^{-1}$ in dogwood (*Cornus stolinifera*) twigs from the same area (M. Gamberg, unpublished, data 1995). Cadmium accumulating in plants such as these, which are winter forage for woodland caribou, combined with naturally high concentrations of cadmium in soil, are the likely source of high cadmium concentrations in the Tay and Finlayson herds.

Mercury levels were generally low in all caribou, with means in kidney ranging from 0.52 $\mu g \cdot g^{-1}$ in Bathurst caribou to 3.94 $\mu g \cdot g^{-1}$ in the Finlayson herd (Figure 3.3.11). Liver mercury concentrations were generally lower than those in kidney, ranging from 0.16 $\mu g \cdot g^{-1}$ to 0.92 $\mu g \cdot g^{-1}$ in the NWT herds (Elkin and Bethke 1995). Methylmercury was measured in the kidney samples from the 1992 Finlayson collection (total mercury averaged 4.07 $\mu g \cdot g^{-1}$), and all levels were less than the detection limit of 0.1 $\mu g \cdot g^{-1}$ (wet wt.). This indicates that virtually all of the mercury present is in the relatively non-toxic inorganic form. The area occupied by the Finlayson and Tay caribou herds in particular is extremely rich in minerals, so it is not surprising to find some metals at elevated levels in these caribou. The mercury levels in the NWT and Yukon caribou were slightly higher than levels found in livers of reindeer from Sweden and Norway, which had levels up to 0.19 $\mu g \cdot g^{-1}$ and

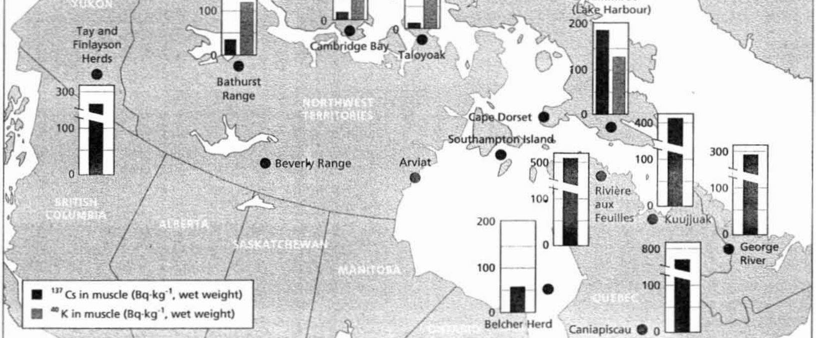


FIGURE 3.3.13

Radionuclides in caribou muscle from the NWT, the Yukon and northern Québec. Results from Elkin and Bethke (1995), Elkin (1996) and Marshall and Tracey (1989).

0.24 μg·g^{-1}, respectively (Froslie *et al.* 1984, Eriksson *et al.* 1990).

Radionuclides

Radionuclide levels were generally low or non-detectable in caribou muscle, with the exception of cesium – 137 (^{137}Cs), cesium – 134 (^{134}Cs) and potassium – 40 (^{40}K) (Figure 3.3.13). ^{137}Cs was the most predominant radionuclide in the NWT herds, with activity ranging from 2.83 Bq·kg^{-1} at Inuvik to 184 Bq·kg^{-1} in Kimmirut caribou. Higher ^{137}Cs levels were found in northern Québec herds in the late 1980s (Marshall and Tracey 1989). Levels of ^{134}Cs ranged from <0.37 Bq·kg^{-1} in Bathurst caribou to <0.71 Bq·kg^{-1} in Cambridge Bay animals. ^{137}Cs and ^{134}Cs are fission products, and most cesium in the Canadian Arctic originated from atmospheric testing of nuclear and thermonuclear devices that began in 1955 and peaked between 1961 and 1963. The levels of ^{137}Cs measured in caribou muscle during the 1960s and 1980s indicate that concentrations in caribou decreased considerably following the moratorium on atmospheric weapons testing in 1963 when levels reached 2000 Bq·kg^{-1} in several NWT herds (Thomas *et al.* 1992). The results from this study are at the lower range of values found from 1986 to 1988, consistent with the view of declining ^{137}Cs levels in the Arctic. The levels of ^{134}Cs in this study were very low, and most likely derived from the Chernobyl nuclear power plant accident, but much lower than observed in reindeer in northern Europe after Chernobyl.

Naturally occurring radionuclides potassium – 40 (^{40}K), lead – 210 (^{210}Pb) and polonium – 210 (^{210}Po), were also detected in the NWT and Yukon caribou herds (Macdonald *et al.* 1996). ^{40}K, a naturally occurring radionuclide with a very long half-life, was also consistently found in all samples at about 100 Bq·kg^{-1}, similar to levels reported for moose in southern Manitoba (Zach *et al.* 1989). Highest levels of ^{210}Pb were found in bone and large differences in concentrations were observed between individuals within the same herd and between herds. The Kimmirut herd had the highest ^{210}Pb and ^{210}Po levels but consistent geographical trends were not observed. Macdonald *et al.* (1996) concluded that radiation doses for Kimmirut caribou, due mainly to ^{210}Po, were high relative to other mammals in the Canadian environment. The animals have presumably adapted to the radiation stress from these natural sources but potential effects of the high doses on the animals are not clear.

Chlorinated dioxins and -furans

A systematic study to quantify concentrations of PCDDs, PCDFs and nPCBs in caribou from across the Canadian North was initiated in 1993 to complement the measurements of other OCs, metals and radionuclides. No data previously existed to document levels of these compounds or TEQs in arctic terrestrial wildlife. The study made use of existing collections of subcutaneous back fat from female caribou in the NWT (Elkin and Bethke 1995). Only one sex was chosen to facilitate comparison among herds. The collections had been made from four herds: Southampton Island, Cape Dorset, Kimmirut and Bathurst. Individual samples were pooled for chemical analysis. In addition, liver, muscle and subcutaneous back or kidney fat samples were taken from 20 female caribou from each of the Finlayson, Tay and Bonnet Plume herds from the Yukon Territory. Fat samples from the Finlayson herd were analysed to determine if there was a relationship between age and PCDD/PCDF levels. Individual samples were pooled according to age. Pooled liver and muscle samples from the Finlayson Age 3 group (a subset of the age 2–6 group in Table 3.3.7) were also analysed to determine contaminant distribution in tissue. The samples were analysed for 27 PCDD and PCDF congeners plus five nPCBS. Further details on sample collection and analysis are given in Hebert *et al.* (1995).

A large number of PCDD/F congeners was determined because it was hypothesized that non-2,3,7,8-substituted congeners, which predominate in the atmosphere, may accumulate in caribou because of the short air-lichen-caribou pathway. Accurate TEQs could be calculated only for caribou from the NWT because data for the mono-ortho-substituted PCBs, IUPAC #105 and #118, were available only from these herds (Elkin and Bethke 1995). The average contribution of CB-105 and CB-118 in the NWT herds was 12%. The TEQs from the Yukon herds were corrected by this percentage to account for the missing CB-105 and CB-118 data.

The data are summarized as total of each isomer group for PCDDs and PCDFs, and individual nPCBs in Table 3.3.7. Levels of PCDDs, PCDFs and nPCBs were extremely low in animals from all herds. Only 2,3,7,8-substituted PCDDs were observed. TCDD was found in only two fat samples (Cape Dorset, 0.73 pg·g^{-1} and Kimmirut, 0.14 pg·g^{-1}) despite very low detection limits ranging from 0.02 to 0.43 pg·g^{-1}. OCDD was the only PCDD that was found in the majority of fat samples, at levels ranging from <0.38 pg·g^{-1} to 4.69 pg·g^{-1} in the Cape Dorset sample. Among PCDF congeners, 1,2,4,7,8-PnCDF was present in all samples except Finlayson at levels ranging from 0.24 to 0.74 pg·g^{-1} . Five other non-2,3,7,8-substituted PCDF congeners, 2,3,6,8-TCDF, 2,3,6,7-TCDF, 1,2,4,6,8-PnCDF, 1,2,4,6,7,8-HxCDF and 1,2,4,6,8,9-HxCDF, were found only in the Kimmirut sample at levels < 1 pg·g^{-1}.

The larger number of PCDD/PCDF congeners detected in two of the NWT herds may reflect differences in the long-range atmospheric transport of these compounds to the western and eastern Arctic.

This is consistent with the observation that OCDD was the predominant PCDD/PCDF congener found in all samples. Highly chlorinated dioxins, such as OCDD, are usually indicative of combustion-related sources (Broman *et al.* 1991), which probably arrive in the Arctic via long-range atmospheric transport (Norstrom *et al.* 1990). The finding of traces of non-2,3,7,8-PCDFs in the Kimmirut sample is also consistent with an atmospheric signal. No detectable PCDDs or PCDFs were found in the Finlayson muscle sample. However, the detection limits were in the 2–18 pg·g^{-1} range because of the low lipid content of the sample (2%).

Non-ortho PCBs were present at low concentrations in all of the caribou from the Yukon and Northwest Territories (Table 3.3.7). PCB congeners #126 and #169 showed some spatial variability with higher levels in the eastern Arctic. This result corroborates the findings of higher concentrations of ortho-substituted PCBs in the eastern Arctic (Figure 3.3.10).

TCDD TEQs on a wet weight basis were calculated for each pooled sample (Table 3.3.7). The TEQ levels found in all of the caribou tissues are extremely low, generally less than 1 pg·g^{-1}, maximum 3.29 pg·g^{-1}. Only Cape Dorset and Kimmirut samples exceeded 1 pg·g^{-1}. By comparison, TEQs in Arctic marine mammals, such as the beluga and narwhal (*Monodon monoceros*), range from approximately 100–500 pg·g^{-1} (Norstrom and Muir 1994). The greatest contribution to overall TEQ levels was from the nPCBs, with CB 126 being of particular importance. Their mean contribution to TEQ levels in fat tissue from all of the herds was greater than 70%.

3.3.2.2 Contaminant Transfer in the Lichen/Caribou/Wolf Food Chain

Caribou are strict herbivores that have a winter diet made up primarily of lichen. In Arctic areas of the NWT, caribou has been shown to be the predominant food item of wolves (*Canis lupus*). This short and simple food chain provides an excellent opportunity to model and quantify the transfer of contaminants through three trophic levels to the top of the chain. To evaluate the transfer and biomagnification of specific contaminants through the lichen→ caribou→ wolf food chain, samples were collected near Yellowknife (Bathurst herd), Cambridge Bay (Victoria Island herd), and Inuvik (Bluenose herd) and analysed for OC, heavy metal and radionuclide contaminant levels.

Study design

Field collections of barren-ground caribou were conducted in cooperation with local Hunters' and Trappers' organizations from Cambridge Bay, Inuvik, and Ndilo/Fort Rae (see Figure 3.3.10). Wolf carcasses were collected by local hunters from these communities, and were submitted frozen for processing and sampling. For both caribou and wolves, samples were collected from 20 animals. Stomach contents were collected from the caribou and wolves for diet analysis. Teeth were collected for aging purposes, and a variety of biological and morphometric measurements were taken. Samples of three common lichen species (*Cladina mitis, Cladina rangiferina, Cetraria nivalis*) that are important in the caribou diet were selected for

TABLE 3.3.7

Concentrations (pg·g^{-1}, lipid wt.) of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and non-ortho substituted PCBs (nPCBs), as well as TCDD TEQs (wet wt.) in female fat collected from three caribou herds in the Yukon Territory during 1993 and from four caribou herds in the Northwest Territories during 1991/92.

Location	Finlayson	Tay	Bonnet Plume	Bathurst	Southhampton Island	Cape Dorset	Kimmirut (Lake Harbour)
Pool	Age 2-6	Age 5.3	Age 6.6	Age 4.1	Age 3.6	Age 3.7	Age 4.3
% Lipid	84.2	84.2	83.8	43.0	92.3	19.2	80.4
PCDD/Fs				**Concentration (pg·g^{-1} lipid wt.)**			
Total TCDD	<0.45	<0.05	<0.11	<0.19	<0.15	0.73	0.14
Total PnCDD	<0.32	0.12	<0.13	<0.40	0.30	1.67	0.31
Total HxCDD	<0.74	0.08	<0.26	<0.64	0.10	1.77	0.57
Total HpCDD	<0.98	0.25	<0.20	0.33	<0.32	0.73	0.39
OCDD	0.38	1.09	0.66	2.14	1.65	4.69	1.13
Total TCDF	<0.30	0.20	<0.48	<0.90	0.16	0.99	0.35
Total PnCDF	<0.58	0.52	0.25	0.74	0.63	1.77	0.77
Total HxCDF	<1.63	0.36	<0.70	<1.12	0.23	1.98	0.61
Total HpCDF	<0.80	0.23	<0.13	<0.23	<0.48	<0.93	0.22
OCDF	<0.20	0.25	<0.06	<0.26	<0.13	<0.21	0.25
nPCBs							
CB 37	6.06	3.98	3.93	7.70	2.60	15.1	2.64
CB 77	3.41	3.04	2.92	5.14	6.45	10.3	5.52
CB 81	0.50	<0.59	0.36	0.58	0.91	2.45	1.74
CB 126	6.40	5.74	9.64	7.16	26.7	74.7	30.7
CB 169	0.86	0.58	0.75	0.88	2.28	11.8	3.42
TEQ (pg·g^{-1} wet wt.)							
Total TEQs	0.62	0.76	0.93	0.33	0.85	1.23	3.29
PCDD/Fs	<0.01	0.18	<0.01	<0.01	0.29	0.46	2.50
nPCBs	0.55	0.49	0.82	0.21	0.49	0.47	0.52

analysis. Lichen samples were collected at three different locations on each caribou range, with four subsites sampled at each location.

Biomagnification from caribou to wolf

The relative tissue concentrations of HCB, ΣPCBs and chlordane, the predominant OC contaminants detected in lichen, caribou and wolf, are given in Figure 3.3.14. Residue levels in caribou and wolf are reported on a lipid weight basis, while lichen residue levels are on a dry weight basis. They clearly show the biomagnification of ΣCHL and ΣPCB from caribou to wolf while ΣHCH and HCB did not show a consistent trend between sites. In the case of chlordane, this biomagnification was due to accumulation of the persistent metabolite, oxychlordane, in wolf relative to caribou. Higher chlorinated PCB congeners were present at much higher proportions, relative to the highly persistent congener PCB 153 (2,4,5, 2',4',5'-hexachlorobiphenyl), in wolf in comparison with caribou or lichen. Similar ΣPCB and ΣCHL levels were found in wolf adipose tissue at all three locations. Concentrations of all OCs in lichen were very low; ΣHCH, the most prominent group was present at levels of 1–2 $ng{\cdot}g^{-1}$.

3.3.2.3 Spatial Trends in Waterfowl and Game Birds

Waterfowl, terrestrial game birds and their eggs are harvested to varying degrees for consumption by Aboriginal people (Coad 1994). A survey of contaminants in harvested avian species in the Canadian Arctic has been part of a larger national survey since 1988. Arctic birds inhabiting terrestrial and freshwater environments can be grouped according to trophic level as follows: Browsers — ground dwellers such as grouse and ptarmigan that feed mainly on terrestrial

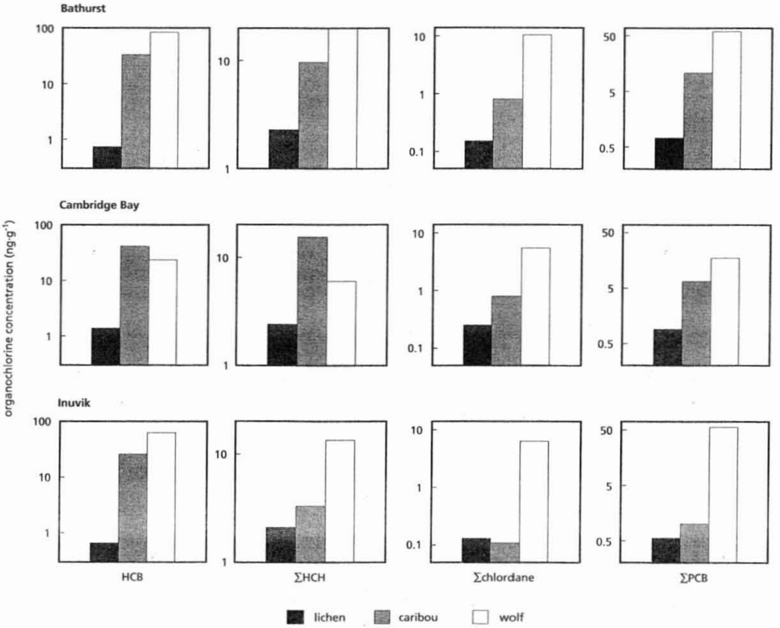


FIGURE 3.3.14

Biomagnification of organochlorines in the lichen→caribou→wolf food web. Residue levels in caribou and wolf are reported on a lipid wt. basis while lichen residue levels are on a dry wt. basis.

vegetation; Grazers — geese that graze mainly on aquatic and terrestrial vegetation; Omnivores — surface-feeding ducks with a varied diet consisting mainly of aquatic vegetation; Molluscivores — diving ducks feeding mainly on invertebrates; Piscivores — diving ducks feeding mainly on fish.

Study Design

The objective was to measure contaminant levels in samples of birds representative of the normal native harvest and submit the results to Health Canada for evaluation of the potential health risks to the human consumer. Beginning in 1988, a different Arctic region was sampled each year: Nunavik Region of northern Québec (1991-92), northern Labrador (1992-93), the Northwest Territories (1993-94), and the Yukon Territory (1994-95). Through the cooperation of the northern communities and First Nations organizations, birds and eggs representative of the native harvest were collected and sent to the National Wildlife Research Centre (NWRC), Hull, Québec, for processing and residue analyses. Egg contents and breast muscle of birds were analysed on a pooled basis (analysis of a composite sample of individuals of the same species collected from the same area within a specified time interval) for OCs, PCB congeners, total mercury, selenium, lead, cadmium and arsenic. Further details on sample size and analysis are given in Braune (1993, 1994).

Northern collection sites were grouped into western Arctic (north of 60°N west of 95°W) or eastern Arctic (north of 55°N east of 95°W, so as to include Hudson Bay, northern Québec and northern Labrador) (Figure 3.3.15). Baker Lake was kept with the eastern sites because of its proximity to Hudson Bay, and Talurjuaq (formerly known as Spence Bay) was kept with the western sites because of the influence of western Arctic waters.

Trophic and spatial trends in organochlorines and metals

Levels of organic contaminants found in breast muscle of harvested birds were generally quite low (Table 3.3.8). The highest OC levels were found in birds feeding at the upper trophic levels, the piscivores and molluscivores. The browsers and grazers contained the lowest levels of organic contaminants. The highest residue levels were found for ΣPCB and ΣDDT in piscivores and molluscivores collected from eastern Arctic communities. Birds collected from eastern locations generally contained higher levels of organic contaminants than comparable collections made from western sites. Many of the bird species are migratory, breeding in the north and overwintering in more temperate, industrialized latitudes. The birds pick up contaminants through the food on their southern

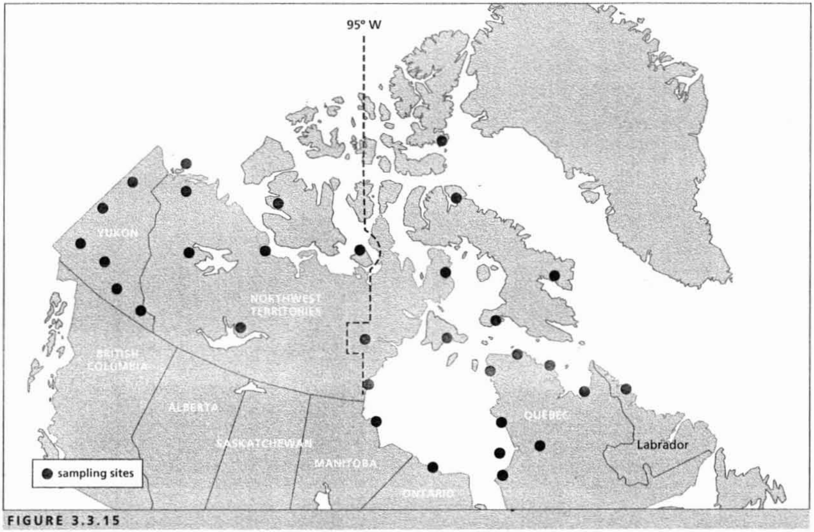


FIGURE 3.3.15

Locations of sampling sites for waterfowl and game birds analysed for organochlorines and heavy metals. Dotted line at 95°W delineates eastern and western collection areas.

TABLE 3.3.8

Range of organochlorine residue levels in breast muscle of waterfowl and game birds harvested in northern Canada in 1988-94 ($ng \cdot g^{-1}$ wet wt.).[1]

Birds[2]	Region[3]	N[4]	ΣPCBs[5]	ΣDDT[6]	ΣCHL[7]	ΣCBz[8]	ΣHCHs[9]	ΣMirex[10]	Dieldrin
Browsers	East	(5, 9, 39)	<1.0–3.1	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
	West	(12, 22, 91)	<1.0–9.6	<1.0–1.5	<1.0–2.5	<1.0–1.9	<1.0	<1.0	<1.0–5.0
Grazers	East	(16, 39, 234)	<1.0–44.9	<1.0–30.0	<1.0–4.5	<1.0–1.6	<1.0	<1.0	<1.0–3.2
	West	(7, 11, 59)	<1.0	<1.0–3.1	<1.0–4.7	<1.0–1.9	<1.0	<1.0	<1.0–2.0
Omnivores	East	(2, 4, 30)	<1.0–87.0	1.0–31.5	<1.0–2.3	<1.0–2.0	<1.0	<1.0–1.6	<1.0–1.3
	West	(8, 31, 149)	<1.0–115	<1.0–650	<1.0–12.8	<1.0–62.4	<1.0–1.0	<1.0–1.6	<1.0 -15.9
Molluscivores	East	(14, 30, 175)	<1.0–1080	<1.0–403	<1.0–102	<1.0–35.9	<1.0–27.8	<1.0–222	<1.0 -120
	West	(12, 42, 178)	<1.0–129	<1.0–127	<1.0–13.2	<1.0–15.4	<1.0–26.5	<1.0–1.7	<1.0–3.8
Piscivores	East	(7, 16, 55)	<1.0–1700	<1.0–951	<1.0–105	<1.0–120	<1.0–2.4	<1.0–268	<1.0 -54.7
	West	(2, 5, 5)	1.1–910	32.0–530	2.9–75.7	1.6–18.3	<1.0–9.9	<1.0–9.2	1.0 -17.4

[1] Detection limit = <1.0 µg·kg^{-1}

[2] Birds are grouped by feeding habit: Browsers (grouse, ptarmigan), grazers (geese, swan), omnivores (blackduck, mallard, teal, pintail, wigeon, shoveller), molluscivores (eider, scoter, scaup, oldsquaw, ring-necked duck, goldeneye, bufflehead, canvasback), piscivores (mergansers, loons)

[3] Birds are grouped into eastern and western collections by collection site (see Figure 3.3.15)

[4] Numbers in brackets refer to: (total # of sites, total # of collections, total # of birds)

[5] ΣPCBs = Sum of 42 congeners: 28, 31, 42, 44, 49, 52, 60, 64, 66/95, 87, 97, 99, 101, 105, 110, 118, 128, 129, 138, 141, 146, 153, 158, 170/190, 171, 172, 174, 180, 182/187, 183, 194, 195, 201, 203, 206

[6] ΣDDTs = Sum of p,p'-DDE, p,p'-DDD and p,p'-DDT

[7] ΣCHL = Sum of oxy-, trans- and cis-chlordane, trans- and cis-nonachlor and heptachlor epoxide

[8] ΣCBz = Sum of 1,2,3,5 and 1,2,3,4 -tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene

[9] ΣHCHs = Sum of α-, β- and γ-hexachlorocyclohexanes

[10] Σmirex = sum of photo-mirex and mirex

TABLE 3.3.9

Range of metal residue levels in breast muscle of waterfowl and gamebirds harvested in northern Canada from 1988–94 ($ng \cdot g^{-1}$ wet wt.)[1].

Trophic[2] Level	Region[3]	N[4]	Mercury	Selenium	Cadmium	Arsenic
Browsers	East	(5, 9, 39)	<30–58	60–241	18–437	2–20
	West	(12, 22, 91)	<60	<30–527	<50–583	<70
Grazers	East	(16, 39, 234)	<130	38–826	<20–40	3–900
	West	(7, 11, 59)	<30–156	105–781	10–30	<100
Omnivores	East	(2,4,30)	202–230	183–520	<250	<40
	West	(8, 31, 149)	24–665	<100–2080	<10–80	<30–110
Molluscivores	East	(14, 30, 175)	35–455	314–1750	20–707	<30–600
	West	(12, 42, 178)	<20–369	<100–5790	<10–510	<30–426
Piscivores	East	(7, 16, 55)	74–1230	373–1240	<30–110	7–723
	West	(5,5,5)	258–1930	254–1510	<65–438	<30–232

[1] Detection Limits – shown as (<value). Detection limits with trace metals analysis are very variable, being highly dependent on the individual sample matrix.

[2] Birds are grouped by feeding habit: (see Table 3.3.8)

[3] Birds are grouped into eastern and western collections by collection site (see Figure 3.3.15)

[4] Numbers in brackets refer to: (total # of sites, total # of collections, total # of birds)

wintering grounds or sites along the migration pathway. Historically, some of the eastern overwintering areas such as the Great Lakes–St. Lawrence River have been much more contaminated than some of the western areas. This is clearly illustrated by mirex, a contaminant characteristic of the lower Great Lakes (Comba *et al.* 1993). Mirex is virtually undetectable in western arctic birds but is quite measurable in eastern arctic birds (Table 3.3.8). Total toxaphene levels in 12 pooled waterfowl samples from the Yukon ranged from non-detectable to 9.4 $ng \cdot g^{-1}$, with highest levels in Oldsquaw.

Metal levels found in breast muscle were also generally quite low (Table 3.3.9). Lead concentrations were not included because the presence of lead shot in muscle precluded accurate determination of natural levels in breast muscle. The distribution of metals across trophic levels is not as clear as for organic contaminants, although levels were still generally higher in the molluscivores and piscivores. In addition to atmospheric transport, metals also occur naturally in the mineral resources of the Canadian North. Some of the activities associated with mineral extraction mobilize the metals making them more bioavailable (Thomas *et al.* 1992). Metals are often transported via riverine sediments. Therefore, those birds that are predominantly benthic feeders may be subject to a greater exposure to metal contamination since they eat prey associated with bottom sediments where metals may settle out. As well, plants absorb metals, particularly cadmium, from soil and water (Singh 1994), which would explain the measurable levels in the browsers and grazers.

3.3.2.4 Spatial Trends of Organochlorines in Mink in the Western NWT

Mink (*Mustela vison*) are top trophic level carnivores found throughout the forested regions of the NWT, and are harvested by local trappers for their valuable pelts. Small mammals and fish form the greatest components of mink diet in most areas (Gilbert and Nancekivell 1982, Eagle and Whitman 1987, K. Poole unpublished data), thus mink are exposed to contaminants derived from both terrestrial and aquatic food webs. Mink readily bioaccumulate environmental pollutants such as PCBs, DDT-related compounds, and methylmercury residues (Platonow and Karstad 1973, Aulerich and Ringer 1977, Hornshaw *et al.* 1983, Wren *et al.* 1987a). Mink are extremely sensitive to PCB contaminants, and are known to experience reproductive impairment or failure as a result of eating food contaminated with relatively low levels of PCBs (reviewed in Ringer 1981, Eisler 1986, Kihlström *et al.* 1992). This unique susceptibility can result in population effects at low levels of environmental contaminants (Wren 1991). As such, mink may be a sensitive indicator to assess trends in environmental contaminants and ecosystem health. A variety of contaminants have been identified in freshwater fish from in and near the Mackenzie River system (Muir *et al.* 1990a, Kuhnlein 1991), providing a source of contaminants for mink.

Study design

A total of 1025 mink was collected from trappers during the winters 1991-92 and 1994-95. Tissue samples from up to 41 mink from each of seven sites (Figure 3.3.16) were analysed for a suite of 63 organochlorine residues in liver samples, including 43 PCB congeners and 20 pesticides, and residues of 10 heavy metals in liver and kidney samples. Pooled samples were examined for PCDD/Fs and toxaphene. Carcasses were collected for three winters from the Mackenzie Delta near Inuvik to examine short-term temporal trends. Stomach contents were examined for diet, teeth were used for aging, and a variety of biological and morphometric measurements were taken. Further details on sample collection and analysis can be found in Poole *et al.* (1995).

Spatial trends

Most of the OC pesticides and PCB congeners detected were found at very low levels in all samples. ΣPCB residues (sum of 43 congeners) ranged from a mean of 4.9 ng·g^{-1} wet wt. in the livers of Inuvik mink (1994 sample) to 73 ng·g^{-1} in the Fort Providence sample (Figure 3.3.16). More highly chlorinated PCB congeners dominated the pattern. ΣDDT ranged from 0.8 ng·g^{-1} in the 1992 Inuvik sample to 9.5 ng·g^{-1} in Fort Providence mink (Figure 3.3.16). ΣPCB and ΣDDT in the Inuvik samples varied among years, but were consistently low relative to other communities. DDT was below detection limits (<0.01ng·g^{-1}) in all but two mink. Of the more toxic nPCB congeners, CB-77 was not detected, CB-169 was only detected in one mink, and residues of CB-126 were found in two mink samples. PCDD/Fs were not detected (<1 pg·g^{-1}) in any of five pooled liver samples examined. Total toxaphene in two pooled liver samples was 1.4 and 7.9 ng·g^{-1} (wet wt.) for animals from Fort Good Hope and Fort Rae, respectively. Heavy metal residues were comparatively low, with the exception of mercury, which had levels (community means) of 0.9–3.3 µg·g^{-1} (wet wt.) in liver. Female mink had higher burdens of ΣHCH and HCB, but levels of other OC groups did not differ between sexes. There were no differences in OC burden between younger (<2 years of age) and older mink.

There was a distinct trend of decreasing contaminant burdens with increasing latitude in many of the groups of OCs, most noticeably for ΣDDT and ΣPCB (Figure. 3.3.16). No spatial pattern was evident in ΣHCH and ΣCHL. No spatial trends in heavy metal burdens were evident. Unlike the OCs, residues of many of the heavy metals are likely due to natural sources and local geology (Wren *et al.* 1986, Barrie *et al.* 1992). The reason why ΣPCB levels in the Fort Providence samples were two to three times higher than mink from any other community is unknown; Fort Providence mink did not have significantly higher residues of other groups of OCs, and mink diet did not differ from other collection sites (K. Poole, NWT Renewable Resources, unpublished data).

Of the three likely sources for contaminants (i.e. long-range transport, local, or natural sources) (Barrie *et al.* 1992), long-range atmospheric transport appears to be the primary source of many of the OCs detected, given the generally uniform distribution of contaminants within trapping areas and the latitudinal trend in residue levels. Local point sources for OC contaminants are not suspected, and the pattern of contaminant burdens in mink collected on and off the Mackenzie River proper or its deltas is not consistent with long-range aquatic transport from southern sources of pollution.

Overall, contaminant levels in western NWT mink were low in comparison with wild mink from other areas of North America, where total PCB (based on Aroclor equivalent or sum of individual congeners) and ΣDDT were generally one to two orders of magnitude higher (summarized in Poole *et al.* 1995). Reproductive impairment and reduced growth and survival of mink kits has been observed at 1200–2000 ng·g^{-1} (wet wt.) Aroclor (Platonow and Karstad 1973, Wren *et al.* 1987b) and at 50,000 ng·g^{-1} (lipid wt.) ΣPCB in liver tissues (Jensen *et al.* 1977). ΣPCB residues observed in NWT mink were considerably lower than these levels. Direct assessment of population reproductive performance from mink harvested

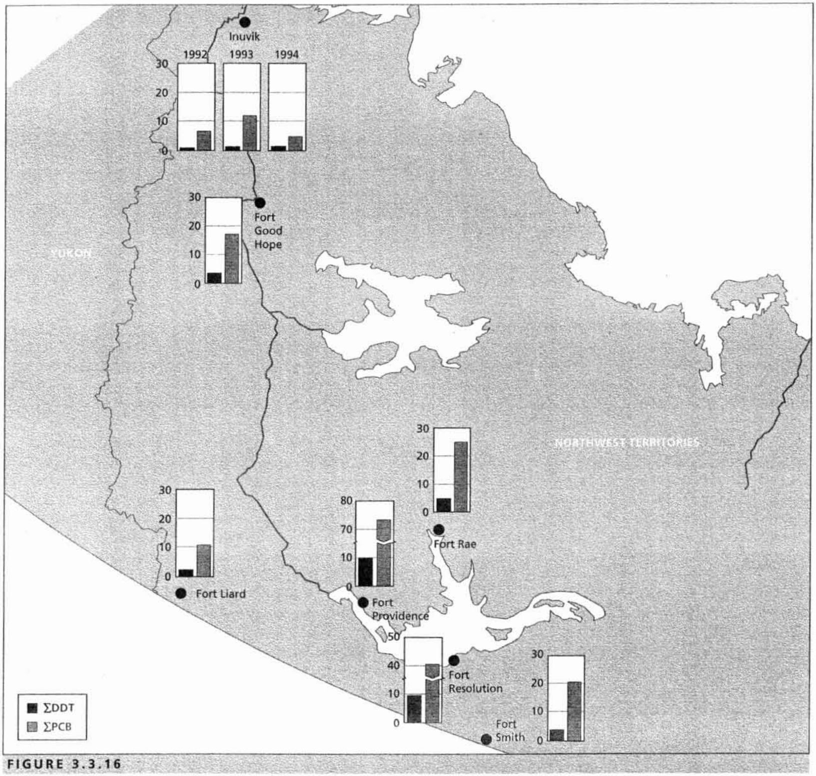


FIGURE 3.3.16

ΣPCB and ΣDDT in mink liver (ng·g^{-1} wet wt.) from various locations in the western NWT. Note that different scales of concentration are used for results from Ft. Providence and Ft. Resolution compared with other locations.

during winter is not possible (Wren 1991), however, population indices derived from age and sex ratios of the harvest (Strickland and Douglas 1987) suggest light harvest pressure and healthy reproductive performance in NWT mink. These indices, coupled with the comparatively low levels of contaminants, suggest little or no effects on reproduction or population health of NWT mink as a result of these contaminants.

3.3.2.5 PCBs and Metals in Plants and Soils at Remote and "Background" Sites Near the DEW Line

As part of environmental assessment and impact studies at Distant Early Warning (DEW) Line sites and other radar facilities across the Canadian Arctic and Labrador, samples comprising a wide variety of species (shrubs, sedges, grasses, wildflowers) were collected at locations near to and remote from the sites. Large amounts of potentially hazardous materials such as PCB-containing devices were stored at many of the radar sites, and no cleanup or maintenance was conducted following their closure. Plants occur at the base of the arctic terrestrial food web, hence they are valuable indicators of contaminants entering the ecosystem. Previous studies have demonstrated that lower plants such as mosses and lichen are good indicators of atmospheric contaminants; for example, in the atmospheric distribution of inorganic and organic compounds in remote and northern areas (Grodinska and Godzik 1991, Ross

1990, Schaug *et al.* 1990, Thomas 1986, Thomas and Schunke 1984). Lichens, in particular, have been studied in recent years with respect to food chain links to caribou and humans in the high Arctic (Nash and Gries 1995a, 1995b, Elkin and Bethke 1995). There has been little information on arctic contaminant concentrations in higher (vascular) terrestrial plants at lower northern latitudes. Vascular plants can be important components in the summer diets of terrestrial herbivores and for other animals that periodically consume plant material (Reimer *et al.* 1993a).

Study design

Over 3500 soil and 1600 vascular plant samples were collected in the immediate vicinity of recently active (main) radar sites as well as at "intermediate" sites, i.e. those abandoned during the 1960s (Reimer *et al.* 1991, 1993a, 1993b, 1993c, 1994, Dodd and Reimer 1992, Dushenko and Reimer 1994, Grundy *et al.* 1994). Background and "remote" locations were also sampled. Background areas refer to locations which are in the vicinity of radar sites but not directly affected by their activities (i.e. within several kms of the site), while remote sites were 20 km or more distant from the radar facilities.

PCBs and lead in plants and soils

The major contaminants found in the plant and soil samples were PCBs and inorganic elements such as lead, copper and zinc. Cadmium and mercury, which are elevated in some terrestrial mammals in the Arctic (Elkin and Bethke 1995), are not found at elevated concentrations in plants and soils at the radar sites. It is therefore unlikely that these stations serve as local sources of these particular contaminants. PCBs and lead, were the primary contaminants along the DEW Line. The average PCB (Aroclor) concentrations for 525 associated plants and soils collected at various background locations and different radar sites ranged from about 1 $ng \cdot g^{-1}$ to 10 000 $ng \cdot g^{-1}$ (Figure 3.3.17a). Average PCB concentrations were higher in plants and soils at radar sites compared with local background and more remote Arctic locations. This is the result of past activities of these sites; such as spills, solid waste disposal and down-the-drain discharge to sewage outfalls. Lower concentrations of PCBs in plants relative to underlying soils indicate that not all of the PCBs occurring in the soils are directly bioavailable to plants and/or a portion of the PCBs bioaccumulated is lost during leaf/plant senescence. The most profound influence is found in sites from the east coast (Labrador coast and Resolution Island) where concentrations in soils and associated plants are the highest, reaching up to 320 000 $ng \cdot g^{-1}$ and 49 000 $ng \cdot g^{-1}$ respectively. The persistence of PCBs at radar stations is also evident based on concentrations in soils (maximum of 1400 $ng \cdot g^{-1}$) and plants (maximum of 3400 $ng \cdot g^{-1}$) at the Intermediate (I) sites. These values are lower than at the more recently active sites (least significant difference, at $P \leq 0.02$), but are still significantly elevated relative to background locations despite their closure over 30 years ago (Fletcher, 1990). Significantly higher concentrations of PCBs are also found in soils and plants from site background areas than in more remote Arctic locations (Figure 3.3.17A) indicating that the sites can serve as local aerial sources of PCBs to the surrounding ecosystem. This has been confirmed by the analysis of congener signatures in both soils (Bright *et al.* 1995a) and plants. The ratio of PCB concentrations in soils to plants also decreases in moving away from sites toward background and remote locations (Figure 3.3.17a). This suggests a decline in the importance of PCBs redistributed to the surrounding tundra from local sources (e.g., spills) relative to long-range atmospheric transport in remote locations.

Similar patterns are also observed for average lead concentrations in 617 associated plants and soils from different radar sites and background areas (Figure 3.3.17b). The highest concentrations occur in plants (maximum 1120 $\mu g \cdot g^{-1}$) and soils (maximum 2780 $\mu g \cdot g^{-1}$) at the Auxiliary (Aux) and larger Main sites, which were closed in the early 1990s (most recent closures). Lead contamination does not appear to be as extensive in the ecosystem as that for PCBs, based on the lower lead concentrations in plants at the I-sites (maximum 70 $\mu g \cdot g^{-1}$). These are similar to levels occurring at background locations (maximum 72 $\mu g \cdot g^{-1}$, Figure 3.3.17b).

Concentrations of PCBs in plants are found to be significantly correlated with soil levels ($r^2 = 0.98$; $p < 0.001$) from background and radar stations (Figure 3.3.17c). The predictability of plant concentrations based on soil PCB levels generally falls within an order of magnitude; much of the variation observed within the central spread is due to differences between the plant species analysed (e.g. longevity, habit), reversed relationships observed between soil and plant concentrations at background and remote locations, and site-specific factors (e.g. habitat).

The above results indicate that radar stations are a significant source of lead and PCBs to the surrounding terrestrial ecosystem. A study examining PCB soil-plant-lemming (*Dicrostonyx groenlandicus*) relationships in the sewage outfall and background areas at the Cambridge Bay radar site was conducted in 1992 to examine bioavailability of the PCBs. Concentrations of PCBs in samples from the sewage outfall were all significantly larger than background values with averages, in some cases, differing by an order of magnitude or more (Figure 3.3.17d). Although the average PCB concentration was much higher in soils (average 1600 $ng \cdot g^{-1}$) than in plants (average 9.3 $ng \cdot g^{-1}$) in the outfall, plant-herbivore food chain biomagnification is indicated by a 6.5-fold increase in

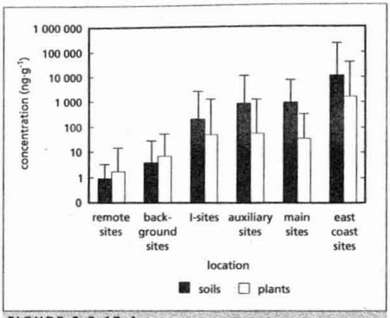


FIGURE 3.3.17 A

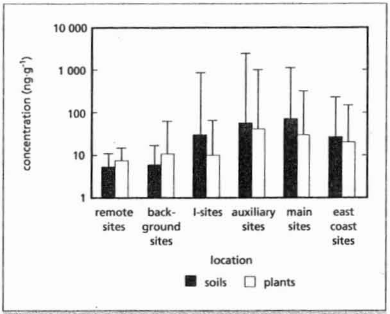


FIGURE 3.3.17 B

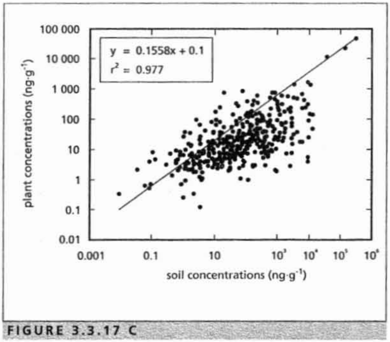


FIGURE 3.3.17 C

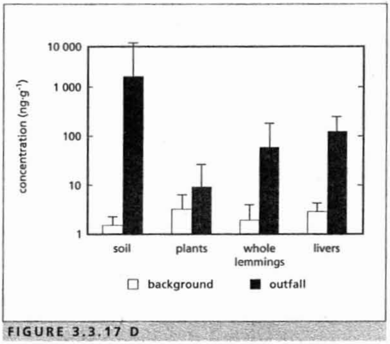


FIGURE 3.3.17 D

Concentrations of Aroclor PCBs and lead in soils and plants associated with radar sites. A. Mean and maximum total PCB concentrations in soils and associated plants from remote (>20 km), background (<20 km), intermediate (I), auxillary, main and east coast DEW line radar sites. Intermediate sites were abandoned in the 1960s. Auxiliary and main sites were used until the 1990s. B. Mean and maximum lead concentrations in soils and associated plants from the same locations as in A. C. PCB soil-plant relationships at background and radar sites. D. Mean and maximum concentrations of PCBs in soil, plants and lemming tissue from background and outfall areas at Cambridge Bay Radar Site.

PCB concentration between lemmings (average whole body concentrations of 61 ng·g^{-1}) and plants. The average PCB level in pooled lemming livers (123 ng·g^{-1}) was double the whole body level. These levels are also considerably higher than the range of average values reported for wild mink, a predator of lemmings, in the western NWT (5.32 to 27.7 ng·g^{-1}, Poole *et al.* 1995). At present, it is not known what the implications of these concentrations in lemmings would be for predators such as the arctic fox, or rough-legged hawks that nest in the area.

3.3.2.6 Contaminant Levels in Traditional Foods in the Yukon

Study design

In 1993/94, a survey was conducted on contaminants in plants used as traditional foods and medicines by the Kaska First Nation in the southeastern Yukon. This survey was expanded the following year to cover the entire Yukon Territory, and to include plants, birds and mammals. In all cases, sampling lists were devised in consultation with local First Nations to ensure that results and conclusions would be locally relevant. Further details on sampling and chemical analysis are given in Gamberg and Mychasiw (1994b). Table 3.3.10 lists the species of plants and animals that were sampled and analysed.

TABLE 3.3.10

Traditional plant and animal samples collected for contaminant analysis from the Yukon.

Plant part	Common Name	Species	N
Vegetation			
Berry	Bearberry	*Arctostaphylos uva-ursi*	2
	Black current	*Ribes hudsonianium*	1
	Blueberry	*Vaccinium uliginosum*	5
	Cloudberry	*Rubus chamaemorus*	1
	Crowberry	*Empetrum nigrum*	4
	Gooseberry	*Ribes oxyacanthoides*	2
	Highbush Cranberry	*Viburnum edule*	4
	Lowbush Cranberry	*Vaccinium vitis-ideae*	5
	Nagoon berry	*Rubus arcticus*	2
	Raspberry	*Rubus ideaus*	5
	Red current	*Ribes triste*	2
	Soapberry	*Sheperdia canadensis*	3
	Strawberry	*Fragaria virginiana*	2
	Wild rose	*Rosa acicularis*	4
thallus	Birch fungus	unknown	1
	Caribou horn	*Cornicularia aculeatum*	3
	Puffball mushroom	*Lycoperdon calvatia*	2
bark	Alpine fir	*Abies lasiocarpa*	1
	Balsam poplar	*Populus balsamifera (bark)*	1
	Tamarack	*Larix laricina*	1
branches	Alpine fir	*Abies lasiocarpa*	1
cones	Spruce	*Picea mariana*	1
foliage, stems	Tamarack	*Larix laricina*	1
	Willow	*Salix* sp.	5
roots	Bear root	*Hedysarum alpinum*	2
foliage	Fireweed	*Epilobium angustifolium*	3
	Wild onion	*Allium schoinopraum*	1
	Wild rhubarb	*Polygonum alaskanum*	2
	Yarrow	*Achillea millefolium*	1
flowers, stems	Yarrow	*Achillea millefolium*	1
flowers	Labrador tea	*Ledum groenlandicum*	1
foliage, stems	Yarrow	*Achillea millefolium*	3
	Bear root	*Hedysarum alpinum*	1
	Sage	*Artemisia frigida*	1
foliage, flowers	Dandelion	*Taraxacum officinale*	3
foliage, stems	Alder	*Alnus incana*	5
	Caribou leaves	*Artemisia tilesii*	7
	Juniper	*Juniperus communis*	3
	Labrador tea	*Ledum groenlandicum*	4
	Shrubby cinquefoil	*Potentilla fruticosa*	5
	Soapberry	*Sheperdia canadensis*	5
foliage, twigs	Red willow	*Cornus stolonifera*	2
Mammals			
	Moose	*Alces alces*	62
	Caribou	*Rangifer tarandus*	24
	Mountain goat	*Oreamnos americanus*	4
	Dall sheep	*Ovis dalli*	5
	Muskrat	*Ondatra zibethicus*	8
	Snowshoe hare	*Lepus americanus*	18
	Beaver	*Castor canadensis*	8
	Porcupine	*Erethizon dorsatum*	4
	Arctic Ground Squirrel	*Citellus parryi*	5
	Lynx	*Lynx canadensis*	1
Birds			
	Spruce grouse	*Dendragapus canadensis*	30
	Blue grouse	*Dendragapus obscurus*	1
	Ruffed grouse	*Bonasa umbellus*	3
	White-tailed ptarmigan	*Lagopus leucurus*	2
	Rock ptarmigan	*Lagopus mutus*	4
	Willow ptarmigan	*Lagopus lagopus*	8
	Ptarmigan	*Lagopus* sp.	6

Vegetation

Most plants analysed contained element levels considered normal for vegetation, although some concentrations ranged into the high or low end of the normal range. Barium was very high in most plants relative to commercial livestock diets. The toxic barium ion is never found free in nature, but occurs mainly as sulphate that is virtually insoluble and nontoxic, or carbonate, which has a low toxicity (Puls 1994). The vegetation in this study was analysed for total barium, so the chemical form is not known. Cadmium and lead were uniformly low in most plants analysed. One exception analysed was a puffball mushroom from Ross River which contained 13.3 µg·g^{-1} cadmium and 8.72 µg·g^{-1} lead (dry wt.). A sample of the same species from Watson Lake contained <0.1 µg·g^{-1} cadmium and <0.50 µg·g^{-1} lead (dry wt.). The mushroom sample from Ross River also contained high levels of aluminium, chromium, cobalt, copper, iron, nickel, selenium, vanadium and zinc, whereas the sample from Watson Lake contained high levels of copper and zinc only. The element levels in these mushrooms appear to reflect the mineral composition of the substrate, and would be expected to be highly variable among locations.

Metal uptake in plants is species-specific and metal-specific, and can be affected by soil pH, organic content, calcium content, cation exchange capacity, oxides of iron and manganese and the presence of ligands in the soil (Crowder 1991). Terrestrial plants generally have highest concentrations in roots, but this is not consistent. Bear root *(Hedysapum alpinum)* plants in this study had higher levels of some elements, such as aluminum, copper and iron, in their roots than in foliage and stems, but both plant parts had similar levels of other elements such as calcium, lead and zinc. Some metals are taken up mostly from the soil, whereas some, such as lead, accumulate primarily from the atmosphere. In these cases, high concentrations may accumulate in the bark of trees. The only measurable level of lead in three samples of tree bark in this study was still very low (0.64 µg·g^{-1} dry wt.), indicating low levels of airborne lead in this area. Cadmium ranged from 0.2 µg·g^{-1} to 1.0 µg·g^{-1} in bark samples, reflecting slightly higher airborne levels of this contaminant.

No unusually high concentrations of any elements were found in the plants analysed in this study. However, it should be noted that element concentrations in vegetation tend to be extremely variable depending on plant species, plant organ and local soil conditions.

Mammals and birds

Levels of OC contaminants (PCBs, chlorinated pesticides) in the terrestrial birds and mammals tested were generally low i.e., <10 ng·g^{-1} wet wt. Only one porcupine (*Erethizon dorsatum*) had levels of chlorinated dioxins and -furans above detection limits >1 pg·g^{-1}. The same was true of metal levels with the exception of mercury and cadmium. Although mercury was elevated in some caribou kidneys (up to 3.04 µg·g^{-1} dry wt.), previous work with the Finlayson caribou indicated that all mercury present was in the inorganic form (Gamberg 1993).

Cadmium levels in muscle tissue of all animals were consistently low (<0.2 µg·g^{-1} dry wt.) with the

TABLE 3.3.11

Cadmium concentration (µg·g^{-1} dry wt.) in birds and mammals from the Yukon.

Species[1]		Kidney				Liver				Muscle		
	N	Mean	±	S.D.	N	Mean	±	S.D.	N	Mean	±	S.D.
Muskrat	8	<0.20		—	8	<0.10		—	8	<0.20		—
Ground Squirrel	5	15.3	±	6.3	5	5.1	±	3.0	5	<0.20		—
Lynx	0	—		—	1	6.2		—	1	<0.20		—
Snowshoe Hare	18	20.7	±	16.2	17	2.4	±	2.1	6	0.02	±	0.01
Grouse	34	80.0	±	232	28	8.1	±	12.4	7	<0.01		—
Beaver	8	130	±	99.1	7	17.0	±	10.2	8	<0.20		—
Ptarmigan	20	143	±	68.4	19	38.8	±	35.5	0	—		—
Porcupine	4	169	±	101	4	38.4	±	24.3	4	0.40	±	0.20

[1] *Refer to Table 3.3.10 for specific names. Grouse and ptarmigan are pooled samples of species listed in Table 3.3.10.*

exception of porcupine, but levels in kidneys and livers ranged from very low in muskrats (*Ondatra zebitheca*), to very high in moose and some grouse (Table 3.3.11). Beaver (*Castor canadensis*) from the Yukon had higher levels of cadmium than beaver from the Sudbury mining area of Ontario, which were considered to have elevated levels (Hills and Parker 1993). Cadmium concentrations in Yukon grouse and ptarmigan (*Lagopus* sp.) (Table 3.3.11) were similar to levels found in ptarmigan in Norway (Fimreite 1993), but some individuals from the Yukon had very high concentrations (up to 1200 µg·g^{-1} dry wt. in kidney). Little comparative data exist for porcupines, snowshoe hares and ground squirrels, but cadmium levels are quite high in porcupine, and in individual snowshoe hares. Muskrats, on the other hand, had uniformly low levels of cadmium.

3.3.2.7 Overall Assessment of Contaminants in Terrestrial Mammals and Birds

The information available on contaminants in arctic terrestrial mammals and waterfowl has increased significantly over the past five years. Previous reviews found relatively little information available on contaminant levels in the terrestrial environment (Thomas *et al.* 1992, Wong 1985). Geographic coverage of major species such as caribou and mink (within NWT), as well as waterfowl and game birds, is now quite complete. Contaminant data are available for a greater number of terrestrial mammals in the Yukon compared with the NWT. PCBs and cadmium appear to be the most prominent contaminants in the species analysed.

Relatively high levels (10–60 µg·g^{-1}) of cadmium were observed in kidney and liver of caribou from the Yukon, the NWT and northern Québec (Crête *et al.* 1989). Cadmium is higher in the western Arctic/Yukon herds but little geographic trend is evident for mercury. Higher cadmium levels (20–190 µg·g^{-1}) in the Finlayson and Tay herds in the southeastern Yukon may be the result of natural mineralization accumulating in some plants (such as *Salix* sp.), and in this way being bioaccumulated by herbivores. There did not appear to be high levels of cadmium particularly associated with industry (mining) or populated areas. Mercury levels in caribou were generally low in liver (means of 0.16–0.92 µg·g^{-1} in the NWT herds) and higher in kidney (means of 0.52–3.5 µg·g^{-1} in all herds). Most of this mercury was found to be in the less toxic inorganic form.

Moose liver and kidneys from Yukon animals had higher cadmium levels than reported in moose from Manitoba (Wotton and McEachern 1988), Québec (Crête *et al.* 1987), Newfoundland (Brazil and Ferguson 1989), Norway (Froslie *et al.* 1986, Scanlon *et al.* 1986) and Sweden (Frank 1986). However, moose from some areas of Ontario (Glooschenko *et al.* 1988) and New Brunswick (Redmond *et al.* 1988) had similar levels of cadmium to those found in Yukon moose.

No clear spatial trends were observed in metals in waterfowl between eastern and western Arctic locations. This contrasts with caribou which clearly had higher concentrations of cadmium in western arctic herds than in those from the east. The waterfowl are migratory and, as noted below for OCs, may reflect exposure in the winter feeding locations.

Radionuclides are important contaminants in caribou and, unlike many other chemicals assessed here, information is available on temporal trends in caribou muscle and lichen over more than 30 years. Current levels of ^{137}Cs in caribou muscle are four to more than 10 times lower than during the 1960s when levels reached 2000 Bq·kg^{-1} in several the NWT herds following atmospheric weapons testing (Thomas *et al.* 1992). Levels of ^{137}Cs also shows a pronounced geographic trend with higher concentrations in northern Québec caribou herds. The levels of ^{137}Cs are also much lower than observed in reindeer in northern Europe after the Chernobyl accident in 1986. Caribou receive relatively high doses of natural radiation. Macdonald *et al.* (1996) concluded that radiation doses for Kimmirut caribou, due mainly to ^{210}Po were high relative to other mammals in the Canadian environment. Highest levels of ^{210}Pb were found in bone. The animals have presumably adapted to the radiation stress from these natural sources but potential effects of the high doses on the animals are not clear.

PCBs were the most prominent organochlorine contaminants and terrestrial mammals. Some clear spatial trends emerge from the data, unlike the situation for metals. Mink in southern NWT (the Great Slave Lake region) had higher PCB concentrations than those from the Mackenzie delta. But PCB concentrations in mink from the NWT were lower than those reported for wild mink in southern Ontario (Poole *et al.* 1995). A significant west to east increase in ΣPCBs, HCB and ΣHCH was found in caribou with highest mean levels in Cape Dorset and Kimmirut herds and lowest in the Inuvik herd. This trend was also observed for PCDD/Fs in caribou. Higher levels of these OCs in the east are probably also the result of the predominate west to east/northeast atmospheric circulation pattern, which delivers these contaminants from industrialized regions of central and eastern North America to the Arctic via long-range atmospheric transport.

Levels of PCDD/Fs were very low (<1 $pg \cdot g^{-1}$ for total PCDD/Fs at most sites) and TCDD TEQs in caribou were attributable mainly to non-ortho and mono-ortho PCB congeners. These results are similar to those from previous studies which found that PCBs were the major contributors to overall TEQ levels in Arctic marine mammals (Daelemans *et al.* 1993, Ford *et al.* 1993).

These results for PCDDs, PCDFs and nPCBs in caribou showed very low levels, which are unlikely to pose a threat to either the caribou sampled in this study or to their human consumers. The levels observed can probably be considered to be background concentrations. TEQs in caribou fat are as low or lower than those reported in fat of domestic animals in Canada (Ryan and Norstrom 1991).

Higher PCBs and other organochlorines, particularly mirex, were also found in waterfowl, especially in molluscivores and piscivores, in eastern, mainly coastal (northern Québec, Nunavik) locations. In the case of birds, however, most overwinter in temperate latitudes and the east-west trends in organochlorines may therefore reflect migratory patterns and winter feeding locations rather than regional contamination differences.

Significant contamination by PCBs and lead of soils and vascular plants is observed in the immediate vicinity and within a 20 km radius of abandoned and recently active DEW line sites in the Canadian Arctic. There is evidence for transfer of PCBs at former DEW line radar sites from plants to lemmings. This raises the possibility that other terrestrial mammals and birds could be contaminated because of feeding, even infrequently, on resident plants or animals at these locations. Arctic char from Char Lake, which is near the airport at Resolute on Cornwallis Island, have much higher ΣPCB than toxaphene, an unusual result which may imply past contamination of the lake. The work on PCBs and lead in plants and soils near DEW line sites demonstrates how these contaminants can move off site over time, presumably via volatilization and redeposition or on soil particles, so that concentrations can be elevated (compared to remote background locations) even several kilometers away. There was no evidence that large mammals such as caribou living in the general area of the DEW line sites had elevated levels of PCBs, however, it should be noted that the studies were not designed specifically to address this question. Further analysis of PCB congener composition in existing caribou and bird data would help to address this concern as would analysis of terrestrial predators such as arctic fox.

- *PCBs and cadmium are the most prominent contaminants in terrestrial mammals and waterfowl species that have been analysed. Geographic coverage of major species such as caribou and mink (within NWT), as well as waterfowl and game birds, is quite complete. Contaminant data are available for a greater number of terrestrial mammals in the Yukon compared with NWT.*

- *Relatively high levels (10-60 $\mu g \cdot g^{-1}$) of cadmium are observed in kidney and liver of caribou from the Yukon, the NWT and northern Québec. Cadmium is higher in the western Arctic/Yukon herds but little geographic trend is evident for mercury. There did not appear to be high levels of cadmium particularly associated with industry (mining) or populated areas.*

- *Radionuclides are important contaminants in caribou. Current levels of ^{137}Cs in caribou muscle are 4- to more than 10-times lower than during the 1960s when levels reached 2000 $Bq \cdot kg^{-1}$ in several NWT herds following atmospheric weapons testing. Levels of ^{137}Cs also shows a pronounced geographic trend with higher concentrations in northern Québec caribou herds. The levels of ^{137}Cs are also much lower than observed in reindeer in northern Europe after the Chernobyl accident in 1986.*

- *Caribou receive relatively high doses of natural radiation relative to other mammals in the Canadian environment due mainly to ^{210}Po. Highest levels of ^{210}Pb were found in bone.*

- *PCB levels in terrestrial mammals show some clear geographic trends. Mink in southern NWT (the Great Slave Lake region) had higher PCB concentrations than those from the Mackenzie delta. A significant west to east increase in PCDD/Fs, ΣPCBs, HCB and ΣHCH was found in caribou with highest mean levels in Cape Dorset and Lake Harbour herds and lowest in the Inuvik herd.*

- *ΣPCB and ΣDDT were higher in piscivorous and molluscivorous birds from the eastern Arctic (Hudson Bay, N. Québec and Baffin Island) than in comparable species in the western Arctic. In the case of birds, the east-west trends in OCs may reflect migratory patterns and winter feeding locations rather than regional contamination differences.*

- *Levels of PCDDs and PCDFs in caribou were very low (<1 pg·g^{-1} for total PCDD/Fs at most sites). Most of the TCDD TEQs in caribou were attributable to non-ortho and mono-ortho PCB congeners. The TCDD TEQ levels observed in caribou fat can probably be considered to be background concentrations because they are as low or lower than those reported in fat of domestic animals in Canada.*
- *Significant contamination by PCBs and lead of soils and vascular plants is observed in the immediate vicinity and within a 20 km radius of abandoned and recently active DEW line sites in the Canadian Arctic.*
- *There is evidence for transfer of PCBs from plants to lemmings at former DEW line radar sites. This raises the possibility that some terrestrial mammals and birds could be contaminated because of feeding, even infrequently, on resident plants or animals at these locations.*
- *There was no evidence that large mammals such as caribou living in the general area of the DEW line sites had elevated levels of PCBs, however, it should be noted that the studies were not designed specifically to address this question.*

3.3.3 Marine Mammals and Sea Birds and their Food Webs

3.3.3.1 Arctic Marine Food Web/ Contaminant Transfer Studies

Although many organochlorine pesticides and PCBs were detected in Arctic marine fauna in the early 1970s, the data base is small and prior to 1990 was restricted almost entirely to tissues from marine mammals (seals and polar bears) and sea birds (Muir *et al.* 1992a). More recent studies supported under the Arctic Environmental Strategy have provided data for fish and lower trophic level organisms that form the basis of the food web leading to humans (Hargrave *et al.* 1992). In the case of metals, a considerable amount of information is available in lower trophic level marine organisms from the work of Macdonald and Sprague (1988) on cadmium and for specific sites such as Strathcona Sound (Bohn and Fallis 1978) but relatively little new (post-1990) information is available.

For both OCs and metals, the distribution and mechanisms for transfer of these compounds between various physical compartments and within the marine food web are poorly understood. Although Arctic marine food web structure is thought to be simply due to the relatively few species that occur in a given region (Section 3.2.4.4), interactions between predators and prey species are sufficiently complex to form networks of potential transfer points (Figure 3.3.18)

Variation of organochlorines in the lower trophic level marine biota

A series of studies conducted since 1986 have extended OC measurements to include organisms from lower trophic levels in the Arctic marine food web. Using facilities on the Canadian Ice Island and at Resolute Bay, OCs have been measured in epontic and under-ice particulate matter, tissues of planktonic and benthic invertebrates, abyssal and coastal marine fish. The OC data (on a lipid weight basis) have been compared with published results from arctic fish and marine mammals to determine levels of biomagnification occurring within the food web. Further details on sampling and analysis are given by Hargrave (1993) and by Hargrave *et al.* (1992).

Toxaphene, PCBs and isomers of DDT and *p,p'*-DDE were the predominant OCs measured in epontic particulate matter, all size classes and taxa of pelagic and benthic zooplankton and amphipods and tissues from arctic cod and char (Table 3.3.12). Concentrations of toxaphene and PCBs were of a similar magnitude and positively correlated as reported in Hargrave *et al.* (1993), but mean PCB: toxaphene ratios were not the same for all taxa. The ratio was closest to unity for zooplankton and deviated the most (values <1) in beluga blubber and fish tissues. The small body size, seasonal cycles of fat deposition and metabolism and short generation time in zooplankton may result in OC body burdens that rapidly approach equilibrium with their food supply and ambient seawater concentrations (Harding 1986). This, however, is not the case for long-lived mammals where selective accumulation and metabolism increase the storage of specific congeners.

Chlordane-related compounds were present in slightly lower amounts than toxaphene and PCBs, while ΣHCH and HCB were the least concentrated. Published data for Arctic marine mammals show a similar proportionality in abundance between these classes of OCs (Norstrom and Muir 1994). The most abundant compounds in air and seawater samples (ΣHCH and HCB) (Chapter 2) are those with the lowest concentrations in Arctic marine biota from all trophic levels.

These data indicate that the biomagnification of various OCs in the marine food web depend on physical as well as biological factors. For example, compounds with low water solubility have a high lipid-water partitioning and are efficiently accumulated across the gills and via the gut of aquatic animals. Isomers and congeners of PCBs and DDT-DDE have water solubilities (10^{-8} to 10^{-10} g·L^{-1}) that are two to three orders of magnitude lower than values for HCH and HCB (Tanabe and Tatsukawa 1983). Although toxaphene is reported to be more water soluble (~10^{-3} g·L^{-1}) (Murphy *et al.* 1987, Bidleman *et al.* 1993) than PCBs, selected toxaphene components may have lower solubility. Less water

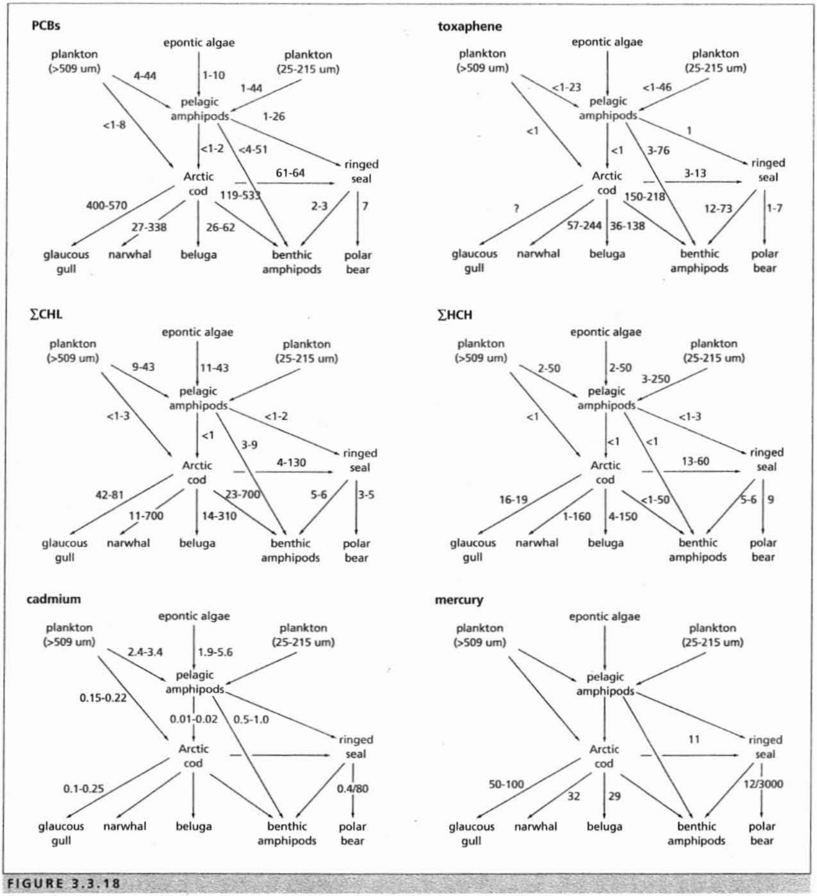


FIGURE 3.3.18

Biomagnification of organochlorines and heavy metals in the arctic marine food web. Biomagnification factors (BMFs) for organochlorines are calculated with lipid-normalized concentrations in predator and prey. For metals, BMFs are based on dry wt. concentrations in whole organisms (invertebrates + fish) and on wet wt. concentrations for fish to muscle of top predators. For polar bear, liver was used instead of muscle.

soluble compounds would tend to rapidly associate with particulate matter on addition to water. Suspended particulate matter concentrations in ice-covered regions of the Arctic Ocean are very low (Gordon and Cranford 1983), but increases occur during late summer due to production by under-ice epontic algae and phytoplankton in the upper water column (Hargrave *et al.* 1989). Rates of inorganic and organic particulate matter sedimentation also increase by an order of magnitude during August (Hargrave *et al.* 1994) and scavenging of particle-reactive OCs from the water column should be maximum at this time of year. Particle-adsorbed OCs are then available for uptake by grazing organisms or sedimentation and subsequent incorporation in the benthic food web. Heavier, less water-soluble

TABLE 3.3.12

Ranges of concentrations (ng·g⁻¹ lipid wt. and wet wt.) of various organochlorine pesticides and PCBs in lower trophic level marine biota.

	Region		Toxaphene	ΣPCB[3]	ΣDDT[4]	ΣCHL[5]	ΣHCH[6]	HCB	Reference
Epontic Particles	Ice Island (Axel Heiberg)	lipid wt.	<100	40–360	20–70	10–40	10–280	6–30	Hargrave *et al.* 1992
	Barrow Strait	lipid wt.	10–140	20–360	150–360	3–11	160–230	10–20	Hargrave 1994 (unpublished)
Zooplankton[1]									
25-125 µm	Ice Island (Axel Heiberg)	lipid wt.	10–890	10–490	8–150	5–150	2–200	1–100	Hargrave *et al.* 1992
>509 µm	Ice Island (Axel Heiberg)	lipid wt.	20–1,360	10–110	10–60	10–50	10–280	10–100	Hargrave *et al.* 1992
>500 µm	Barrow Strait	lipid wt.	200–1,400	4–20	2–20	8–107	90–180	5–130	Hargrave 1994 (unpublished)
Amphipods[2]									
Pelagic	Ice Island (Axel Heiberg)	lipid wt.	460	<440	<350	330	500	170	Hargrave *et al.* 1992
Pelagic	Barrow Strait	lipid wt.	50–800	1–230	3–60	4–78	70–390	1–40	Hargrave 1994 (unpublished)
Benthic (*Eurythenes*)	Arctic Ocean	lipid wt.	3,000–35,000	5,700–34,000	2,200–25,900	750–4,460	6–420	60–260	Hargrave *et al.* 1992
Benthic	Barrow Strait	lipid wt.	170–3,800	50–1,900	15–1,590	30–1,320	120–7,800	10–210	Hargrave 1994 (unpublished)
Fish									
Arctic cod	Lancaster Sound	lipid wt.	339–639	66–95	66–120	55–105	39–49		Muir & Lockhart 1994
		wet wt.	23–47	4–7	5–6	4–8	3–4		" "
	Barrow Strait	lipid wt.	300–3,900	14–230	15–255	30–600	40–150	30–190	Hargrave 1994 (unpublished)
Turbot	Cumberland Sound	lipid wt.	1,766–3,452	914–1,619	626–1,044	505–956	59–117	163–276	Muir & Lockhart 1996
		wet wt.	318–408	163–216	105–133	96–119	10–15	27–36	" "
	Beaufort sea	lipid wt.	1,725–3,217	875–1,605	659–1,251	658–1,246	78–147	144–273	Muir & Lockhart 1996
		wet wt.	282–427	140–216	111–163	111–163	13–19	26–36	" "
Greenland cod	Wellington Bay	wet wt.	—	0.79–2	—	—	—	—	Bright *et al.* 1995a,c
Four-horn sculpin	Wellington Bay	lipid wt.	—	171–521	92.9	450	100	—	Bright *et al.* 1995a,cc
		wet wt.	—	2.4–7.3	1.3	6.3	1.4		" "
Four-horn sculpin	Cambridge Bay	lipid wt.	—	2500	1225	255	95	—	Bright *et al.* 1995a,c
		wet wt.	—	50	24.5	5.1	1.9		" "
Four-horn sculpin	Hall Beach	lipid wt.	—	—	135	140	110	—	Bright *et al.* 1995a,c
		wet wt.	—	—	2.7	2.8	2.2		" "
Bivalves									
clams	Sanikiluaq	lipid wt	—	61.8	33.7	56.2	<5.6	—	Cameron & Weis 1993
		wet wt	—	1.1	0.6	1	<0.1		" "
Septentrion sp.	Manitounuk Sound	lipid wt	155	129.5	13	12.4	15.5	<0.5	Muir *et al.* 1995c
		wet wt	3	2.5	0.25	0.24	0.3	<0.01	" "

[1] *Zooplankton in the smaller size class were dominated by* Microcalanus sp.*and copepodites of* Calanus hyperboreus *with adults of the latter species in the larger size class.*

[2] *Pelagic amphipods were* Parathemisto sp. *and* Gammarus wilkitzkii. *Benthic amphipods included* Onisimus sp., Tmetonyx cicada, *and* Anonyx nugax

[3] *As Aroclor 1254 equivalents for epontic particles, zooplankton and amphipods. All others as sum of 80 PCB congeners.*

[4] *Sum of p,p'-DDT + p,p'-DDE + o,p'-DDE for epontic particles, zooplankton and amphipods. All others include o,p'-DDT.*

[5] *sum of cis- + trans-chlordane + cis-nonachlor + oxychlordane + heptachlor + heptachlor epoxide*

[6] *sum of α-, β- and γ-HCH*

compounds are preferentially accumulated by the lipid and organic carbon surfaces of suspended particulate matter and microalgae. In the complex mixture of toxaphene, which contains 6-carbon ringed compounds with a methylene bridge (bornanes) with 6–10 chlorine molecules, there is accumulation of the heavier Cl_8 and Cl_9 bornanes in arctic zooplankton and benthic amphipods in comparison to those in seawater (Bidleman *et al.* 1989). Comparisons with lower trophic levels and fish also show that more short-lived species may have lower concentrations (Hargrave *et al.* 1993). Zooplankton and pelagic amphipods generally have a one- to two-year life cycle and these species, feeding on microalgae and suspended particulate matter, accumulate less of the most abundant OCs than do more long-lived fish and benthic amphipod species.

The combined importance of life span and food source in affecting OC body burdens is evident in studies of the large benthic lysianassid amphipod *Eurythenes gryllus*, a benthic scavenger that may contain concentrations that equal or exceed those in mammals (Table 3.3.12). The life span of these organisms is not well known but it may exceed 10 years (Hargrave *et al.* 1992) — much greater than annual values for pelagic crustaceans. The food of *Eurythenes* is thought to be tissues of dead animals and the high levels of OCs could be explained if carcasses of marine mammals are a major component of their diet. In this case, although they consume non-living animals, lysianassid amphipods function as top predators in the Arctic marine food web. In shallow water, smaller-sized amphipods in pelagic and epontic populations are an important food resource for fish and seals. *E. gryllus* is restricted in its distribution to deep water where there is a refuge from predation due to the low abundance of fish.

There is very limited information on levels of OCs or PAHs in Arctic marine fish. Results for samples collected and analysed since 1990 are listed in Table 3.3.12. Results from earlier studies have been reviewed by Muir *et al.* (1992a). That review showed that toxaphene was the major OC contaminant in marine fish. In arctic cod collected at three eastern Arctic locations in the mid-1980s, toxaphene levels were five- to ten-fold higher than DDT or PCB residues.

Highest concentrations of OCs in arctic fish are found in turbot (Greenland halibut). These predacious, bottom feeding fish have a relatively fatty muscle compared with whitefish, char or sculpins. Samples from the eastern Canadian Arctic and the eastern Beaufort Sea both had mean toxaphene concentrations of 376 $ng \cdot g^{-1}$ (wet wt.) (Table 3.3.12). These levels are three to five times higher than in sea run char muscle and 15 to 20 times higher than arctic cod (whole fish) reflecting the higher trophic position of the turbot.

Metals in lower trophic level marine organisms

The data on metals in lower marine food organisms in the Canadian Arctic are very limited with the exception of the work on cadmium reported by Macdonald (1986) and Macdonald and Sprague (1988). They studied the food chain accumulation of cadmium in various trophic level samples, from phytoplankton to arctic cod, in the northern Baffin Island, Lancaster Sound and Jones Sound regions. Cadmium concentrations in invertebrates correlated with trophic level with the highest levels in carnivorous amphipods. Arctic cod were the exception, having lower cadmium than most crustaceans while feeding at the same trophic level. Highest concentrations of cadmium were observed in *Parathemisto libellula* (6.3 $\mu g \cdot g^{-1}$) and phytoplankton (4.7 $\mu g \cdot g^{-1}$), and the lowest in arctic cod (0.40 $\mu g \cdot g^{-1}$ whole body) and the amphipod *Mysis litoralis* (0.17 $\mu g \cdot g^{-1}$). Bohn and McElroy (1976) also found higher cadmium concentrations in zooplankton (2.8 $\mu g \cdot g^{-1}$) than in arctic cod (0.62 $\mu g \cdot g^{-1}$ whole fish) from Strathcona Sound. Concentrations of cadmium in arctic cod liver were about 10 times higher than in cod liver from the northern Baltic sea. Cadmium levels in carnivorous amphipods were similar to those found in other amphipods (Family: hyperiidea) in the north Pacific Ocean and were attributed to the carnivorous diet of these animals.

TABLE 3.3.13

Ranges of PCB concentrations in various environmental samples from Cambridge Bay, NWT (sum of 47 congeners; $ng \cdot g^{-1}$ wet wt., unless indicated otherwise).

	N	Cambridge Bay Concentration	N	Reference Area[1]
Water ($ng \cdot L^{-1}$)	8	0.045–1.8	2[2]	<0.008
Sediment ($ng \cdot g^{-1}$)	49	0.14–45	2	0.052–0.44
Bivalves				
Arctica islandica	2	<2.4[3]–1.9		not sampled
Mya truncata	4	0.89–2.2	1	<0.62[3]
Mytilus edulis	2	2.7–3.2		0.99
Sea Urchins				
S. droebachiensis	14	<1.0[3]–210	2	24–26
Amphipods	2	32–36		not sampled

[1] *From Queen Maud Gulf or Wellington Bay, Victoria Island, NWT*

[2] *Ice Island data from Hargrave* et al. *(1988). See also Table 3.3.12*

[3] *Concentrations < detection limit (DL) in some samples; a maximum DL is given.*

In Strathcona Sound, sea urchins in the vicinity of the Nanisivik lead/zinc mine had lead concentrations of 0.4 to 32.5 µg·g^{-1} (dry wt.), a definite indication of local contamination by lead (Fallis 1982). Lead concentrations in fucoid algae (*Fucus distichus*) from the same area averaged 0.8 µg·g^{-1} and did not vary with distance from the mine (Bohn 1979).

A substantial amount of metals data is available on lower trophic level organisms from western Greenland which may be relevant to the eastern Canadian Arctic because of similar geology and ecology (Dietz *et al.* 1995, Riget *et al.* 1995). Similar to observations in the eastern Canadian Arctic, cadmium, mercury and selenium levels are relatively high in marine organisms. Lead levels in marine organisms from Greenland were relatively low except in molluscs (medians ranging from 0.08–1.7 µg·g^{-1}) and algae (0.18–0.36 µg·g^{-1}).

Cadmium concentrations increased in the order fish <crustaceans <toothed whales/seals/birds. Cadmium concentrations were higher in algae from the open sea and lowest in the inner regions of large fiords (Riget *et al.* 1995).

There are limited data on mercury concentrations in invertebrates from western Greenland. Mercury concentrations were low in molluscs (means of 0.011–0.023 µg·g^{-1}) and in most crustaceans (medians of 0.04–0.88 µg·g^{-1}). Highest concentrations were found in deep sea prawns from Baffin Bay.

3.3.3.2 PCBs and Other Organochlorines in Nearshore Benthic Animals of Cambridge Bay

A case study was carried out in Cambridge Bay, NWT, during August in 1991, 1992 and 1993 to identify the scale of local redistribution into the sea of PCBs and metal contaminants from the local DEW line site and hamlet (see Sect. 3.2.5 for additional information on DEW line contamination) (Bright *et al.* 1995b). A secondary objective was to examine the uptake in coastal benthos of PCBs and other OCs from sources other than local shoreline sites, including long-range transport and ocean-dumped debris. The biomagnification of organic contaminants in foodwebs tied to benthic environments has been examined in more detail in temperate coastal marine environments (e.g., New Bedford Harbour, Connolly 1991, or Puget Sound, Varanasi *et al.* 1987) or in freshwater systems (Macdonald *et al.* 1993, Kidd *et al.* 1995a) but has received little study in Arctic coastal environments.

PCB levels in benthic animals collected in Cambridge Bay, and from two reference areas in Queen Maud Gulf (Fig. 3.3.19) are listed in Table 3.3.13. There are few comparable data on OC levels in benthic marine invertebrates or bottom-dwelling fish in the Canadian Arctic because most samples have been collected at sites remote from radar facilities and harbours (Table 3.3.12). Scallops, *Chlamys islandicus*, collected from near the Thule Airforce Base in Greenland (Kjolholt and Hansen 1986) contained from 1 to 6 ng·g^{-1} (wet wt.) PCBs, a similar range to that observed in bivalves from Cambridge Bay. Sculpin, *Myoxocephalus quadricornis* or *M. scorpius*, (whole tissue) collected in Cambridge Bay had relatively high levels of PCBs compared to samples collected from areas primarily influenced by atmospheric contaminant sources (Wellington Bay; Hall Beach) (Table 3.3.12).

The spatial variation in sculpin of PCBs, DDT and toxaphene in upper Cambridge Bay (Fig. 3.3.19) suggested that:

1. The bioaccumulation of PCBs and DDT residues in upper Cambridge Bay is attributable to local inputs (Bright *et al.* 1995b); and,
2. Historical ocean disposal, although extensive in Cambridge Bay, has not contributed appreciably to marine contaminant inputs.

   Bright *et al.* (1995 a,c) observed that all OCs were elevated in sculpins collected closer to areas of riverine input (Figure 3.3.19), both in Cambridge Bay and in Wellington Bay (a reference area in Queen Maud Gulf). This suggests that riverine input to coastal areas of atmospherically-transported contaminants may be a significant source to nearshore marine areas.

The OCs with the highest detectable concentration in mussels, *Mytilus edulis*, were hexachlorocyclohexane (ΣHCHs: α–HCH + β–HCH + γ–HCH), at approximately 0.7 ng·g^{-1} (ng·g^{-1} wet wt.) and dieldrin (0.2 to 0.4 ng·g^{-1}). Organochlorines detected in sea urchins, excluding DDT group compounds and PCBs (which were present mainly as a result of local sources), included HCB (0.1 to 0.6 ng·g^{-1}), HCHs (primarily α–HCH: 0.6 to 3.4 ng·g^{-1}), nonachlors (including *trans* and *cis*: 0.4 to 0.9 ng·g^{-1}), other chlordanes (oxychlordane, *trans*- and *cis*-chlordane: 0.05 to 0.9 ng·g^{-1}) and dieldrin (0.1 to 0.6 ng·g^{-1}). Benthic amphipods, *Orchomene* sp. contained approximately 9 to 10 ng·g^{-1} of HCB, 6.4 to 6.9 ng·g^{-1} α–HCH, 4.6 to 5.9 ng·g^{-1} chlordanes, 9.2 to 10.8 ng·g^{-1} nonachlors, and 2.3 to 2.8 ng·g^{-1} dieldrin. The major OC groups found in whole tissues of higher benthic predators such as sculpin or Greenland cod, both within Cambridge Bay and at the reference sites, included PCBs (Tables 3.3.12, 3.3.13), DDT and related products (from 0.9 to 61 ng·g^{-1}), and toxaphene, or polychlorobornanes (total toxaphene levels ranged from 1.5 to 27 ng·g^{-1}) (see also Bright *et al.* 1995b). Toxaphene in sculpin or Greenland cod was invariably dominated by two compounds: 2-exo,3-endo,5-exo,6-endo,8,8,10,10-octachlorobornane and 2-exo,3-endo,5-exo,6-endo, 8,8,9,10,10,-nonachlorobornane. Coplanar PCBs detected in four-horn sculpins included CB77 (3,3',4,4'-TCB) and CB126 (3,3',4,4',5-PCB). The percent composition of these coplanar PCBs in sculpin whole tissue was

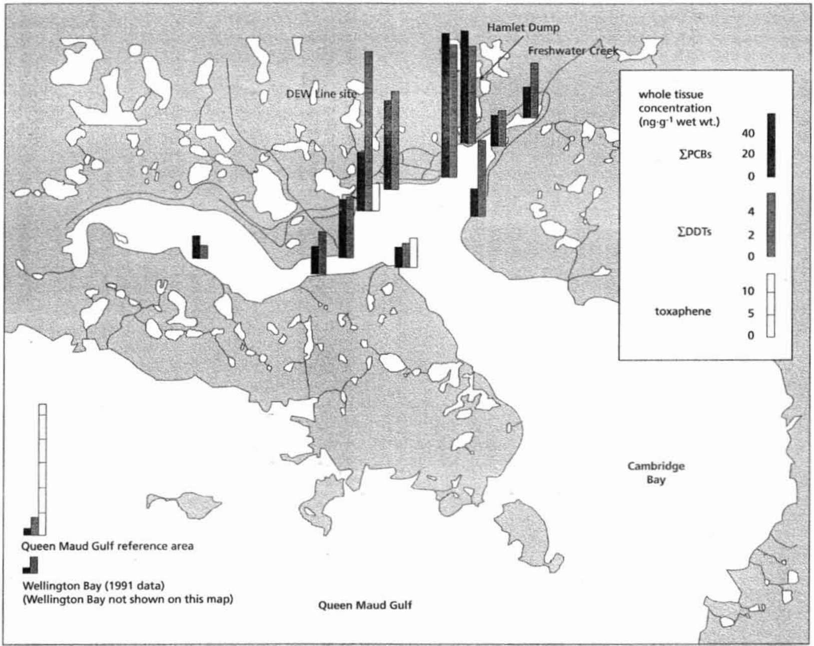


FIGURE 3.3.19

Spatial variation of PCBS, DDTs and toxaphene (analysed at a small number of sites) levels in four-horn sculpin whole tissues collected from Cambridge Bay (1993).

uniform across a wide range of PCB levels: CB77 and 126 comprised 0.017% and 0.028% of ΣPCB congener concentrations in Cambridge Bay four-horn sculpins.

Knowledge of the specific source and composition of PCB inputs into Cambridge Bay is useful for allowing a better understanding of mechanisms of uptake into a benthic food web. There was a shift from OCs with greater aqueous solubility (such as HCH) in primary consumers (filter-feeding clams) to OCs with lower aqueous solubility and higher lipophilicity (higher K_{ow}) in predatory fish. This shift is also found in the pelagic marine food web from pelagic zooplankton to predatory marine mammals in the Arctic (Norstrom and Muir 1994). The composition of PCBs (as 47 individual congeners) in four-horn or short-horn sculpins was different than that of the original source which was primarily the Aroclor 1254 technical mixture used in electronic equipment at the DEW Line site or found in the hamlet dump. The different congener composition in the biota may be due in part to metabolic modification in fish and preferential excretion of congeners which are more readily oxidized by the mixed function oxygenase enzyme system. Metabolic modification of PCBs in fish has been reported by several researchers including Sijm *et al.* (1992), Bruggeman (1983), Boon *et al.* (1989), and Varanasi *et al.* (1987). The congener pattern could also differ because of greater movement of more volatile and water soluble congeners (i.e. di-, tri- and tetrachlorobiphenyls) from the dump sites to the marine environment and because of contributions from atmospheric sources.

The levels in Arctic marine animals of non-ortho-substituted (or coplanar) PCBs, i.e., those with dioxin-like toxicity, is a more direct measure of potential for toxic effects in the animals themselves or their predators.

Greenland cod (*Gadus ogac*) are generally not considered to be a component of benthic food webs. They exhibit a much broader range of movement than most benthic fish, and probably spend only a small portion of the year in coastal inlets. In

Cambridge Bay, however, Greenland cod were observed to feed predominantly on small (<15 cm length) sculpins in shallow areas (based on the analysis of stomach contents). Elevated levels of PCBs in four-horn sculpins, therefore, are accompanied by elevated levels in Greenland cod relative to fish from outside of Cambridge Bay. Levels of OCs in four-horn or short-horn sculpins probably reflect the high affinity of these contaminants for sediments, and the trophic status of these fish within benthic communities. The limited mobility of sculpins makes them suitable bioindicators of contaminant transfer pathways in association with both local coastal inputs and terrigenous runoff of previously deposited atmospheric contaminants.

The biomagnification of PCBs and other OCs in Greenland cod that feed in Cambridge Bay provides one example of the transfer of persistent OCs from near-shore sediments to more pelagic predators that feed on benthic fauna in the Arctic.

3.3.3.3 Spatial Trends of Organochlorines and Metals in Marine Mammals

In comparison with the relatively limited data on OC and heavy metal contaminants in the lower marine food chain organisms, there exists a large database on OC and heavy metals contaminants in Arctic marine mammals (Muir *et al.* 1992a). Most of these data were generated in the period 1985-1991 by the DIAND Arctic Contaminants Program, which preceded the NCP. A review of the data to 1991 concluded that this dataset was limited spatially and temporally (DIAND 1991). The following sections summarize the existing data to 1991 and review the more recent results obtained since then, which have filled in some of the knowledge gaps on spatial and temporal trends.

Spatial trends of organochlorines in beluga, ringed seal and walrus

The most extensive measurements of OCs, especially of PCBs and DDT-related compounds, have been on blubber samples from beluga whales, ringed seals (Table 3.3.14), and polar bears. In small-toothed arctic whales (beluga and narwhal) toxaphene is the most prominent OC. A mean concentration of 9.2 $\mu g \cdot g^{-1}$ toxaphene was found in male narwhal blubber from Baffin Bay (Muir *et al.* 1992b) and similar or slightly higher levels were found in male belugas (6.0–14.5 $\mu g \cdot g^{-1}$) (Muir *et al.* 1990b). The toxaphene pattern in beluga and narwhal blubber is dominated by two components, an octachlorobornane and a nonachlorobornane, which account for about 70% of total toxaphene. These two components were also prominent in zooplankton and predatory amphipods analysed by Bidleman *et al.* (1989). The structure of these two chlorobornanes has been identified (Stern *et al.* 1992) and analytical standards have been prepared (Burhenne *et al.* 1993). Toxaphene concentrations in ringed seals are much lower (0.2–0.4 $\mu g \cdot g^{-1}$) and show a different pattern of components compared with whales or fish (Bidleman *et al.* 1993). Chlorobornanes are also present in polar bear tissues (Zhu and Norstrom 1994) at levels similar to ΣDDT and ΣHCH. This work was carried out with a single sample. No spatial trends in toxaphene residues in polar bears have been determined.

Few statistically significant differences in the mean concentrations of the major OCs were found in the five beluga stocks sampled in the period 1992-95 (Figure 3.3.20). ΣPCBs and ΣDDT levels showed no geographical trends, however, toxaphene was significantly higher ($p < 0.05$) in male beluga from the three eastern arctic stocks than in samples from Beaufort Sea animals. No significant differences were found among mean levels in females within the five stocks. Levels of OCs in belugas from western Greenland were similar to those in the southeast Baffin animals (Stern *et al.* 1994). PCB concentrations averaged about 6 $\mu g \cdot g^{-1}$ in male belugas, and 4 $\mu g \cdot g^{-1}$ in blubber of females (Figure 3.3.20). These levels are about 12 times lower (in males) than in blubber of dead beluga from the St. Lawrence estuary.

Organochlorine levels in blubber of ringed seals collected in the period 1984-1989 from 10 locations (including the western and central High Arctic, northwest and northeast Hudson Bay, Baffin Bay, Ungava Bay) have been summarized in Muir *et al.* (1992a). PCBs, ΣCHLOR and ΣDDT are the most prominent contaminants, while toxaphene (or chlorobornanes) is present at lower concentrations. Concentrations of PCBs and DDT in ringed seals from 12 locations sampled in the mid- and late-1980s (Muir *et al.* 1995b, Cameron *et al.* 1996, Cameron and Weis 1993) were quite similar in males and females from all locations. Significantly higher average concentrations were found in Hudson Bay males than in males from more northerly locations (Muir *et al.* 1994). More recent analyses of ringed seal blubber samples from Arviat, Pangnirtung and Inukjuak (all collected during spring 1992) also found higher levels of OCs in Hudson Bay animals (Figure 3.3.20). Concentrations of PCBs in females from Arviat and Inukjuaq were similar (1115±425 and 1301±1560 $ng \cdot g^{-1}$, respectively) while females from Pangnirtung had much lower levels (467±195 $ng \cdot g^{-1}$) (Table 3.3.14). Cameron *et al.* (1996) also found significantly higher concentrations of ΣPCB in male ringed seals from the Belcher Islands than from Holman in the western Arctic.

Walrus blubber samples from East Hudson Bay (Inukjuak and Akulivik), Foxe Basin (Igloolik and Hall Beach) and East Baffin Island (Loks Land) were recently found to have higher than expected levels of PCB congeners (ortho and non-ortho substituted) and other persistent OCs (DDT, toxaphene,

TABLE 3.3.14

Mean concentrations (ng·g⁻¹ ± S.D. wet wt.) of organochlorines in marine mammal samples from arctic waters (nd = not determined or data not available).

Species	Region	Location	Tissues[1]	Sex	Date	N	% Lipid	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Ref[2]
Ringed Seal	W. Hudson Bay	Arviat	B	F	1992	24	91.1 ± 4.7	63.2 ± 35.4	335 ± 204	806 ± 330	840 ± 398	1115 ± 425	297 ± 304	91.1 ± 54.6	2
				M		30	92.4 ± 3.4	68.9 ± 35.6	336 ± 127	1596 ± 1119	1662 ± 1103	2066 ± 1390	661 ± 1995	107 ± 72.5	2
	E. Baffin Is.	Pangirtung	B	F	1994	10	95.4 ± 2.4	37.6 ± 9.4	179 ± 21.8	322 ± 129	359 ± 166	467 ± 195	175 ± 65.8	71.3 ± 33.5	2
		(Cumberland S.)		M		10	95.3 ± 2.7	36.1 ± 7.1	210 ± 36.9	470 ± 324	703 ± 890	675 ± 597	180 ± 83.9	64.4 ± 21.2	2
	E. Hudson Bay	Inukjuaq	B	F	1989-92	7	92.9 ± 5.3	54.8 ± 53.0	276 ± 231	1008 ± 1338	1006 ± 1164	1301 ± 1560	307 ± 327	82.0 ± 100	2
				M		4	94.7 ± 2.0	61.6 ± 17.5	275 ± 123	708 ± 453	1143 ± 681	1234 ± 636	221 ± 112	86.6 ± 102	2
	N. Baffin Is.	Arctic Bay	B	F	1993	10	93.0 ± 1	80.5 ± 20.4	201 ± 67.9	299 ± 92.3	189 ± 58.7	296 ± 66.8	177 ± 80.6	64.5 ± 27.4	2
				M		10	91.2 ± 3	111 ± 50.5	250 ± 98.6	1272 ± 978	1542 ± 1905	1435 ± 1203	547 ± 441	115 ± 61.9	2
	Lancaster S.	Barrow Strait	B	F	1993	10	92.7 ± 0.9	87.7 ± 26.8	310 ± 141	466 ± 119	334 ± 108	535 ± 154	146 ± 47.3	78.4 ± 36.6	2
				M		10	92.9 ± 1.1	103 ± 19.0	384 ± 102	478 ± 156	365 ± 168	655 ± 184	163 ± 97.1	93.9 ± 37.9	2
	SW Ellesmere Is.	Eureka	B	F		7	91.5 ± 2.1	187 ± 96.2	255 ± 108	613 ± 189	673 ± 261	1267 ± 315	441 ± 136	96 ± 30.2	
				M		9	90.8 ± 1.9	185 ± 80.8	302 ± 121	1000 ± 344	1033 ± 266	2055 ± 707	561 ± 226	130 ± 44.2	
	Amundsen Gulf	Holman	B	M	1989	4		nd	117 ± 38.5	143 ± 49.8	269 ± 211	241 ± 44.3	nd	nd	3
	Holman			F	1991						217 ± 170	366 ± 110	nd	nd	4
	S. Hudson Bay	Sanikiluaq	B	M	1989	3	87.2	nd	435 ± 175	393 ± 96.7	1652 ± 700	1284 ± 323	nd	187 ± 113	5
				F	1991	6		nd	69.4 ± 17.3	225 ± 55.7	168 ± 24.8	562 ± 417	nd	40.8 ± 18.1	3
				M		10		nd	68.3 ± 31.7	358 ± 102	261 ± 59.9	1157 ± 416	nd	30.9 ± 33.8	
Harp Seal	Hudson Strait	Salluit	B	F		8	nd	nd	nd	nd	486 ± 289	897 ± 295	nd	nd	1
Beluga	S. Beaufort Sea	Mackenzie Delta	B	F	1992-95	9	89.3 ± 2.9	899 ± 171	337 ± 83.7	2406 ± 422	3271 ± 1210	5515 ± 1824	6061 ± 2298	397 ± 94.0	2
				M	1992-95	26	89.9 ± 2.6	956 ± 128	334 ± 93.7	2486 ± 319	3443 ± 641	5010 ± 1618	6335 ± 2021	410 ± 124	2
	S. Beaufort Sea	Husky Lakes	B	M	1989	8	83.9 ± 5.8	798 ± 309	429 ± 116	2003 ± 699	3262 ± 1548	4879 ± 1557	6859 ± 3166	479 ± 189	2
				F	1989	5	80.3 ± 4.1	315 ± 159	361 ± 157	683 ± 288	598 ± 275	1506 ± 640	2571 ± 1060	207 ± 89.1	
	S. Hudson Bay	Sanikiluaq	B	F	1994	10	93.4 ± 2.2	254 ± 122	335 ± 86.9	1450 ± 943	1697 ± 1391	2185 ± 1559	7185 ± 3614	306 ± 202	
				M		7	94.6 ± 0.8	460 ± 255	442 ± 133	4167 ± 1585	11200 ± 8231	6768 ± 2346	14150 ± 7041	923 ± 360	
	SE Baffin Is.	Kimmirut	B	F	1993-94	5	93.0 ± 1.3	664 ± 388	255 ± 139	2890 ± 1310	4134 ± 2150	4643 ± 2210	8300 ± 3563	786 ± 398	
				M		6	91.2 ± 3.9	1305 ± 781	515 ± 163	4236 ± 1669	6964 ± 3303	6794 ± 2171	14550 ± 6361	1447 ± 661	
	Cumberland S.	Pangnirtung	B	F	1993-94	2	93.3	446	366	3658	7164	6415	13600	736	
				M		3	92.8 ± 3.2	808 ± 389	494 ± 155	3465 ± 884	4559 ± 1417	1505 ± 1200	12400 ± 3777	932 ± 339	
	S. Beaufort Sea	Mackenzie Delta	M	M+F		20	4.3 ± 1.4	34.0 ± 12.1	21.2 ± 9.0	133 ± 54.2	202 ± 104	246 ± 102	372 ± 172	24.0 ± 9.84	1
	NW Greenland	Nuussuaq	M	M+F		20	10.2 ± 4.3	81.6 ± 76.7	29.7 ± 23.2	353 ± 208	413 ± 266	373 ± 196	1526 ± 899	85.9 ± 60.7	6
			K	M+F		20	3.4 ± 1.4	34.0 ± 18.5	8.3 ± 6.5	85.2 ± 46.6	97.6 ± 63.5	122 ± 68.6	328 ± 214	6.98 ± 3.01	6
Narwhal	Landcaster S.	Creswell Bay	B	M	1991	8	91.4 ± 1.9	267 ± 116	85.0 ± 16.7	1994 ± 380	3232 ± 872	3195 ± 765	7927 ± 2411	228 ± 52.1	1
Walrus	N. Hudson Bay	Foxe Basin	B	M	1988	12	80.3 ± 5.3	23.2 ± 7.6	99.5 ± 15.1	312 ± 64.4	32.8 ± 8.9	199 ± 46.2	451 ± 116	154 ± 42.0	7
				F		12	80.9 ± 5.2	15.3 ± 4.1	62.6 ± 12.7	166 ± 98.9	20.8 ± 12.9	90.0 ± 54.6	201 ± 123	71.8 ± 37.0	
	E. Baffin Is.	Loks Land	B	M+F		6	82.4 ± 6.0	54.8 ± 33.9	156 ± 81.5	2083 ± 2186	605 ± 709	1643 ± 1577	359 ± 202	284 ± 184	7
	E. Hudson Bay	Inukjuak	B	M	1989	4	84.5 ± 4.7	66.6 ± 33.3	267 ± 149	6293 ± 4236	4609 ± 3165	11512 ± 8465	3493 ± 2443	911 ± 436	7
				F		9	82.4 ± 4.2	61.9 ± 47.9	214 ± 114	2753 ± 1784	2164 ± 924	4790 ± 2376	1451 ± 954	464 ± 428	
	NE Hudson Bay	Akulivik	B	M	1989	8	83.2 ± 5.5	23.4 ± 12.6	116 ± 76.8	1169 ± 2105	561 ± 1087	1201 ± 2056	420.5 ± 286	251 ± 225	7
				F		2	87.4	16.4	126	238	45.5	195	606	209	

[1] *B = blubber; M = muktuk, K = kidney*

[2] *References*
1. Beck et al. *1994*
2. Muir 1994, 1996
3. Cameron et al. *1996*
4. Addison 1995
5. Cameron and Weis 1993
6. Stern et al. *1994*
7. Muir et al. *1995c*

chlordanes, dieldrin, mirex) when compared with previous studies of walrus from Greenland and Alaska (Muir *et al.* 1995c). Samples from 19 of 53 individuals had concentrations of ΣPCBs greater than 1000 ng·g^{-1} (wet wt.); the remaining individuals had much lower concentrations (50–600 ng·g^{-1}). Local contamination was ruled out because levels of all OCs were elevated in each animal from Inukjuak and elevated levels were also found in animals from other areas of the eastern Canadian Arctic. Skaare *et al.* (1994a) also found a wide range of concentrations of ΣPCB, ΣDDT and chlordane-related compounds in walrus skin/blubber biopsy samples from Svalbard. Concentrations of ΣPCB (mean 11.5 µg·g^{-1}, median

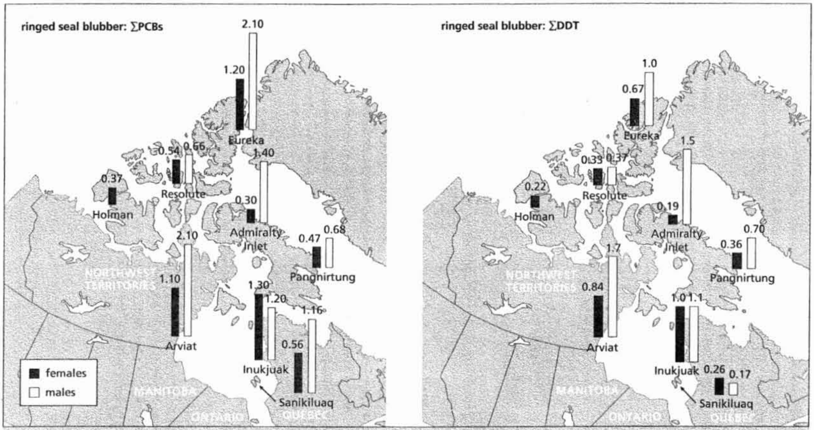


FIGURE 3.3.20 A

ΣPCBs and ΣDDT in ringed seal blubber (collected 1989-94) from Muir (1994, 1996), Cameron et al. *(1996) and Addison (1995). Concentrations µg·g^{-1} wet wt.*

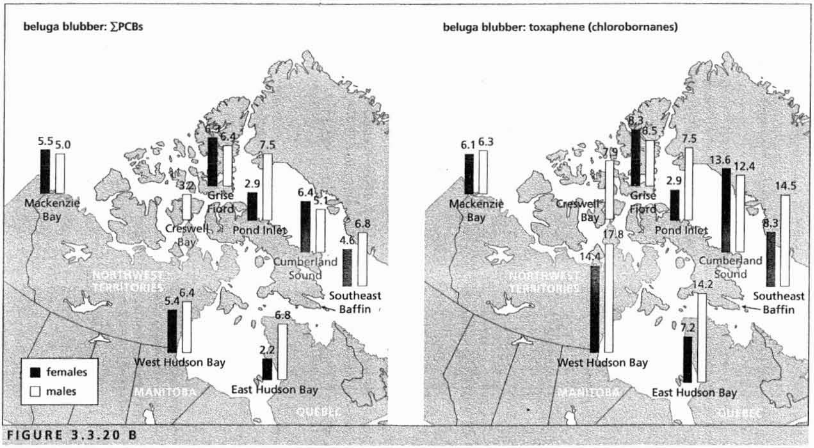


FIGURE 3.3.20 B

ΣPCBs and toxaphene in beluga blubber collected 1992-95 (Muir 1994, 1996). Concentrations µg·g^{-1} wet wt.

8.8 μg·g^{-1} lipid) in Svalbard walrus were similar to those from E. Hudson Bay but much higher than Foxe Basin animals (Table 3.3.14). The results suggest that the walrus with elevated OCs are feeding at a higher trophic level than those with low levels and are probably utilizing ringed seals for a portion of their diet.

Effects of age and sex on organochlorine levels

Age and sex are important factors to be taken into account when comparing geographical and temporal trends in marine mammals. Most OCs, except HCH and mirex, are lower in adult females than in male whales and seals because of elimination of these lipophilic compounds via lactation (Addison and Smith 1974). But trends with age vary with species and sex. Stern *et al.* (1994) found no significant trend of OC levels with age in male beluga (70 animals were analysed) and a substantial decrease in females after five or six years of age corresponding to the time of first parturition. The variation of ΣPCBs and ΣDDT with age in Arviat ringed seals is illustrated in Figure 3.3.21. Concentrations increase with age in males but not in females. This has been observed previously (Addison and Smith 1974) but the large number of samples from Arviat (35 males, 30 females) enabled a thorough test of the age/concentration relationship. Geographical comparisons are thus best done with adult female ringed seals and with male belugas.

Organochlorines in other marine mammal tissues

Fewer analyses of kidney, liver and muktuk than of blubber of marine mammals have been conducted. This reflects the fact that OC concentrations are uniformly lower in tissues other than blubber corresponding to lower lipid content in these tissues. Low ng·g^{-1} (wet wt.) levels of PCBs and OC pesticides were found in liver and muscle samples of beluga and narwhal (Muir *et al.* 1990b,1992b). In beluga liver, PCBs and toxaphene concentrations averaged 124 and 120 ng·g^{-1} wet wt., respectively, in males, unlike blubber where toxaphene was more prominent. On a lipid weight basis (beluga liver and muscle averaged 3.4 and 1.3 % extractable lipid, respectively) levels of OCs were comparable to, or higher than, those found in blubber (Muir *et al.* 1990b).

Muktuk from the Beaufort Sea contained low ng·g^{-1} levels of all major classes of OC contaminants (PCBs, chlorobenzenes, OC pesticides) (Table 3.3.14). The lipid content of muktuk ranged from 3% to 7% which indicates it was relatively free of underlying blubber. Toxaphene was the major OC contaminant in beluga muktuk with mean concentrations of 372 ng·g^{-1} wet wt. Muktuk from western Greenland had similar concentrations of OCs to those in Beaufort Sea beluga samples (Stern *et al.* 1994). In both locations, PCBs were higher in muktuk from males than from females similar to observations for blubber. Cameron and Weis (1993) also found relatively high ΣPCBs, ΣDDT and chlordane-related compounds in beluga muktuk from Sanikiluaq. Beluga kidney from Greenland animals had lower levels of PCBs and most other OCs than muktuk and showed unexpected differences between the sexes with about two-fold lower levels in females.

Spatial trends in heavy metals in marine mammals

Measurements of metal contaminants in tissues of Arctic marine mammals collected in the 1970s and 1980s indicated that mercury, cadmium and lead were relatively high in some marine mammals but information on spatial trends was limited (Muir *et al.* 1992a). The impetus for a systematic investigation of metal contaminants in marine mammals in the Canadian Arctic derived largely from the early indication that mercury levels in marine mammal tissues were relatively high, i.e. exceeding the Health Canada consumer guideline (for fish flesh) of 0.5 μg·g^{-1} wet wt. In the case of cadmium in narwhal (Wagemann *et al.* 1983), the levels were sufficiently high to also raise concerns that cadmium might be sublethally toxic to the animals themselves.

Metals in Ringed Seals

Recent work, summarized in Wagemann *et al.* 1995 and Wagemann 1994, has resulted in a large metals data base for ringed seals from the western Arctic, and more limited information is available for the rest of northern Canada. Animals were sampled (and analysed) in different years and from different areas; 117 from the western Arctic (1987/88), 35 from the central Arctic (1993), and 40 from Hudson Bay and the eastern Arctic. The resulting data base showed significant geographic trends for mercury and cadmium. Ringed seals in the western Arctic had

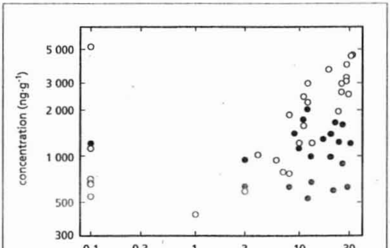


FIGURE 3.3.21

Variation of ΣPCB with age in ringed seals (W. Hudson Bay, Canada). A total of 24 females and 30 males were analysed. ΣPCB in males was significantly correlated with age (log ΣPCB (y) vs. log age (x); $r^2 = 0.88$; $P<0.01$). Similar correlations were observed for ΣDDT and ΣCHL.

higher concentrations of total mercury in the liver than those in the eastern and central Arctic (Figure 3.3.22A), and a lower concentration of cadmium in the liver and kidney, than those in the eastern Arctic and Hudson Bay (Figure 3.3.22B).

Although the mean ages of the western (7.4±5.1 years) and eastern (6.1±4.6 years) arctic ringed seals were similar, they differed in total mercury concentration in the liver by nearly a factor of four (32.9 and 8.34 $\mu g \cdot g^{-1}$ wet wt., respectively) (Table 3.3.15). Most mercury in the liver was inorganic mercury and was positively correlated with selenium. The mean methylmercury concentration in the liver (0.87 and 0.75 $\mu g \cdot g^{-1}$ wet wt., western and eastern Arctic, respectively), and in the muscle (0.43 and 0.45 $\mu g \cdot g^{-1}$ wet wt., respectively) was not significantly different in animals of different regions. Methylmercury represented approximately 90% to 100% and 2% to 9% of the total mercury in muscle and liver, respectively.

The spatial trend for cadmium was opposite to that for mercury, i.e. it was higher in the eastern Arctic than the western Arctic for ringed seals (Table 3.3.15) and the rate of accumulation of cadmium in the liver was higher in the eastern Arctic (Wagemann *et al.* 1996b).

Metals in belugas

In 1993, beluga were sampled during Native hunts at four locations (27 animals), and in 1994 at two locations (46 animals) in the western Arctic. Belugas were also sampled in 1981 (35 animals) and in 1984

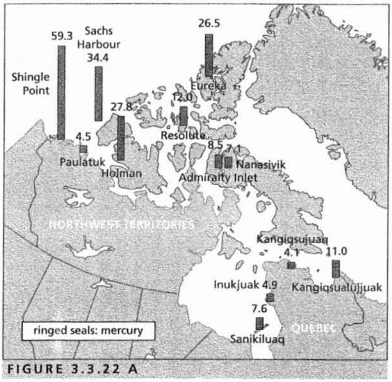


FIGURE 3.3.22 A

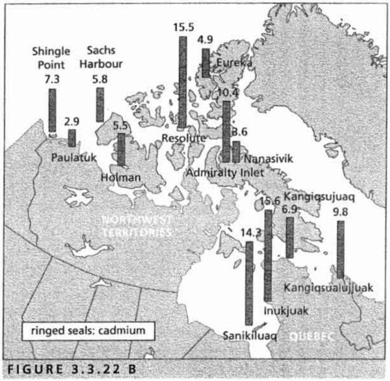


FIGURE 3.3.22 B

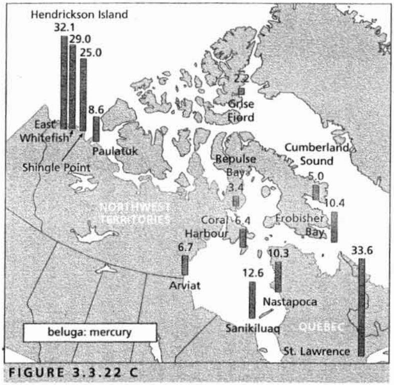


FIGURE 3.3.22 C

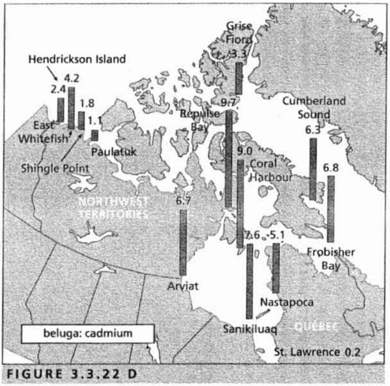


FIGURE 3.3.22 D

Mean mercury and cadmium in ringed seal and beluga liver ($\mu g \cdot g^{-1}$ wet wt.). (Wagemann et al., *1996b)*

TABLE 3.3.15

Mean concentrations (µg·g⁻¹ ± S.D. wet wt.) of metals in various marine mammal tissues from the Canadian Arctic. Average age in years is given for beluga and ringed seal and average length in cm is given for narwhal (Wagemann *et al.* 1996b).

Species	Location	Years	Age in Years	(N)[3]	Tissue[1]	N[4]	Cadmium	Copper	Lead	Mercury[2]	Selenium	Zinc
Beluga	Western Arctic	1993-1994	19.3 ± 6.6	(71)	MK	28-65	0.002 ± 0.001	0.47 ± 0.08		0.78 ± 0.41	4.02 ±1.17	65.8 ± 4.8
					M	76	0.019 ± 0.015	0.74 ± 0.16		1.34 ± 0.67	0.41 ± 0.088	25.7 ± 6.8
					L	77	2.27 ± 1.04	11.3 ± 7.09		27.1 ± 24.7	18.8 ± 13.9	27.9 ± 5.01
					K	78-79	9.68 ± 3	2.1 ± 0.28		4.91 ± 2.84		26.4 ± 3.88
	Eastern Arctic	1984-1994	11.9 ± 6.0	(143)	MK	43-45	0.009 ± 0.01	0.51 ± 0.08		0.59 ± 0.22	4.75 ± 1.43	86.6 ± 14.4
	(Hudson Bay,				M	136-138	0.079 ± 0.27	0.98 ± 0.2		0.94 ± 0.44	0.4 ± 0.17	24.4 ± 6.12
	Southeast Baffin)				L	139	6.51 ± 4.88	19.2 ± 32.7		8.4 ± 8.25	5.35 ± 3.18	28.8 ± 8.06
					K	135-139	22.4 ± 13	2.78 ± 0.49		3.1 ± 1.71		29.4 ± 1.71
	Whole Arctic	1984-1994			MK	60			0.01 ± 0.04			
					M	212			0.01 ± 0.04			
					L	215			0.02 ± 0.03			
Ringed Seal	Western Arctic	1987-1993	7.4 ± 5.1	(145)	M	118-113	0.041 ± 0.04	1.09 ± 0.16	0.05 ± 0.05	0.41 ± 0.29	0.51 ± 0.14	27.1 ± 5.31
	(Sachs Harbour,				L	140-142	5.6 ± 3.14	10.8 ± 6.65	0.08 ± 0.12	32.9 ± 35.2	15.2 ± 12.9	41.5 ± 5.78
	Holman)				K	140-144	21.1 ± 14.2	6.48 ± 2.08		2.05 ± 1.34		38.7 ± 10.7
	Eastern Arctic	1989-1993	6.1 ± 4.6	(114)	M	35-61	0.098 ± 0.09	1.2 ± 0.2	0.01 ± 0.01	0.39 ± 0.17	0.33 ± 0.04	23.2 ± 6.09
	(N.Québec,				L	115-133	11.9 ± 9.2	13.4 ± 7.8	0.01 ± 0.01	8.34 ± 7.03	4.81 ± 3.12	47.9 ± 8.49
	Arctic Bay, Resolute)				K	35	47.7 ± 23.3	6.02 ± 1.68		1.49 ± 0.58		50.4 ± 11
	Eureka	1994	12.9 ± 12.2	(18)	M	16-18	0.032 ± 0.03	1.35 ± 0.033		0.66 ± 0.42	0.33 ± 0.098	22 ± 4.86
					L	18	4.65 ± 3.78	8.8 ± 7.22		26.5 ± 36.9	14 ± 16.7	44.6 ± 7.87
					K	17-18	15.6 ± 13.8	5.54 ± 2.4		3.17 ± 1.91		40.8 ± 7.42
			Length (cm)									
Narwhal	Eastern Arctic	1992-1994	420 ± 57	(26)	MK	47-48	0.018 ± 0.01		0.002 ± 0.002	0.59 ± 0.18		
	(Creswell Bay)				M	55-56	0.21 ± 0.19		0.008 ± 0.002	1.03 ± 0.37		
					L	55	29.7 ± 25.4		0.026 ± 0.018	10.8 ± 8.05		
					K	55	54.1 ± 24.1			1.93 ± 1.12		
	Arctic (combined)				MK	47-48		0.34 ±0.057			5.05 ± 2.38	64.8 ± 9.92
					M	55-56		0.79 ± 0.18			0.4 ± 0.043	24.9 ± 8.85
					L	55		6.3 ± 2.59			7.35 ± 4.37	40.3 ± 14.8
					K	55		2.93 ± 0.62				34 ± 7.15
Walrus	Eastern Arctic	1992			M	90-113	0.17 ± 0.12			0.11 ± 0.14		
	(Foxe Basin/eastern			L	125-130	11.1 ± 6.4			1.43 ± 1.21			
	Hudson Bay)				K	108-112	57.6 ± 30.4			0.31 ± 0.11		

[1] *MK=muktuk, M=muscle, L=liver, and K=kidney*

[2] *Total mercury*

[3] *Number of samples used to calculate mean age*

[4] *Number of samples analysed. Range gives variation in number of samples analysed for the various metals. See Wagemann* et al. *1996b for further detail.*

(8 animals) in the Mackenzie Delta. Fewer metal contaminants data are available for belugas from the eastern Arctic.

The mean total metal concentrations ($\mu g \cdot g^{-1}$ wet wt.) in liver, kidney, muscle, and muktuk of all western Arctic belugas collected between 1993 and 1994 are shown in Table 3.3.15. The average mercury concentration (1.34 $\mu g \cdot g^{-1}$ wet wt.) in muscle of belugas (all years combined) from the western Arctic was more than twice the Health Canada guideline for total Hg in fish muscle (0.5 $\mu g \cdot g^{-1}$ wet wt.). Animals approximately four years and older had mercury concentrations in muscle >0.5 $\mu g \cdot g^{-1}$ wet wt. Total mercury in muscle was positively correlated with the age of animals — a significant finding. The total mercury concentration in muktuk, 0.78 $\mu g \cdot g^{-1}$ wet wt., (1993 and 1994 combined) was lower than in muscle, but still exceeded 0.5 $\mu g \cdot g^{-1}$ (wet wt.) in 61% of the animals. Animals 16 years and older appeared to have mercury concentrations in muktuk >0.5 $\mu g \cdot g^{-1}$ wet wt. Muktuk was not collected prior to 1993, neither in the western nor eastern Arctic. The total mercury concentration was approximately 18 times higher in the liver and nearly four times higher in the kidney than in the muscle. The concentrations, in muscle, liver and kidney, were strongly, positively correlated with the age of the animals.

Approximately, 100% of the total mercury in muscle was methylmercury (1.32 $\mu g \cdot g^{-1}$ wet wt.). Of the animals sampled, 92% had a methylmercury concentration in muscle >0.5 $\mu g \cdot g^{-1}$ wet wt. In muktuk the methylmercury concentration was lower (0.69 $\mu g \cdot g^{-1}$ wet wt.), and fewer samples exceeded the 0.5 $\mu g \cdot g^{-1}$ level. The methylmercury concentration in liver (1.9 $\mu g \cdot g^{-1}$ wet wt.) was approximately 1.4 times higher than in muscle. On average, 6% to 12% of the total mercury in the liver was methylmercury.

The zinc concentration was approximately three times higher and selenium nine times higher in muktuk than in muscle, (Table 3.3.15) making muktuk a good source for consumers of these two essential elements. (See Chapter 4, Section 4.2.5.3).

Individual strata (4) of the skin from 27 western arctic belugas, were analysed for mercury which increased progressively in each stratum in the outward direction (0.29 $\mu g \cdot g^{-1}$ wet wt., in the dermis, and 1.5 $\mu g \cdot g^{-1}$ wet wt., in the *stratum externum*). The thickness and density of each skin stratum was measured, the surface area of an average-size beluga was estimated, and the excretion rate of mercury through molting calculated (14 100 $\mu g \cdot year^{-1}$). Approximately 20% of the total mercury in the skin was estimated to be lost annually through molting.

Similar to results for ringed seals, the spatial trend for cadmium (Figure 3.3.22D) in beluga liver was opposite to that for mercury (Figure 3.3.22C), i.e. higher concentration and rate of accumulation in the eastern Arctic than the western Arctic (see Section 3.4.1.2 of this chapter).

Metals in narwhal

Results for a large sample of narwhal tissues from Pond Inlet (63 animals) in 1979 were reported by Wagemann *et al.* (1983). Data from this sample have not been subsequently verified with an equally large sample. However, the small sample collections in 1992–94 (Table 3.3.15) tend to confirm earlier results (Wagemann *et al.* 1983) indicating that narwhal from Pond Inlet had high cadmium concentrations in liver and kidney. In the 1992 animals (Pond Inlet, 9 animals) the cadmium concentration in both organs were even higher than observed by Wagemann *et al.* (1983) averaging 47.1 $\mu g \cdot g^{-1}$ wet wt. in the liver, 75.8 $\mu g \cdot g^{-1}$ wet wt. in kidney. A larger sample size should be analysed to confirm these high levels.

The mean total mercury concentration in muscle of narwhal from Pond Inlet (1.27 $\mu g \cdot g^{-1}$ wet wt.), exceeded considerably the guideline for fish (0.50 $\mu g \cdot g^{-1}$ wet wt.). In liver, the mean concentration of total mercury in narwhal from Pond Inlet was 10.8 $\mu g \cdot g^{-1}$ wet wt. These results were derived from 55 animals. In muktuk, the mean total mercury concentration ranged from 0.61 $\mu g \cdot g^{-1}$ wet wt. (Iqaluit) to 0.34 $\mu g \cdot g^{-1}$ wet wt. (Repulse Bay) and probably reflects age differences between the sample populations. As in beluga, mercury was not homogeneously distributed throughout the muktuk. The outermost strata (degenerative epidermis and stratum externum) had the highest concentration. The mean methylmercury concentration in muscle of narwhal (three sites combined) was 0.81 $\mu g \cdot g^{-1}$ wet wt., significantly exceeding the 0.50 $\mu g \cdot g^{-1}$ wet wt. guideline, and representing 90% to 100% of the total mercury concentration in this tissue. Methylmercury in liver of narwhal was 1.58 $\mu g \cdot g^{-1}$ (wet wt.) (Repulse Bay), and 3.02 $\mu g \cdot g^{-1}$ (Iqaluit), representing 37% to 51 % of the total mercury in this tissue, but these mean percentages depend to some extent on the mode of calculation (Wagemann *et al.* 1996a). The age of narwhal could not be determined in any of the investigations which limited interpretation of the data. Methylmercury was not determined in muktuk.

The concentration of lead in liver (0.004 $\mu g \cdot g^{-1}$ wet wt.) and muscle (0.004 $\mu g \cdot g^{-1}$ wet wt.) in the narwhal sampled in 1992 was very low, as in the 1979 animals (Wagemann *et al.* 1983). Hard tissues (where lead accumulates preferentially) were not analysed for lead.

Metals in walrus

Heavy metals and selenium data for walrus harvested at Arviat in 1982 to 1988 (113 animals), at Hall Beach in 1989 (17 animals), at Akulivik in 1990 (4 animals) and at Inukjuak in 1990 (9 animals), and data for clams (*Mya truncata*) and cockles (*Serripes groenlandicus*) collected at Igloolik have been published by Wagemann and Stewart (1994). Liver, kidney and muscle from walrus and various tissues of clams and cockles were analysed. In clams, lead was high, up to 3 $\mu g \cdot g^{-1}$ (wet wt.). Lead was also high in walrus tissues and showed a geographic trend of increasing concentration from north to south (0.07 to 0.17 $\mu g \cdot g^{-1}$ wet wt., in liver; 0.012 to 0.024 $\mu g \cdot g^{-1}$ wet wt., in muscle). The mercury concentration in all walrus tissues was low compared with that in belugas and ringed seals, and in liver showed a geographic trend similar to that for lead. Cadmium in liver and in kidney of walrus was high compared with that in belugas and ringed seals, especially at the most northern sampling sites. The mean metal concentrations ($\mu g \cdot g^{-1}$ dry wt.) varied with sampling site. Overall concentrations (all years and locations combined) are shown in Table 3.3.15.

Walrus tissues were not analysed for methylmercury. Associations among metals and between metals and age were found in walrus as in other marine mammals, notably between mercury and selenium, and mercury and cadmium in liver, cadmium and zinc in liver and kidney, and mercury and cadmium with age in kidney.

Spatial trends of organochlorines and metals in polar bears

The polar bear is an excellent candidate biomonitor for the Arctic marine environment. Polar bears are the principal mammalian predators at the top of the Arctic marine food chain, and are distributed widely throughout the Arctic and subarctic circumpolar regions. Their diet consists mainly of ringed seal with variable percentages of bearded seal (*Erignathus barbatus*) in some areas (Smith 1980; Stirling and Archibald 1977). Ringed seals do not move far (Smith and Hammill 1980) and therefore integrate OC contamination in their diet over a limited area. Polar bears are distributed into relatively discrete populations (Taylor and Lee 1995, Bethke *et al.* 1995) although individuals can wander over several thousand kilometres in a year (Ramsay and Stirling 1986, Messier *et al.* 1992). The polar bear is at trophic level 5 compared to particulate organic matter in seawater (Hobson and Welch 1992). Biomagnification factors of OCs are in the order of 3 to 7 per trophic level on a lipid weight basis (Norstrom 1994), therefore biomagnification factors from particulate matter to polar bear lipid can be as high as $7^5 \approx 17\,000$.

Polar bears are philopatric and there is little exchange among populations, although the winter ranges may overlap significantly in some areas such as Hudson Bay (Derocher and Stirling 1990). The average marine area over which a polar bear population integrates contaminants is variable, ranging from 150 000–350 000 km^2 in the Bering and Chukchi Seas (Garner *et al.* 1995) to less than 25 000 km^2 in the Canadian Arctic archipelago (Schweinsburg *et al.* 1982). The polar bear is therefore a meso-scale indicator of Arctic and subarctic marine pollution.

PCBs and *p,p'*-DDE were first reported in polar bear tissues in the early 1970s by Bowes and Jonkel (1975), a few years after PCBs were first identified as environmental contaminants (Jensen *et al.* 1969). Between 1982 and 1984, a systematic survey of the geographical distribution of OCs in polar bear fat and liver throughout the NWT was undertaken. Relatively high levels of PCBs and the pesticide metabolite oxychlordane were found in polar bears, along with lesser amounts of chlordane-related compounds, *p,p'*-DDE, dieldrin, HCB and α–HCH (Norstrom *et al.* 1988, Muir *et al.* 1988b). Levels of OCs tended to be highest in the southeast range of sample collection (Hudson Bay), and lowest in the northwest (Beaufort Sea).

Study design

Samples were collected from 586 polar bears by necropsy or biopsy between the spring of 1989 to the spring of 1993 from 18 regions in the western hemisphere from Wrangel Island in the East Siberian Sea, westward through the range of the bear in Alaska, Canada and eastern Greenland to Svalbard in the east, a span of 200° of longitude encompassing the Western Hemisphere Arctic. Sampling locations in the Canadian Arctic are described in Table 3.3.16. The regions in Canada were chosen to follow the latest Management Zones in Canada (Taylor and Lee 1995). In some cases the Zones were subdivided. The Wrangel Island (R1) and Chukchi/Bering sea (R2) groups (Figure 3.3.23) are considered to be the same population, but were separated because of the different sex and season characteristics of the samples. These samples were analysed for concentration of 35 individual OCs using the method of Zhu *et al.* (1995). The results for the other OCs were summed according to membership in three OC classes: 19 polychlorinated biphenyl congeners (ΣPCB), 12 chlordane-related compounds and metabolites (ΣCHL), and *p,p'*-DDE. More volatile OCs, chlorobenzenes (ΣCBz) and hexachlorocyclohexanes (ΣHCH) were analysed separately. Samples were also analysed for methyl sulfone metabolites of PCBs ($\Sigma MeSO_2$-PCB). Analysis of covariance was carried out with classification factors for three sex categories (males, females with cubs and females) and 18 regions, age as a covariate, and interaction of age and sex with Region. Further details on the design of spatial trend and results for ΣPCB, ΣCHL, *p,p'*-DDE and dieldrin are presented in Norstrom *et al.* (1996).

Spatial trends in organochlorines

A significant negative effect of age was found on levels of ΣPCB and ΣCHL in subadults (<5 yrs), therefore comparison of OC levels among regions was restricted to the 345 adult samples. The sample characteristics and geometric mean concentrations of OCs in adipose tissue (lipid basis) for Canadian Arctic locations are given in Table 3.3.16. Levels of

TABLE 3.3.16

Concentrations of organochlorines (geometric means, ng·g^{-1}, lipid wt.) in adult polar bear fat from the Canadian Arctic. Data are from Norstrom *et al.* (1996).

	Location[1]	Sample size[2] (ng·g^{-1}, lipid wt.)				Concentration			
		M	F	FC	Total	ΣPCB	ΣCHL	*p,p'*-DDE	Dieldrin
R3	McLure Strait + adjacent Arctic Ocean	11	14	–	25	20 256	2 956	285	145
R4	Amundsen Gulf and Beaufort Sea to 135°W	3	8	1	12	5 191	1 453	73	85
R5	Viscount Melville Sound west of 100°W	9	12	–	21	8 632	1 952	112	96
R6	Queen Maud Gulf and Larsen Sound	4	5	4	13	4 566	2 141	52	147
R7	Barrow Strait and Cornwallis Island	8	2	–	10	4 280	1 766	113	157
R8	Gulf of Boothia	5	4	1	10	3 062	1 500	72	142
R9	Baffin Bay north of 72°N, Lancaster Sound, Jones Sound, Thule and Ellesmere Island	9	6	3	18	5 985	1 987	184	185
R10	Southern Baffin Bay and Northern Davis Strait	4	1	–	5	6 819	4 055	386	
R11	Foxe Basin and Hudson Strait west of 72.5°W	18	8	3	29	5 565	2 073	215	233
R12	Western Hudson Bay (Cape Churchill)	33		–	33	5 942	1 689	238	159
R13	Eastern Hudson Bay (Belcher Island)	9	3	–	12	10 873	4 632	560	335
R14	Davis Strait (below Arctic circle) and Hudson Strait east of 72.5°W	3	4	2	9	7 049	2 074	219	190

[1] *Only regions within Canada are listed. All regions, R1 to R16 are shown in Figure 3.3.23.*

[2] *M=Males, F=Females, FC=Females with cubs. All samples obtained in 1990 or 1991.*

ΣPCB were 46% higher in males than females and there was no trend with age. Levels of ΣCHL were 30% lower in males than females and there was a negative trend with age in both sexes ($\log_e$ΣCHL = –0.021 age). There was a sex effect on *p,p'*-DDE levels in females with cubs, but no consistent age effect. Concentrations of ΣPCB, ΣCHL and *p,p'*-DDE in individual adult female bears therefore were standardized to adult males using factors derived from the least-square means of each sex category.

The sex-standardized geometric mean levels of ΣPCB and ΣCHL in polar bear fat for 16 circumpolar regions are plotted as bars in Figure 3.3.23. In this figure, the ΣCHL results have also been age-standardized to the median age of 11 using an analysis of covariance model intercept for each region, and a slope with age of –0.021. One of the most important findings was the relatively uniform distribution of OC levels over much of the study area. This is a clear indication of extensive transport and deposition of OCs to all areas of the Arctic and subarctic. Median (range) levels for the whole data set were as follows: ΣPCB, 7119 (1228–70 421) ng·g^{-1}, ΣCHL, 1988 (207–15 013) ng·g^{-1}, *p,p'*-DDE, 190 (24–2821) ng·g^{-1}, DIEL, 149 (7–835) ng·g^{-1}.

Geometric means of the standardized data on a lipid weight basis were compared among Regions. The geographical distribution of ΣPCB is notable for the peaks occurring in R1, R3, R15 and R16 (Table 3.3.16, Fig. 3.3.23). The higher ΣPCB levels in R1 may have been because this group was composed almost entirely of females with cubs just emerging from dens. Polischuk (1995) has found that ΣPCB levels in fat of females emerging from dens after six months fasting are two times higher than in the previous autumn. ΣPCB levels were similar (20 256–24 316 ng·g^{-1}) in bears from Svalbard (R16), East Greenland (R15) and the Arctic Ocean near Prince Patrick Island in Canada (R3), and significantly higher than most other areas. There were few other significant differences in ΣPCB levels apart from some between R5 and R13 and the regions with the lowest mean levels: R2, R6, R7 and R8. There were no significant trends in geographical disposition of ΣPCB within the band of Regions across the North American coast and the Canadian Archipelago which had the lowest levels of ΣPCB (R2, R4, R6–R12, R14, Figure 3.3.23). Nevertheless, average levels in this band were higher in Baffin Bay and Hudson Bay (6270 ± 631 ng·g^{-1}, R9–12, R14), than in the western regions (3970 ± 1030 ng·g^{-1} R2, R4, R6–8), indicating a slightly increasing west-east gradient.

Significant differences in ΣCHL occurred mainly between southern Hudson Bay (R13, highest) and Wrangel Island, East Siberian Sea (R1, lowest) and other regions (Table 3.3.16, Figure 3.3.23). Levels were about 50% higher than average in the three Regions which had highest ΣPCB concentrations, but the highest level was found in southeastern Hudson Bay (R13), which also had the highest *p,p'*-DDE and dieldrin levels among regions. ΣCHL was the most uniformly distributed of the OCs in Table 3.3.16. This result is in accord with the finding of lower geographical variation of ΣCHL in air and seawater in the Northern Hemisphere than in tropical areas (Iwata *et al.* 1993). The lower ΣCHL levels in polar

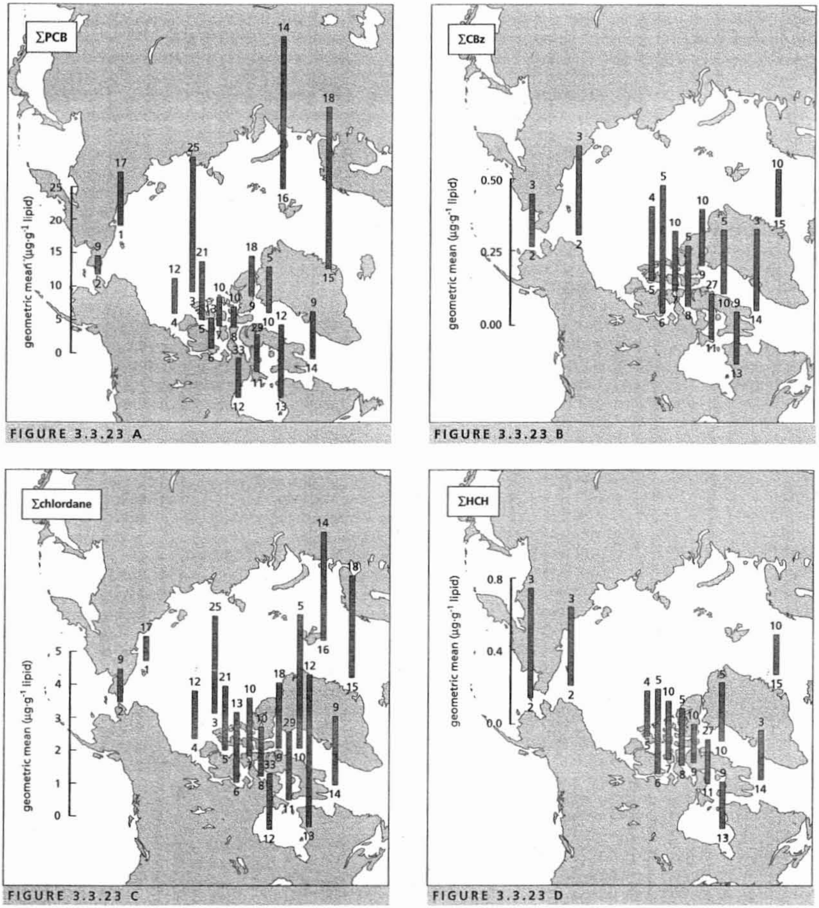


FIGURE 3.3.23 A

FIGURE 3.3.23 B

FIGURE 3.3.23 C

FIGURE 3.3.23 D

Circumpolar view of spatial trends of organochlorines in polar bear fat from 16 regions (Norstrom et al. 1996). Numbers above each bar represent number of samples (N) at each site and numbers below are the region (see Table 3.3.16). Results for ΣPCB and ΣChlordane are based on larger numbers of individual samples than those for ΣCBz and ΣHCH.

bears from R1–R2 indicate that ΣCHL loading is less in the Chukchi and Bering Seas than in the rest of the Arctic.

DDE levels in R2, R4, R6 and R8 were significantly lower than most other regions (Table 3.3.16). DDE levels were higher in the eastern regions, with peaks in R1 and R3 in the west and R13 in the east. As for ΣPCB, the high *p,p'*-DDE in R1 may be because this group of samples was biased towards fasting females. The high *p,p'*-DDE in R10 is probably spurious, because it is driven by one bear out of the five samples from this area. Dieldrin levels displayed a more pronounced west-east increasing gradient than the other OCs (Table 3.3.16). Levels in R1 and R2 were significantly lower than most other regions. The dieldrin distribution strongly suggests an influence of North American sources. This is probably an indication of atmospheric transport to this area after volatilization from soil and plant surfaces in North America during the summer. Eastern Hudson Bay is the only area in the study that is in the direct path of average continental airflow in the summer (Barrie *et al.* 1992). Although atmospheric circulation in Regions further north is less well connected with mid-latitude North American air, it is possible that occasional incursions add to background levels in the eastern Canadian Arctic, explaining the generally higher concentrations of ΣCHL, *p,p'*-DDE and dieldrin in polar bear from these areas compared with those further west, although ecological factors cannot be ruled out. Barrie *et al.* (1992) observed that the patchiness of detection of ΣDDT in air may be an indication of a more episodic incursion of ΣDDT in the Arctic environment.

Geographical distribution of ΣHCH and ΣCBz in composite male adipose tissue samples is shown in Figure 3.3.23. ΣCBz levels were very uniform across the whole study area, with the exception of a peak in R6. This result suggests that ΣCBz is well equilibrated in surface waters throughout the Arctic. Levels of α-HCH were also similar among areas, but unlike all other OCs, the highest levels were found in the western regions, especially the Bering and Chukchi Seas (R2). This is a clear indication of ongoing contributions from Asia and Southeast Asia (Barrie *et al.* 1992). The relatively high level of both ΣHCH and ΣCBz in R3 compared with nearby regions was also noted in 1984 (Norstrom *et al.* 1988).

Methyl sulfone-PCB and -DDE metabolites were found in polar bear fat at levels similar to those of ΣHCH, ΣCBz, *p,p'*-DDE and dieldrin (Bergman *et al.* 1994). Letcher *et al.* (1994) determined $\Sigma MeSO_2$-PCB levels PCBs in the same composite polar bear fat samples analysed for ΣHCH and ΣCBz. They found the geographical distribution to be very similar to that of ΣPCBs. Concentrations of $\Sigma MeSO_2$-PCB were 4% to 8% of ΣPCB. The ratio of $\Sigma MeSO_2$-PCB to ΣPCB decreased from west to east, due to an increase in the proportion of higher chlorinated PCB congeners. Most of the PCBs that form methyl sulfone metabolites are tetra- to hexachloro-substituted.

In addition to the OC residues listed above, several others have been identified in polar bear tissues but there is much more limited information on their geographical distribution and none on temporal trends. These OCs include PCDDs and PCDFs (Norstrom *et al.* 1990); tris (*p*-chlorophenyl) methanol (Jarman *et al.* 1992), photoheptachlor (Zhu *et al.* 1994) and toxaphene (Zhu and Norstrom 1994).

Spatial trends of metals in polar bears

A detailed survey of metals in polar bear livers was conducted between 1982 and 1984 using samples from the 12 polar bear management zones in the Canadian Arctic (Norstrom *et al.* 1986a, Braune *et al.* 1991). As a top predator, with a wide and well-characterized distribution, the polar bear is a excellent candidate for assessing spatial trends on metals in the Arctic marine food chain. Other studies have examined metals in polar bear hair (Eaton and Farant 1982, Renzoni and Norstrom 1990) but the work conducted during the 1980s is currently the most recent on metal contaminants in polar bear tissues.

A total of 124 samples (60 from the western and central Arctic and 58 from the eastern Arctic and Hudson Bay) were analysed for 24 metals. Details of the sampling and analytical procedures employed for determination of metals in polar bear liver are given in Norstrom *et al.* (1986a). Statistical comparisons were made between sampling areas after adjusting for age- and sex-related variation, where required, by analysis of covariance. In the western and central Arctic silver, arsenic, calcium, copper, iron, potassium, magnesium, manganese, sodium, phosphorus, strontium and zinc showed no significant spatial variation (five areas). However, when combined with results from the eastern Arctic most metals showed some variation with sampling region. Only cadmium, mercury and selenium increased significantly with age. Iron was the only element to show significant variation with sex of the animal; it was lower in female livers. Figure 3.3.24 shows age and sex adjusted mean concentrations of cadmium and mercury in polar bear liver from each management zone. Accumulation rates of cadmium in bears from the western Arctic were nearly three times lower than those found in the eastern Arctic. The opposite was found for mercury and selenium: accumulation rates were 7 and 10 times greater, respectively, in bears from the western Arctic than in those from Hudson Bay. Mercury and selenium were highly correlated in polar bear liver (r = 0.97). A high partial correlation (with age as covariable) of mercury and selenium was also found indicating that accumulation with age was through a common biochemical mechanism.

High relative levels of various elements in polar bear liver were not strongly associated with mining operations such as those near Nanisivik or on Little Cornwallis Island, or with known mineral deposits. The large differences in accumulation rates for cadmium and mercury observed in polar bears from western and eastern sampling zones, were similar to those found in ringed seals and beluga (see Section 3.3.3.3, Spatial trends in heavy metals in marine mammals, and Figure 3.4.3, Section 4.1.2). This gradient was also found for mercury in polar bear hair samples (Eaton and Farant 1982, Renzoni and Norstrom 1990). Levels of mercury and selenium were highest in samples from areas bordering the Beaufort Sea. High mercury appears to be related to elevated natural levels associated with the sedimentary geology of the western Arctic. Higher accumulation rates of cadmium in the eastern Arctic may also be the result of the mineralogy of this region, since ringed seals, the main dietary item of polar bears, show the same trend. However, differences in the diet of ringed seals such as lower proportions of amphipods in the diets of ringed seals in the western Arctic, may also play a role (Braune *et al.* 1991, Macdonald 1986).

3.3.3.4 Spatial Trends of Organochlorines and Metals in Sea Birds

Surveys of OC levels in eggs and tissues of arctic sea birds conducted in the mid-1970s showed that sea birds breeding in the high Arctic were contaminated with a similar suite of organic contaminants as those breeding in temperate regions (Noble and Elliott 1986). Arctic sea bird species monitored for contaminants since that time were chosen to represent different trophic levels, feeding strategies and overwintering habits.

The northern fulmar (*Fulmaris glacialis*) and black-legged kittiwake (*Rissa tridactyla*) are both offshore surface-feeders, but the fulmar is also a scavenger that includes carrion in its diet. The thick-billed murre (*Uria lomvia*) and black guillemot (*Cepphus grylle*) are both pursuit divers, omnivorous in diet, feeding on fish and amphipods. The murre feeds both coastally and offshore whereas the guillemot feeds in the more shallow waters of the sublittoral zone. The glaucous gull (*Larus hyperboreus*) is an opportunistic surface-feeder, scavenger and predator.

All of the species except for the black guillemot disperse widely after breeding. Large numbers of the northern fulmar, thick-billed murre and glaucous gull overwinter off Newfoundland. The black guillemot is relatively sedentary, overwintering at the edge of the winter pack ice (Godfrey 1986).

Study design

In 1993, eggs of the thick-billed murre, black guillemot, northern fulmar, black-legged kittiwake and glaucous gull were collected from several Arctic sites (Figure 3.3.25). Chicks of all five species and adult birds of all but the glaucous gull were collected from Prince Leopold Island. Additionally, thick-billed murre chicks were collected from Coats and Coburg Islands, thick-billed murre adults were collected from Coats Island,

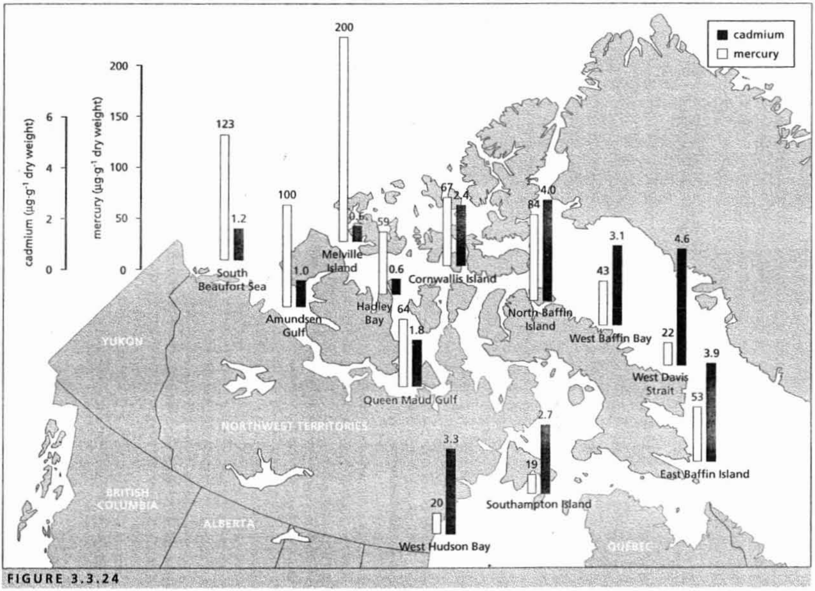


FIGURE 3.3.24

Concentrations (age and sex adjusted) of cadmium and mercury in polar bear liver from 12 polar bear management zones in the Canadian Arctic (Braune et al. *1991).*

and black guillemot chicks and adults were collected from the Nuvuk Islands.

Eggs were collected by hand, one per nest, and the contents analysed on the basis of 5 pools of 3 eggs each totaling 15 eggs per species per location. Exceptions are noted in Table 3.3.17. Ten chicks were collected per species per location except for thick-billed murres from Coburg Island (N=6), black-legged kittiwakes from Prince Leopold Island (N=1), and glaucous gulls from Prince Leopold Island (N=4). Ten adult birds were collected per species per location except for black guillemots from Prince Leopold Island (N=5).

Egg contents, chick carcasses and livers of adults were analysed for OC pesticides, PCB congeners, total mercury, lead, cadmium, and selenium. Further details on collection, storage and analytical methods are given by Braune (1993, 1994b).

Interspecific and spatial trends

Analyses of variance on residue levels found in eggs of sea birds from Prince Leopold Island indicate statistically significant differences among the five species for HCB, ΣChlordanes, *p,p'*-DDE, ΣPCBs, selenium and total mercury. Organochlorine residue levels were four to ten times higher in Glaucous Gulls than in the other species (Figure 3.3.26a). Similar patterns were found in the chicks and the adults. Studies of OCs in sea birds in the Svalbard area have also shown the Glaucous Gull to contain higher residue levels than the other species (Daelemans *et al.* 1992, Norheim and Kjos-Hanssen 1984, Savinova *et al.* 1995).

Total mercury residue levels were also highest in the glaucous gull. This pattern for mercury and OC levels is to be expected since the glaucous gull is highly predatory, often feeding on the eggs and chicks of other birds. Selenium levels were slightly higher in eggs of black-legged kittiwakes and northern fulmars, both surface feeders, but highest in chicks of the glaucous gull, which probably feeds its chicks the eggs and chicks of other birds. Similar inter-specific patterns of mercury and selenium were observed in Greenland sea birds (Nielsen and Dietz 1989).

Lead and cadmium were not detected in any of the eggs collected, and lead was detected in only minimal quantities in the chicks and the livers of adult birds. Cadmium was, however, detected in all of the chicks. Therefore, cadmium and to a lesser extent, lead are being taken up by the chick after hatch (Table 3.3.18) indicating that these contaminants are being picked up from the local arctic food base.

Analyses of variance on residue levels in eggs of thick-billed murres collected from two high Arctic colonies (Prince Leopold Island, Coburg Island) and two low Arctic colonies (Coats Island, Digges Island) show significant differences for HCB ($P<0.001$), ΣPCBs ($P=0.02$) and mercury ($P<0.001$). The eggs from the low Arctic colonies generally contained higher residue levels of OCs than the eggs from the high Arctic sites (Figure 3.3.26b) whereas mercury levels were higher in the eggs from the high Arctic colonies. Consistently higher levels of mercury were also found in birds from northern Greenland compared with southern Greenland (Nielsen and Dietz 1989). Patterns of residue levels in eggs of a resident arctic species, the black guillemot, collected from the same two low Arctic and one of the high Arctic colonies (Prince Leopold Island) sampled for thick-billed murres corroborate the results found for thick-billed murres.

Organochlorine residue levels in eggs of glaucous gulls from the two high Arctic colonies (Prince Leopold Island, Browne Island) were significantly higher ($P<0.001$) than levels in eggs from the two

TABLE 3.3.17

Mean residue levels of organochlorines in eggs of arctic sea birds collected in 1993 ($ng\cdot g^{-1}$ wet wt.).

Species	N[1]	Location	% Lipid	ΣCBz[2]	ΣHCHs[3]	ΣCHL[4]	ΣDDT[5]	Dieldrin	ΣPCBs[6]
Thick-billed Murre	5(3)	Coats Island	14.5	124	22.8	58.2	326	21.2	360
	5(3)	Digges Island	12.5	129	17.8	63.0	311	26.3	434
	5(3)	Coburg Island	13.5	78.1	18.4	39.0	309	14.2	420
	5(3)	Prince Leopold Island	12.2	53.9	19.9	41.9	201	12.5	233
Black Guillemot	4(3)	Nuvuk Island	9.4	106	19.1	141	150	19.5	577
	5(3)	Walrus Island	11.7	91.8	17.5	110	144	12.0	300
	4(3)	Prince Leopold Island	10.1	79.0	24.6	67.6	163	15.4	318
Northern Fulmar	5(3)	Prince Leopold Island	11.9	69.4	4.8	136	490	12.9	563
Black-legged Kittiwake	5(1)	Coburg Island	10.0	45.5	5.2	61.0	124	15.8	602
	5(3)	Prince Leopold Island	8.5	43.1	5.1	24.4	122	7.8	470
Glaucous Gull	5(2)	Browne Island	8.4	143	69.5	295	831	22.7	1262
	5(2)	Prince Leopold Island	8.9	235	65.8	388	2370	27.8	3330
	5(3*)	Anderson River Delta	8.4	61.9	21.7	48.0	144	8.2	316
	5(3)	Coppermine	8.6	52.4	20.2	77.7	339	7.6	462

[1] *N — Number of pools (number of eggs in each pool) -*- one of these pools contained only 2 eggs*

[2] *ΣCBz = Sum of 1,2,3,5 and 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene*

[3] *ΣHCHs = Sum of α-, β- and γ- hexachlorocyclohexanes*

[4] *ΣCHL = Sum of oxy-, trans- and cis-chlordane, trans- and cis-nonachlor and heptachlor epoxide*

[5] *ΣDDTs = Sum of p,p'-DDE, p,p'-DDD and p,p'-DDT*

[6] *ΣPCBs = Sum of 42 congeners: 28, 31, 42, 44, 49, 52, 60, 64, 66/95, 70, 74, 87, 97, 99, 101, 105, 110, 118, 128, 129, 137, 138, 141, 146, 149, 151, 153, 158, 170/190, 171, 172, 174, 180, 182/187, 183, 185, 194, 195, 200, 201, 203, 206*

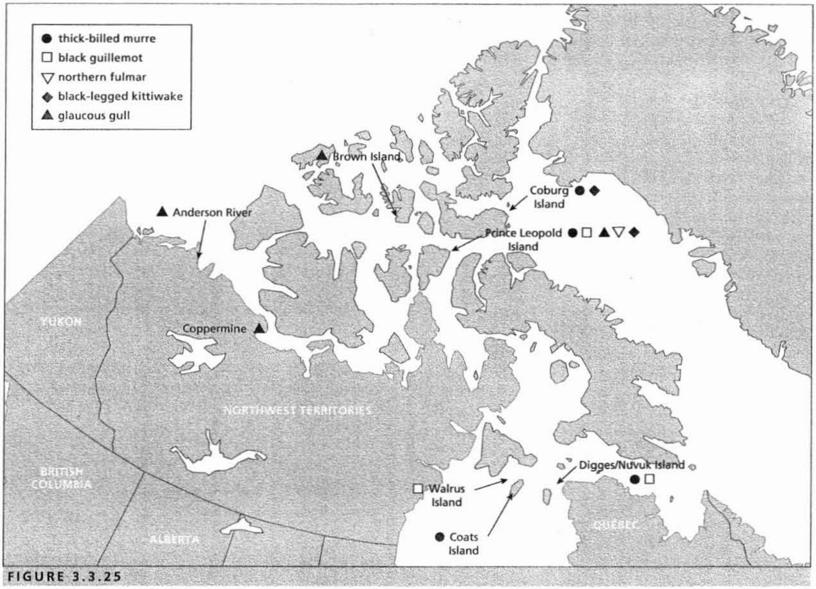


FIGURE 3.3.25

Collection sites of sea bird eggs (1993).

western low Arctic colonies (Anderson River, Kuglutuk — formerly known as Coppermine) (Table 3.3.17). The birds in the western low Arctic feed lower on the food chain (i.e., vegetation, goose eggs and goslings) (Barry and Barry 1990) compared with the more predatory birds on Prince Leopold Island, which feed on eggs and nestlings of other sea birds as well as fish (Nettleship *et al.* 1990).

3.3.3.5 Migratory Marine Species

Although the source of persistent OC contaminants present in biota throughout the Arctic ecosystem is thought to be mainly long-range atmospheric dispersion (Barrie *et al.* 1992) migratory species may bring contaminants with them from more contaminated regions in the south. Migratory birds and harp seals (*Phoca groenlandica*) are the best documented examples of this.

Migratory birds pick up contaminants through the food on their southern wintering grounds or sites along the migration pathway. Birds that breed in the north and overwinter in more temperate, industrialized latitudes may contain higher levels of contaminants in their tissues than those birds that overwinter in the north. The contaminants are taken up from the food on the overwintering grounds and transported north each spring when the birds migrate back to their breeding grounds. This has significant implications for arctic predators, including humans, for which the migrating birds provide a food source. Contaminant levels in common and king eiders (*Somateria mollissima; S. spectabilis*) collected as part of a survey of contaminants in harvested avian species (see Section 3.3.2.3), illustrate this trend as do peregrine falcons (sections 3.4.2.2, 5.3.1).

There are several population groups of eiders throughout Arctic Canada and Hudson Bay, some that overwinter in the north and others which migrate to more temperate latitudes (Figure 3.3.27) (Abraham and Finney 1986, Barry 1986, Reed and Erskine 1986). The Low Arctic and High Arctic eiders migrate to overwinter in waters as far south as Newfoundland and the Gulf of St. Lawrence. The Gulf of St. Lawrence receives the effluent flow from the Great Lakes and St. Lawrence River, waters known to be contaminated (Gagnon *et al.* 1990, Comba *et al.* 1993). The other eider groups, the western arctic birds in particular, overwinter in less contaminated northern waters. This is reflected in the tissue residue concentrations of the birds (Figure 3.3.28).

3.3.3.6 Biomagnification of Organochlorines and Metals in the Arctic Marine Food Web

Organochlorines

Information on likely predator-prey linkages in the Arctic marine food chain (Figure 3.3.18) was combined with data in Table 3.3.12, and other tables in this section, to construct a simplified food web diagram for calculation of biomagnification factors (BMFs) (the ratio of concentrations in predator:prey) for OCs accumulated by organisms from various trophic levels (Figure 3.3.18). High BMF values (>100) were observed by Muir *et al.* (1988b) for heavier PCB congeners (containing six or seven chlorines) and similar values are apparent in the data for ΣPCB summarized here (Figure 3.3.18). The ranges of values for toxaphene, reported in Hargrave *et al.* (1993) using some of the data summarized in Table 3.3.12, are similar in magnitude to those for PCBs with the highest values for links between arctic cod (or char at the same trophic level) and their potential predators (mammals and benthic lysianassid amphipods).

Ranges and maximum BMFs for predator-prey links between lower trophic level organisms (plankton-amphipods) (10–250) and between fish and marine mammals (1–300) are similar for all OC compounds. Apparently, short life span in smaller invertebrates that might limit OC bioaccumulation, as discussed above, does not result in lower BMFs. This effect could account for levels of OCs in relatively short-lived benthic amphipods that exceed those in long-lived fish and mammals. Low BMFs between arctic cod (or char), and their main diet of pelagic amphipods and epontic algae, may be reassessed after analyses of samples that have been collected seasonally in Barrow Strait. It will also be possible to calculate BMFs for specific PCB congeners to examine the selective accumulation known to occur. With data presently available, however, the observed patterns of biomagnification between organisms and for various compounds are sufficient to account for high (µg·g-1) OC levels observed in marine fish and mammals consumed by northern people.

TABLE 3.3.18

Total mean residue burdens (µg) of metals in eggs and chicks of sea birds collected from Prince Leopold Island in 1993.

Species/Sample		N[1]	# Eggs or Chicks/Pool	Mean[2] Wt. (g)	Total Hg	Pb	Cd	Se
Thick-billed Murre	Egg	5	3	75.1	22.7	—	—	51.6
	Chick	1	10	129.0	44.8	—	12.7	55.2
Black Guillemot	Egg	4	3	42.4	25.6	—	—	24.1
	Chick	1	10	226.0	106.0	4.2	29.0	104.0
Northern Fulmar	Egg	5	3	79.7	25.5	—	—	86.9
	Chick	1	10	156.0	80.2	2.6	35.3	140.0
Black-legged	Egg	5	3	37.8	5.3	—	—	37.6
Kittiwake	Chick	1	1	102.0	16.5	8.7	9.5	48.2
Glaucous Gull	Egg	5	2	98.5	113.0	—	—	63.3
	Chick	1	4	838.0	1180.0	—	18.4	880.0

[1] *Number of pools*

[2] *Mean egg weight refers to egg contents. Mean chick weight includes carcass and feathers*

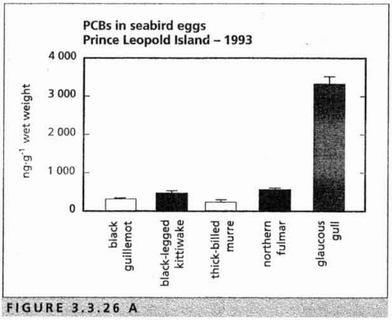


FIGURE 3.3.26 A

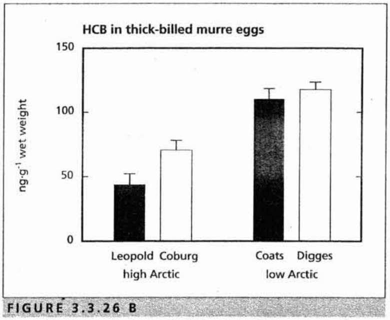
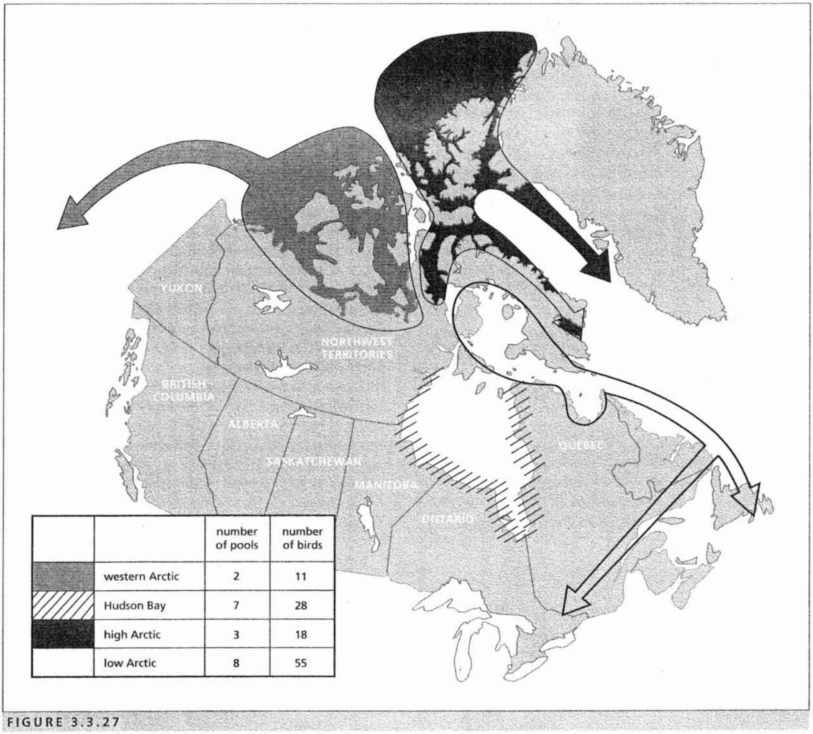


FIGURE 3.3.26 B

Mean concentrations (± S.E.)(ng·g⁻¹ wet wt.) in sea bird eggs. (A) ΣPCBs in eggs from Prince Leopold Island. (B) HCB in thick-billed murre eggs from four Arctic colonies.



		number of pools	number of birds
▒	western Arctic	2	11
▨	Hudson Bay	7	28
■	high Arctic	3	18
□	low Arctic	8	55

FIGURE 3.3.27

Migration pathways and number of samples collected of eiders in the Canadian Arctic.

The biomagnification of persistent OCs such as CB153 (2,4,5,2',4',5'-hexachlorobiphenyl) can also be viewed from a trophic level perspective (Figure 3.3.29) using the same sources of information used in Figure 3.3.18. Trophic level data from Hobson and Welch (1992) were used to assign levels for each species/genus. Trophic level is strongly correlated with log CB153 (lipid weight) concentrations illustrating the biomagnification up the food chain of this PCB congener which is essentially non-biodegradable in the marine food web.

Metals

Less information is available on metals in lower marine food web organisms with which to evaluate biomagnification. Using estimated concentrations of heavy metals in sea water which were made during the 1980s (from the review by Muir *et al.* 1992a) BMFs for heavy metals from seawater to narwhal liver were 4 x 10^6 for cadmium, 3 x 10^5 for mercury and 1.9 x 10^3 for lead, for eastern Arctic marine food chains. By comparison the BMFs for cadmium and mercury from water to hepatic tissues of antarctic minke whales (*Balaenoptera acutorostrata*) were 5.5 x 10^5 and 4.3 x 10^4, respectively (Honda *et al.* 1987). BMFs for cadmium and mercury in the Arctic marine food web are shown in Figure 3.3.18. These are the ratio of concentrations in predator or higher trophic organism to prey calculated using dry weight ÷ dry weight, or wet wt. ÷ wet wt. concentrations with whole organism (invertebrates and fish, or muscle concentrations in marine mammals). Where information from the Canadian Arctic was not available, for example, with mercury in some invertebrates, data from Greenland (Dietz *et al.* 1995) was used. The BMFs for cadmium from seals to polar bears is <1 if based on the ratios of concentrations in the liver of each species. The bears consume mainly the skin and blubber of seals, which have low levels of cadmium and mercury (Johansen *et al.* 1980). BMFs for seals to bears of 77 for cadmium and 3000 for mercury can be calculated assuming levels of 0.02 $\mu g \cdot g^{-1}$ for both cadmium and mercury, which are typical of levels of these metals in blubber of eastern Arctic ringed seals (Wagemann 1995). These high BMFs are more typical of the transfer of cadmium and mercury from prey to predator.

3.3.3.7 Overall Assessment of Contaminants in Marine Mammals and Sea Birds

A large amount of information is presently available on OCs and metals in marine biota from the Canadian Arctic. Most of this information is for marine mammals (including polar bears) and sea birds. Geographic coverage of marine mammal and sea bird populations is very good. All major beluga, ringed seal and polar bear stocks along with several major sea bird colonies have been sampled for OC and heavy metal contaminants. Studies on contaminants in walrus are limited to Foxe Basin and northern Québec stocks, while migratory harp seals have only been studied recently at one location. Contaminant measurements in bearded seal, harbour seal, bowhead whale and killer whale tissues from the Canadian Arctic are very limited or non-existent.

There is much less information on contaminant levels and very limited geographic coverage for marine fish and invertebrates. Contaminant data from the marine food web is important because, combined with information of trophic levels of each organism, it leads to an understanding of the pathways of bioaccumulation of the contaminants. The marine food web data is also important because some animals, such as mussels and amphipods, may be harvested by Northerners or may be fished commercially (turbot, prawns). Most of the information for OCs in lower food chain organisms comes from work at the Ice Island (near Alex Heiberg) and in Lancaster Sound at Resolute in the central archipelago. In addition, the studies of the PCB contamination near DEW line sites have contributed

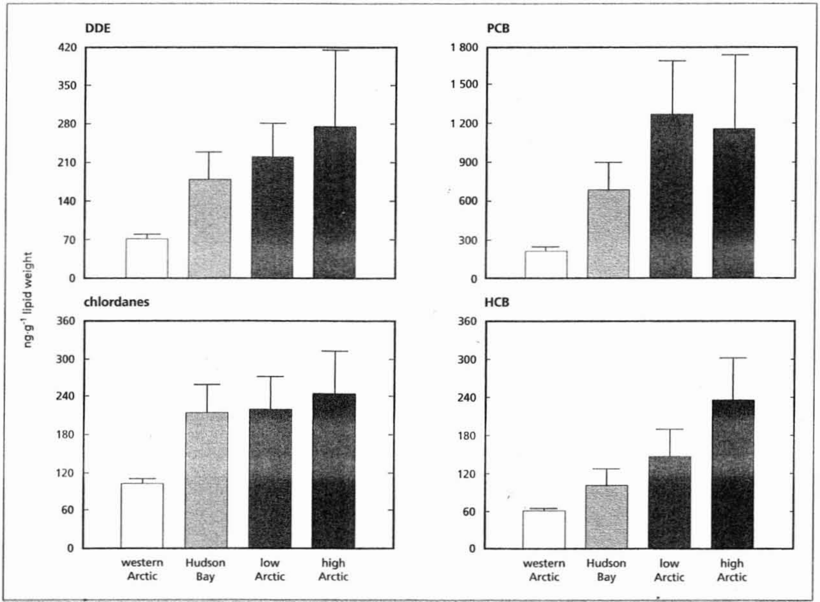


FIGURE 3.3.28

Mean concentrations (± S.E.) (ng·g⁻¹, lipid wt.) of DDE, PCB, chlordane and HCB in common and king eiders from four locations in the Canadian Arctic.

to the data available on levels in nearshore benthic organisms such as sculpins and bivalves. Much of the data on metals in invertebrates comes from studies in northern Baffin Island and Lancaster Sound carried out during the 1970s and 1980s prior to and following development of mines in the region. There is virtually no contaminant information on invertebrates and marine fish from Hudson Bay, Hudson Strait or areas south and west of Lancaster Sound in the central Arctic archipelago.

There is evidence of PCB contamination at some locations in the Canadian Arctic due to pollution from military radar facilities and other activity. The detailed study of PCBs in nearshore areas of Cambridge Bay illustrates how PCBs can be transported from dump sites to the immediate marine environment. But these studies also show that the PCB contamination is quite localized when considered on a broad regional scale. There is no evidence that marine mammals frequenting the marine waters within the general area of these sites have elevated PCB contamination resulting from these sites. In fact, PCB concentrations in blubber of marine mammals and fat of polar bears are remarkably similar across the Canadian Arctic. However, it should be noted that no specific studies were carried out to examine marine mammals near these sites. The work near and at Cambridge Bay also showed that PCBs were elevated in nearshore areas close to riverine inputs in pristine areas (Wellington Bay) which may indicate that runoff of snow-derived PCBs and/or food web differences in these areas contributes to the regional variability of contaminant levels.

The major OC contaminants in marine biota were toxaphene and PCBs. DDT and chlordane related contaminants are also important especially in polar bears and seals. Toxaphene predominates in lower food web organisms and does not biomagnify to the same extent as PCBs or some chlordane components in polar bear. Toxaphene concentrations were particularly elevated in turbot muscle (300–400 $ng \cdot g^{-1}$ wet wt.). The turbot, or Greenland halibut, is a predacious desmersal fish, which, along with other fish such as redfish (*Sebastes marinus*), is a significant dietary item of beluga and narwhal in the Baffin Bay, Davis Strait area. The presence of these species may explain why toxaphene and other OCs are slightly higher in beluga and narwhal from Cumberland Sound and Greenland compared with stocks in the western Arctic. Slightly higher concentration of PCBs are observed in ringed seals from Hudson Bay compared with populations in the western or central Arctic. Concentrations of PCBs in polar bear fat parallel this spatial trend. These west-to-east spatial trends are

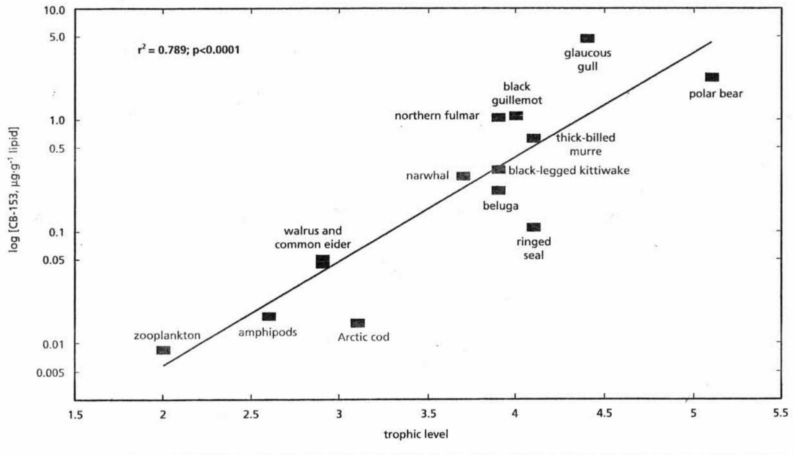


FIGURE 3.3.29

Correlation between logarithm of CB153 concentrations in marine biota (lipid weight basis) and trophic level in the arctic marine food chain. Trophic level data are from Hobson and Welch (1992). The trophic level of walrus was assumed to be the same as for common eider because both are primarily molluscivores. The trophic level of arctic cod was assumed to be an average of large and small fish. CB 153 data for invertebrates, fish and marine mammals are from Hargrave et al. *(1992), Muir* et al. *(1988b) and Norstrom and Muir (1994), respectively. Data for common sea birds are from Braune (1994b).*

difficult to identify within the Canadian Arctic but become obvious when examining the circumpolar data. In both ringed seals and polar bears (Figure 3.3.23) higher concentrations of PCBs and other OCs are seen in animals from east Greenland Svalbard and northern Norway. Pfirman *et al.* (1995) suggest that release of particulates by melting in the marginal ice areas in the Greenland and Barents seas may be an important mechanism of focusing contaminants from a wide area of the Arctic into these areas. The existence of similar areas in Baffin Bay/Davis strait and M'Clure Strait might be an additional explanation for elevation of OC contaminants in cetaceans from this area.

While concentrations of most OCs are quite similar spatially in most species in the Canadian Arctic an exception is the polar bears from Region 3 (M'Clure Strait). This population showed elevated levels in samples collected in 1990-91 and between 1982 and 1984. These bears may be influenced by a different food chain structure in a permanent ice environment, or by some as yet unknown feature of atmospheric or oceanic transport. There is anecdotal information that bearded seals may be more abundant in this area, and there is the possibility that the base of the ringed seal food chain is more epontic than pelagic compared with areas further east (Welch *et al.* 1992). Polar bears from M'Clure Strait also had exceptionally high levels of mercury in liver compared with polar bears in nearby regions. Unfortunately there is no seal or lower food web contaminant data from this area for corroboration.

Other exceptions to the thesis of lack of spatial trends of OCs in marine biota are in sea birds and walrus. Glaucous gulls from Prince Leopold Island and Brown Island in the central Arctic had much higher OC levels (by three to ten times) than populations from the western Arctic. This was not the case for thick-billed murre and black guillemot, however, these species were available only in the eastern/central Arctic and Hudson Bay. Different feeding habits on the breeding grounds as well as winter feeding areas for the eastern and western glaucous gulls are the possible explanation for these differences.

The remarkable variation of OCs in walrus provides an example of the influence of diet on contaminant levels in top predators. About 35% of the 53 samples analysed had elevated concentrations of ΣPCBs. Local contamination was ruled out because levels of all OCs were elevated in each animal from Inukjuak, and elevated levels were also found in animals from other areas of the eastern Canadian Arctic such as at Loks Land at the tip of Frobisher Bay. Predation by walrus on seals is well known but the fraction of walruses at Inukjuak and Loks Land that appear to prey on seals (85% and 66%, respectively, based on the elevated PCB levels) is higher than is reported in the literature. Lowry and Fay (1984) report that from the 1940s to the 1980s, the percent of walruses harvested in the Bering and Chukchi Seas that contained seal tissue in their stomachs, ranged from 0 to 11.4%. Parts of the seals that were eaten were mainly skin and blubber. This may explain why mercury and cadmium were not elevated in the same animals (Wagemann and Stewart 1994).

Unlike the organochlorines, contamination of marine biota by heavy metals shows consistent large regional differences. Studies of polar bears and of ringed seals and beluga all indicate higher mercury contamination of these species in the western Arctic. In polar bear, levels of mercury and selenium were highest in samples from areas bordering the Beaufort Sea. High mercury appears to be related to elevated natural levels associated with the sedimentary geology of the western Arctic. The spatial trend for cadmium was opposite to that for mercury — that is, higher in the eastern Arctic than in the western Arctic for belugas and the rate of accumulation of cadmium in the liver was higher in the eastern Arctic.

Higher accumulation rates of cadmium in the eastern Arctic may also be the result of the mineralogy of the eastern Arctic. Differences in the diet of ringed seals, such as lower proportions of amphipods in the diet of ringed seals in the western Arctic, may also play a role. But with three species that have different dietary preferences showing the same trend, the explanation that differences are due to the different geology of the eastern and western Arctic seems stronger. These regional differences have not been confirmed with actual measurements in lower food web marine organisms, sea water or sediments.

In general, levels of cadmium in kidney and liver of Canadian Arctic marine mammals are as high or higher than levels observed in the same or similar species in more temperate waters, but are similar to concentrations found in some terrestrial animals such as caribou and moose. This was first noted in the early 1980s in the case of narwhal (Wagemann *et al.* 1983) and has been confirmed as additional species have been examined in the Canadian Arctic and in Greenland (Hansen et al 1990, Dietz *et al.* 1995). Cadmium concentrations are also elevated — i.e. $>0.5\ \mu g \cdot g^{-1}$ — in muscle of beluga and ringed seals. The high cadmium appears to be the result of accumulation from natural sources. Although cadmium concentrations are not elevated in sea bird eggs and chicks due to lack of maternal transfer, levels in adult sea birds are comparable to levels found in marine mammals. Cadmium concentrations are remarkably variable in lower food chain organisms (Macdonald and Sprague 1988). Arctic cod have very low cadmium whilst the carnivorous amphipod *Parathemisto libellula* and phytoplankton have elevated levels. Although concentrations of cadmium in arctic cod liver are low, they are about 10 times higher than in cod liver from the northern

Baltic Sea. Cadmium levels in amphipods were similar to those found in hyperiid amphipods in the north Pacific Ocean.

In the case of lead and mercury, higher levels are found in beluga from more contaminated environments. Belugas from the St. Lawrence estuary had higher lead and mercury concentrations in kidney and liver tissues than beluga from five Arctic locations (Wagemann *et al.* 1990). This is consistent with the greater industrial contamination of the St. Lawrence estuary that is also observed in the case of PCBs in beluga.

The relatively high levels of mercury and methylmercury in skin or muktuk of beluga is a new observation. Previous measurements of metals in skin of Arctic marine mammals were limited to samples from harbour porpoise (*Phocena phocena*) (Dietz *et al.* 1995). An estimated 20% of the total mercury in the skin is lost annually through molting, thus a major route of elimination of mercury by cetaceans has been identified.

With the completion of work on beluga muktuk, information is now available for levels of metals for most marine mammal tissues, which may be used in traditional native diets. Additional studies of metal contaminants in muktuk need to be made in eastern Canadian marine mammal populations. Information is more limited on OC levels in muktuk and other tissues of marine mammals. Levels clearly vary with fat content of muktuk, thus OC concentrations in blubber may be used to estimate concentrations in muktuk with good accuracy. Whether OC levels in other marine mammal tissues can be estimated from blubber levels has not been thoroughly investigated. Additional measurements of OCs in muktuk are needed which take into account any variations, among individuals or communities in methods of preparing muktuk, which might affect fat content of this food.

In the case of ringed seals and beluga, geographic comparisons can also be made with recent measurements on the same species in more highly contaminated locations. Ringed seals from the Baltic Sea have well-documented, elevated levels of PCBs and other OCs. Concentrations have declined since the 1970s (Blomkvist *et al.* 1992). PCB concentrations in juvenile ringed seals (1 yr old) sampled in the 1980s from the Baltic Sea averaged 17 $\mu g \cdot g^{-1}$ (Blomkvist *et al.* 1992), about 10 to 20 times higher than levels in ringed seals from the Canadian Arctic. Male beluga from southern Hudson Bay (Sanikiluaq) sampled in 1994 had mean ΣPCB concentrations of 6.8 $\mu g \cdot g^{-1}$ compared with levels of 79.2 $\mu g \cdot g^{-1}$ in male beluga sampled in 1993/94 from the St. Lawrence estuary. The difference of about 12-fold is lower than observed in the mid-1980s (about 25-fold) because of slightly higher mean levels in the Hudson Bay animals and declining concentrations in the St. Lawrence males.

- *Geographic coverage of levels of persistent OCs and metals in marine mammal and seabird populations in the Canadian Arctic is very good. Most stocks/populations of major species (e.g. beluga, ringed seals, walrus, polar bears, thick-billed murres, glaucous gull) have been studied. Little or no data is available on contaminants in minor species such as harbour seals, harp seals.*
- *There is much less information on contaminant levels and very limited geographic coverage for marine fish and invertebrates.*
- *There is evidence of PCB and DDT contamination of the nearshore marine environments at some locations in the Canadian Arctic due to pollution from military radar facilities and other activity. The detailed study of PCBs in nearshore areas of Cambridge Bay illustrates how PCBs can be transported from dump sites to the immediate marine environment. But these studies also show that the PCB contamination is quite localized when considered on a broad regional scale.*
- *The major OC contaminants in marine biota were toxaphene and PCBs. DDT and chlordane related contaminants are also important especially in polar bears and seals. Toxaphene predominates in lower food web organisms and does not biomagnify to the same extent as PCBs or some chlordane components, in polar bear.*
- *Slightly higher concentrations of PCBs are observed in ringed seals and polar bears from Hudson Bay compared to populations in the western or central Arctic. These west to east spatial trends are difficult to identify within the Canadian Arctic, but become obvious when examining the circumpolar data for these species. This trend parallels observations for PCBs and other OCs in caribou and waterfowl.*
- *Glaucous gulls from Prince Leopold Island and Brown Island in the central Arctic had much higher OC levels (by 3 to 10 times) than populations from the western Arctic. Different feeding habits on the breeding grounds as well as winter feeding areas for the eastern and western glaucous gulls are the possible explanations for these differences.*
- *The remarkable variation of OCs in walrus, in which about 35% of the 53 samples analysed had elevated concentrations of* $\sum$*PCBs* ($<1\ \mu g \cdot g^{-1}$)*, provides an example of the influence of diet on contaminant levels in top predators. The reason for the elevated levels is almost certainly from predation on seals. Local contamination was ruled out because levels of all OCs were elevated in each animal from Inukjuak, and elevated levels were also found in animals from other areas of the eastern Canadian Arctic.*

- *Contamination of marine biota by heavy metals shows consistent large regional differences. Polar bears, ringed seals and beluga all have higher mercury contamination in the western Arctic. In polar bears, levels of mercury and selenium were highest in samples from areas bordering the Beaufort Sea. High mercury appears to be related to elevated natural levels associated with the sedimentary geology of the western Arctic.*
- *The spatial trend for cadmium was opposite to that for mercury, i.e. higher in the eastern Arctic than the western Arctic for belugas and the rate of accumulation of cadmium in the liver was higher in the eastern Arctic. Again the geographical trend is probably related to the different mineralogy of the eastern Arctic.*
- *Cadmium levels in kidney and liver of Canadian Arctic marine mammals are as high or higher than those observed in the same or similar species in more temperate waters, but are similar to concentrations found in some terrestrial animals such as caribou and moose. Cadmium concentrations are also elevated i.e.* $>0.5\ \mu g \cdot g^{-1}$*, in muscle of beluga, ringed seals and adult sea birds.*
- *Relatively high levels of mercury and methylmercury are found in skin or muktuk of beluga. Since approximately 20% of the total mercury in the skin was lost annually through molting, a major route of elimination of mercury by cetaceans has been identified.*
- *Additional studies of metal and OC contaminants in muktuk need to be made in eastern Canadian marine populations because information is limited. These studies should take into account any variations, between individuals or communities, in methods of preparing muktuk, which might affect fat content of this food.*
- *Comparisons with other locations: PCB concentrations in juvenile ringed seals (1 yr old) sampled in the 1980s from the Baltic averaged 17* $\mu g \cdot g^{-1}$ *— about 10 to 20 times higher than levels in ringed seals from the Canadian Arctic. Male beluga from southern Hudson Bay (Sanikiluaq) sampled in 1994 had mean* $\sum$*PCB concentrations of 6.8* $\mu g \cdot g^{-1}$ *compared with levels of 79.2* $\mu g \cdot g^{-1}$ *in male beluga sampled in 1993-94 from the St. Lawrence estuary. Belugas from the St. Lawrence estuary had higher lead and mercury concentrations in kidney and liver tissues than arctic beluga.*

3.4 Temporal Trends of Contaminants in Biota

3.4.1 Marine Mammals

3.4.1.1 Organochlorines

Two groups of organochlorine compounds, the DDT-group of insecticides and the PCBs, are of particular interest for two reasons. First, there has been little local use of these compounds in the high Arctic and so the presence of these compounds in arctic biota indicates the global or at least hemispheric scale of the transport and distribution of these compounds. Second, the use of both groups of compounds in the western world essentially stopped during the early 1970s; this represents a "signal" that should be reflected in changes in OC concentrations in biota. The interval between the "signal" and the "response" will provide some information about time scales over which we can expect ecosystems to recover from the impact of these compounds.

PCBs and DDT in Holman Island ringed seals
A population of arctic ringed seals at Holman, NWT, has been studied continuously since the late 1960s to provide information about arctic seal population dynamics. This population was first sampled in the early 1970s to measure OC concentrations in the seal blubber. The first analyses (from samples taken in 1972) have provided a "base-line" with which to compare later sampling and analyses. These results also showed two other points: first, that variables such as age, sex and condition must be eliminated or at least recorded when interpreting blubber OC concentrations in seals (Addison and Smith 1974), and second, that OC concentrations in the Holman ringed seals were about one order of magnitude *lower* than those in seals from the more contaminated east coast of Canada. A re-sampling and re-analysis of this population in 1981 showed the following main points (Addison *et al.* 1986):

i) between 1972 and 1981, PCB concentrations in the Holman ringed seals fell by about 40%;
ii) DDT group concentrations in this population had not fallen significantly over that interval, and the metabolite *p,p'*-DDE represented the same proportion of the DDT-group as it had in the early 1970s.

The rate of decline in PCB concentrations was very similar to that seen in east coast harp seals (*Pagophilus groenlandicus*) from the Gulf of St. Lawrence (Addison *et al.* 1984), and almost certainly indicates a response to the ban on PCB manufacture and use during the early 1970s. Similar declines in PCB concentrations have been seen in fish and in birds from various sites in the Northern Hemisphere. The absence of any change in DDT-group concentrations was puzzling. In east coast seals, the DDT-group declined over a similar interval to about 25% of its concentrations in the early 1970s. Furthermore, in east coast seals sampled in the early 1980's, more of the DDT-group was present as *p,p'*-DDE, a persistent metabolite of *p,p'*-DDT which tends to accumulate as the DDT-group becomes "older". Thus the trends in DDT-groups concentrations and composition were consistent with the restrictions placed on DDT use in the early 1970s. Why did this trend not appear in arctic ringed seals? The simplest explanation was that DDT-group compounds continued to be transported to Arctic ecosystems at concentrations similar to those in the 1970s. The original source of the DDT, which appears to be transported to the Arctic is still not clear, but the far east, where DDT is still used extensively, seems to be the most probable.

A further sampling and analysis of the Holman ringed seal population was carried out in 1991, and archived samples from 1981 were re-analysed at the same time (Addison 1995a,b). Results are presented in Figure 3.4.1. (Since the original 1972 and former 1981 analyses had been carried out by the same methods, there were two sets of data (1972-1981 and 1981-1991) each of which was internally consistent; by comparing current and previous 1981 data, the earlier data were corrected to be consistent with the modern analyses. This was required only for PCBs and not for the DDT-group.) The 1981 and 1991 samples did not differ significantly ($p>0.05$) in their total DDT-group concentrations, in *p,p'*-DDE concentrations, nor in PCB concentrations (for convenience, individual PCB congeners were summed as Aroclor

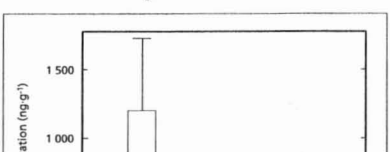


FIGURE 3.4.1

Temporal trends in ΣDDT and ΣPCB in blubber of female ringed seals from Holman in the western Canadian Arctic (Addison et al. *1986, Addison 1995b). Vertical bars represent one standard deviation.*

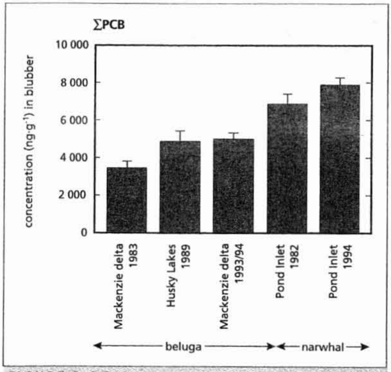


FIGURE 3.4.2 A

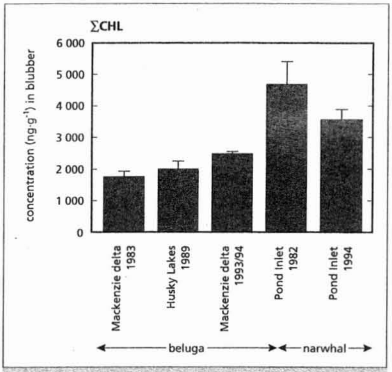


FIGURE 3.4.2 B

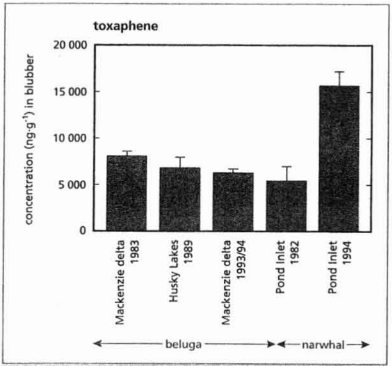


FIGURE 3.4.2 C

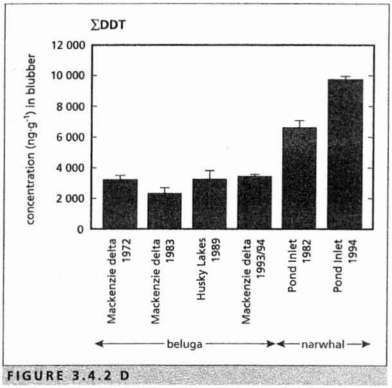


FIGURE 3.4.2 D

Trends in ΣPCB, ΣDDT, ΣCHL and toxaphene over a 10-year period (20-yr for DDT in the Mackenzie Delta) for male narwhal and beluga from the Beaufort Sea stock. Bars represent means ± S.E., N=8-26. Data for beluga from 1972 are from Addison and Brodie (1973).

1254 for comparisons with earlier data). Concentrations of *p,p'*-DDT concentrations fell significantly ($p<0.05$). HCB concentrations fell by about half ($p<0.05$) between 1981 and 1991 while α-HCH and γ-HCH concentrations remained unchanged (data not shown). Taking all these data together, we can summarize the trends in Holman Island ringed seals as follows:

i) between 1972 and 1991, PCB concentrations in blubber declined to about 30 % of their initial value and most of this decline took place from 1972 to 1981;
ii) DDT-group concentrations have declined slightly over the same interval; most of the decline has occurred during the 1980s rather than the 1970s.

Other ringed seals, beluga and narwhal populations
In the Canadian Arctic trends over 6 to 10 years have recently been examined in female ringed seals and male narwhal and beluga (Figure 3.4.2). Concentrations of ΣDDT, ΣPCB, ΣCHL and toxaphene have not declined significantly in female ringed seal blubber from Cumberland Sound (E. Baffin Island) or Barrow Strait in Lancaster Sound over six and nine years, respectively. However levels of these OC groups in animals from western Hudson Bay (Rankin Inlet) from 1986 were about 50% lower than observed in the (spring) Arviat samples collected in 1992, except for toxaphene. This difference is not consistent with trends elsewhere in ringed seals (for example, at Holman Island) or in polar bears, and may be due to

different feeding habits of the Arviat group. In male narwhal from the Lancaster Sound region concentrations of ΣCHL and ΣPCB have not declined significantly while ΣDDT and toxaphene are higher than in 1982 samples. Three sets of samples of male beluga (N=8 to 26 each time) from the Mackenzie delta region have been analysed since 1983 using identical methodology. Results for DDT in these animals from this stock were available from 1972 (Addison and Brodie 1973). No significant decline was observed in any of the four major OC groups over the 10-year period, and over 20 years in the case of DDT results.

3.4.1.2 Metals in Ringed Seals and Beluga

Mercury data for belugas sampled in 1993–94 and 1981–84 in the western Arctic, and 1993–1994 in the eastern Arctic were compared (Wagemann *et al.* 1996b). Results for all locations were pooled into two regional groups, "eastern" and "western" Arctic, because mean concentrations within each region were not significantly different (see Figure 3.3.22 for locations). The mean concentration in tissues of the recently sampled animals in the western Arctic was significantly higher (27.1±24.7 $\mu g{\cdot}g^{-1}$ wet wt., in the liver) than in belugas sampled a decade ago (11.8±12.1 $\mu g{\cdot}g^{-1}$ wet wt.) (Figure 3.4.3C). In the eastern Arctic, the mean concentration in the liver was also significantly higher in the more recently collected belugas (10.2±8.00 $\mu g{\cdot}g^{-1}$ wet wt.) than in belugas collected a decade ago (6.10±8.37 $\mu g{\cdot}g^{-1}$ wet wt.) (Figure 3.4.3C). These comparisons are confounded by differences in mean ages of the belugas between the two periods. To assess whether the concentration differences were due to age effects, it was necessary to compare the slopes of the regressions of concentration on age (Figure 3.4.3B).

In the western Arctic, the regression slope (mercury in liver on age of animals) of the belugas sampled in 1993-94 was significantly higher ($p=0.05$) than the regression slope for belugas sampled a decade ago (1981-84) (Figure 3.4.3B). A similar trend was observed in the eastern Arctic where the regression slope for the recently sampled belugas was also significantly higher than for belugas sampled a decade ago. This means that mercury in the liver accumulated at a higher rate (approximately two times) in more recent times in belugas that were ≥ 8 years old than it did in belugas sampled a decade ago, in both the western and eastern Arctic.

Ringed seals in the western Arctic had the same trend as belugas. The mean mercury concentration in the liver of 32.9±36.9 $\mu g{\cdot}g^{-1}$ wet wt. for those sampled between 1987 and 1993, was significantly ($p=0.05$) higher than for those sampled in 1972-73 (22.9±28.7 $\mu g{\cdot}g^{-1}$ wet wt.) (Figure 3.4.3C). The regression slope

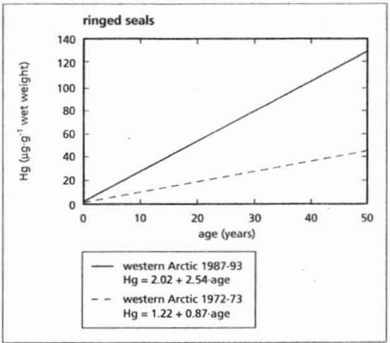


FIGURE 3.4.3 A

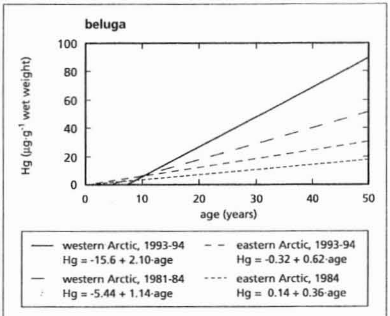


FIGURE 3.4.3 B

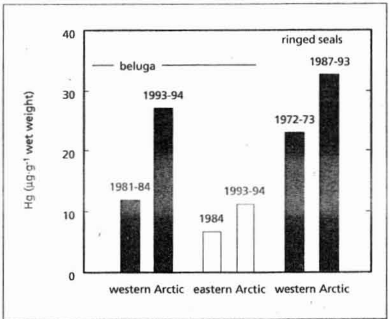


FIGURE 3.4.3 C

Relationship of mercury concentrations ($\mu g{\cdot}g^{-1}$) and age in: A. ringed seal liver, B. beluga liver and C. mean concentrations of mercury in ringed seal and beluga liver.

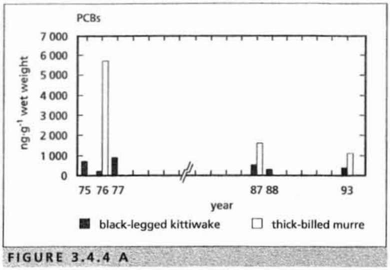


FIGURE 3.4.4 A

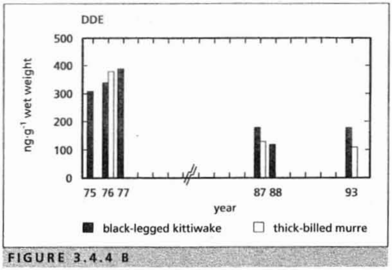


FIGURE 3.4.4 B

PCB and p,p'-DDE trends in arctic sea bird eggs from Prince Leopold Island (1975-1993).

for the more recently collected ringed seals was also significantly higher than for ringed seals collected 15 to 20 years ago. This indicates that ringed seals sampled in the period between 1987 to 1993 in the western Arctic accumulated mercury in the liver at a rate approximately three times higher, at all ages, than ringed seals sampled 15 to 20 years ago (Figure 3.4.3A). For the eastern Arctic, similar temporal data were not available for ringed seals. The regression results also indicate that the current rates of accumulation of mercury by belugas in the western and eastern Arctic were significantly different ($p<0.05$).

Cadmium accumulation rates in belugas from the eastern Arctic did not differ significantly between 1993-94 and 1981-84 sampling periods. There were no data available for cadmium from animals sampled in 1981-84 in the western Arctic. The spatial trend for cadmium was opposite to that for mercury (i.e. higher in the eastern Arctic than the western Arctic for both belugas and ringed seals) and the rate of accumulation of cadmium in the liver was higher for both species in the eastern Arctic.

3.4.2 Birds

3.4.2.1 Sea Birds

The sea bird egg has been shown to be an efficient, conservative tool for monitoring OC levels in the marine environment (Gilbertson *et al.* 1987). Sea bird colonies in the high Arctic tend to be large, dominated mainly by thick-billed murres, northern fulmars, and black-legged kittiwakes, with smaller numbers of other gulls (Brown 1986). These species represent different foraging habits as described in Section 3.3.3.4. At the high Arctic colony of Prince Leopold Island (see map, Figure 3.3.25, Section 3.3.3.4), OC residues have been monitored in eggs and livers of adult birds of thick–billed murres, northern fulmars and black-legged kittiwakes since the mid-1970s. Collection and analytical protocols for samples collected during 1975-77 and 1987 followed methods described in Nettleship and Peakall (1987). Methods for the collection and analysis of the 1993 samples are described in Section 3.3.3.4.

TABLE 3.4.1

Historical trends of organochlorine residue levels in livers of adult northern fulmars from Prince Leopold Island (ng·g^{-1} lipid wt.)

Date	N	Lipid	*p,p'*-DDE	PCB[1]
1975	7	4.2	5 950	25 700
1976	7	8.1	6 170	24 300
1987	8	3.6	3 890	15 600
1993	10	3.3	5 060	10 200

[1] *PCB measured as Aroclor 1254:1260 (1:1)*

Comparison of OC levels in eggs of black-legged kittiwakes and thick-billed murres on Prince Leopold Island show that OCs (i.e. *p,p'*-DDE, PCB as Aroclor 1254:1260, oxychlordane, HCB, dieldrin, heptachlor epoxide), in general, have declined from the mid-1970s to the late 1980s (Noble and Burns 1990). By 1993, most of the residue levels have continued to decrease or have reached a threshold where they are leveling off. This is clearly illustrated by the pattern of decline in PCBs and *p,p'*-DDE in eggs of thick-billed murres and black-legged kittiwakes (Figure 3.4.4). Residue levels measured in livers of adult northern fulmars in the mid-1970s, mid-1980s (Nettleship and Peakall 1987, Noble and Elliott 1986) and again in 1993 also indicate a decline in residue levels (Table 3.4.1). The declining levels of OCs in these migratory species (see Section 3.3.3.4) reflect an overall reduction in the OC levels of the North Atlantic where many of these birds overwinter. Declines in organic contaminants have been observed in both inshore and continental shelf sea birds on both the Atlantic and Pacific coasts of Canada (Elliott *et al.* 1992b), in the Barents Sea area (Savinova *et al.* 1995) and the Baltic Sea (Bignert *et al.* 1995). Declining levels of organic contaminants are also reflected in other northern biota (Andersson *et al.* 1988).

TABLE 3.4.2

Organochlorine residues (µg·g^{-1} wet wt.) in Peregrine Falcon eggs from Rankin Inlet.

Compound	1991-1994 (n=20)		1982-1986			Difference	
	Range	mean[1]	Range	mean[1]	n	Differences	p
PCB[2]	1.67–45.6	8.31	1.95–47.8	8.74	36	t_{54}= 0.36	0.72
HCB	0–0.165	0.030	0–0.15	0.03	22	t_{40}=–0.374	0.711
HCH	0–0.028	0.002	—	<0.02	19		
Oxychlordane	0.038–0.909	0.211	0.08–0.80	0.21	21	t_{39}= 0.617	0.541
Hept epoxide	0.045–1.39	0.265	0.09–5.92	0.02	36	t_{54}= 1.655	0.104
Dieldrin	0.045–1.80	0.361	0.13–1.66	0.41	36	t_{54}= 1.298	0.200
p,p'–DDE	0.76–28.1	4.45	1.79–29.3	7.59	36	t_{54}= 2.849	0.006
p,p'–DDD	0–0.203	0.012	0–0.37	0	22	t_{40}= 1.295	0.203
p,p'–DDT	0–0.033	0.004	0–1.41	0	36	t_{54}=2.021	0.048
Mirex	0.062–1.82	0.496		NM[3]			

[1] *Geometric mean concentrations*

[2] *As Aroclor 1254:1260(1:1)*

[3] *Not measured*

3.4.2.2 Peregrine Falcon

Populations of the arctic-breeding tundra peregrine falcon (*Falco peregrinus tundrius*) have been monitored since the mid-1970s for possible effects of OC pesticides. However, no temporal studies of contaminant levels for a single population of *F. p. tundrius* exist, and the data available are confused because data sets were collected in a haphazard manner, and are temporally discontinuous and spatially disjoint (Peakall *et al.* 1990, Thomas *et al.* 1992).

Between 1991 and 1994 samples of peregrine eggs, eggshell, blood plasma, tissue and prey species were collected to establish the present level of contamination of a population of tundra peregrine falcons and their prey species, and to delineate temporal trends by comparing present levels with contaminant levels measured between 1982-1986 for the same population studied by Court *et al.* (1990). Further details on sampling and analysis are given by Johnstone *et al.* (1996).

A total of 28 eggs, representing 20 clutches were collected for analysis between 1991 and 1994. Organochlorine residue levels in egg contents are summarized in Table 3.4.2. Mean *p,p'*-DDE levels show a decline from 7.6 µg·g^{-1} in the 1980s to 4.5 µg·g^{-1} in the 1990s. No other significant improvements in residue levels were detected between decades. The mean *p,p'*-DDE level of 4.5 µg·g^{-1} is below that reported in other recent studies of tundra peregrines. Ambrose *et al.* (1988) studying *F. p. tundrius* in Alaska, reported mean *p,p'*-DDE levels of 9.3 µg·g^{-1}, almost twice the mean level found in Rankin Inlet. For *F. p. tundrius* in Canada, Peakall *et al.* (1990) reported a mean of 6.8 µg·g^{-1} for 26 eggs collected between 1980 and 1986. Since the use of some of these OC chemicals were restricted, or even banned in the 1970s, recovery of some of the affected populations has been noted (Court 1993, Shank *et al.* 1993, Peakall 1990, Newton 1979). Recent reviews show a clear trend of decreasing OC contamination of the American peregrine falcon (*Falco peregrinus anatum*) from the late 1960s through to the present day (Court 1993, Peakall *et al.* 1990).

3.4.3 Fish

3.4.3.1 Organochlorines

Temporal trends in OCs in Lake Laberge were examined by analysis of archived round whitefish. These fish were collected in 1974 and stored frozen at -20 to -40°C in sealed plastic bags. Eleven samples from 1974 were analysed for PCBs and toxaphene, and compared with results from 20 lake whitefish collected in 1992 using lipid-normalized, log-transformed concentrations (Figure 3.4.5). Whitefish collected in 1974 had higher concentrations of ΣDDT, ΣHCH and dieldrin and lower concentrations of toxaphene, chlordane and ΣPCBs than those from 1992 on a lipid weight basis. The higher DDT in 1974 is in agreement with results for sediment that showed higher DDT concentrations at that time in sediments than in present day sediments. Age was

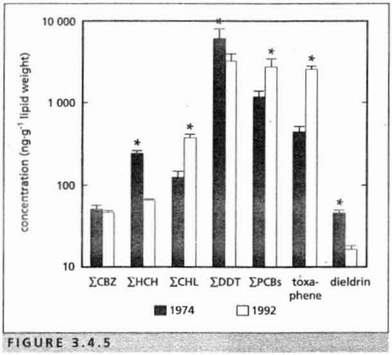


FIGURE 3.4.5

Organochlorines (geometric means±S.E.) in whitefish muscle from Lake Laberge (1974, N=11 and 1992, N=20). Samples from 1974 are from round whitefish; those from 1992 are lake whitefish. Significant differences (*) between years (p<0.05) were observed from ΣHCH, ΣCHL, ΣDDT, ΣPCB, toxaphene and dieldrin.

unlikely to have influenced the results because both groups had the same average age (11 years). Migration of round whitefish from Lake Laberge and dietary differences between the two species could have influenced the comparison.

Burbot were first collected at Fort Good Hope on the Mackenzie River for contaminant analysis in 1986 and in 1989 as part of work that tried to explain reports by local residents about the poor quality of some burbot livers. Collection of additional samples in 1994 permitted comparison over the eight-year period using lipid-normalized log-transformed results (Figure 3.4.6). Toxaphene, ΣHCH and chlordane levels have undergone significant declines over the eight-year interval. No significant declines were observed for ΣDDT, ΣPCBs, ΣCBz, or dieldrin.

3.4.3.2 Metals

Walleye *(Stizostedion vitreum)* from Lac Ste. Therese, in northwestern NWT, were analysed for mercury in 1975, 1980 and 1992. Mean lengths of the fish (465–504 mm) did not differ significantly among years and mean mercury concentrations were also not significantly different (DIAND 1994). There is little additional temporal trend information on mercury in fish. Lockhart (1995) in a review of DFO Fish Inspection records of mercury in anadromous char since the 1970s concluded that there was no obvious increase in mercury over a 20-year period. Clearly a future priority is to determine if levels of mercury are increasing in fish as they appear to be in lake sediments.

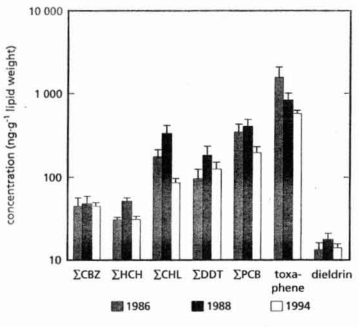


FIGURE 3.4.6

Temporal trends of organochlorines in burbot liver collected from the Mackenzie River at Ft. Good Hope. Bars represent geometric means±S.E. of lipid-normalized concentrations. Significant declines were observed (1986/88 vs. 1994) for ΣCHL and toxaphene.

3.4.4 Overall Assessment of Temporal Trends

Many of the temporal trend data for contaminants in Canadian Arctic biota are confounded by changes in analytical methodology, as well as by variability due to age/size, or to dietary and population shifts. The best data are the long-term studies (20 years) of OCs in ringed seal blubber at Holman Island and in sea birds at Prince Leopold Island in Lancaster Sound. Both studies show declining concentrations from the 1970s to 1980s, then a more recent leveling off. These declining concentrations of ΣDDT and PCBs in marine biota are consistent with a general decline of OC concentrations that has been observed in marine mammals and sea birds from eastern Canadian waters (Addison *et al.* 1984, Beck *et al.* 1994, Westgate 1995, Elliott *et al.* 1989), and in lake trout in Lake Ontario (Borgmann and Whittle 1991). Beck *et al.* (1994) reported a significant ($p<0.05$) 1.7-fold decline in PCBs in male harp seals from Les Escoumins in the St. Lawrence estuary between 1982 and 1989, but no decline of ΣDDT over the same period ($p>0.05$). These authors cautioned that their earlier results may have underestimated *p,p'*-DDT and PCBs so that the decline between 1982 and 1989 could have been greater. The decline in concentrations of DDT and PCBs in harp seals, as well as in lake trout from Lake Ontario, was greater during the 1970s immediately following bans on use of DDT and open use of PCBs, than during the 1980s. Similarly, the decline in concentrations of PCBs in Holman Island ringed seals was more pronounced between 1972 and 1981 than from 1981 to 1989. ΣDDT showed a slower decline than PCB in Holman Island ringed seals over the period 1972 to 1981, but a faster decline in the 1980s.

The lack of declines of ΣDDT and PCB concentrations in eastern Arctic ringed seals, narwhal and sea birds during the 1980s and early 1990s is consistent with the slow rate of decline observed elsewhere in southern Canada. The lack of decline of ΣDDT in male beluga from the Mackenzie delta between 1972 and 1993/94 was surprising, but probably related to methodological differences between more recent (capillary GC separation, and earlier (packed column GC) determination of DDT-related compounds.

The lack of any significant change in ΣDDT and ΣPCB concentrations in arctic beluga over an eight-year period contrasts with a small but significant decline in ΣPCB in St. Lawrence (male) beluga over the same period. The average difference in PCB concentrations between Hudson Bay animals and male St. Lawrence beluga in the mid-1980s was about 25-fold. It is now about 12-fold. The smaller difference is because of slightly higher mean levels in the Hudson Bay animals resulting from analysis of a larger and probably more representative sample, combined with a slower decline in the arctic animals, and declining concentrations in the St. Lawrence males (1.9-fold over 12 years) (Muir *et al.* 1996).

The decline of ΣDDT in peregrines was greater than that observed for arctic sea birds. It may reflect declining concentrations in their wintering grounds. Recent reviews have shown a clear trend of decreasing PCB and OC pesticide contamination of the peregrine falcon from the late 1960s to the 1990s (Court 1993, Peakall *et al.* 1990). In the tundra peregrine, the decline is less pronounced. The effect of continuing use or persistence of OC pesticides within the wintering ground of the arctic-breeding peregrines and their prey remains unclear. The dietary intake of OCs by peregrines is related to the species available as prey, and their contaminant load. Consequently, it is difficult to distinguish spatial trends related to differences in prey use between populations, from temporal trends in contaminant levels, when samples are collected opportunistically from scattered populations (Peakall *et al.* 1990, Thomas *et al* . 1992). Future work must focus on repeated sampling of selected populations to establish true temporal trends in contaminant levels in peregrines and other birds of prey. In summary, the ban placed on PCB manufacture and use in the early 1970s led to a reduction of PCB concentrations in seals and sea birds during the 1970s. This decline cannot be easily discerned by comparing concentrations in samples from the 1980s and early 1990s. The results imply continued inputs of PCBs and DDT to Arctic marine food webs large enough to maintain concentrations at steady state since the early 1980s.

The above discussion has focused on DDT and PCBs. With the exception of limited data on chlordane, and chlorobenzenes in sea birds and polar bears, there is less information on temporal trends of other OCs in marine biota. Limited data for the 1980s to early 1990s suggest few significant declines in concentrations in marine mammals or sea birds. Sex and age are not major confounding factors in the temporal trend studies of OCs in marine mammals or sea birds because comparisons are restricted to female seals and male beluga, and bird eggs, all of which show no correlations of contaminant levels with age.

Temporal trends of mercury and cadmium in ringed seals and beluga have recently been determined for the first time. The results show higher mean concentrations of mercury in more recent (1993/94) samples than in earlier collections (1981-83 in eastern Arctic, 1972-73 in western Arctic). These results are confounded by age differences in the samples analysed. When the slopes of concentrations on age are compared, the same conclusion is reached: rates of accumulation are higher in present day animals than 10 to 20 years ago. The fact that cadmium concentrations in the same animals (eastern Arctic only) showed no change over a 10-year period further confirms the elevation in mercury because it rules out the possibility of dietary changes, for example, a possible shift to invertebrates which may have higher mercury and cadmium than arctic cod. No temporal trend data are available for metals in sea birds or polar bears.

The apparent increasing levels/rates of accumulation of mercury in ringed seals are perhaps the most striking new result in this assessment of temporal trends. They are consistent with observations of increasing anthropogenic mercury deposition in lake and marine sediments (Lockhart 1995, Lockhart *et al.* 1995) in the eastern Arctic and in Greenland (Dietz *et al.* 1995). Over the longer term there has been an increase of mercury in human hair in Greenlanders and in seal hair from Greenland, of about three- to four-fold (Hart Hansen *et al.* 1991) over the past 400 years. Whether these increases are due entirely to anthropogenic sources or to a combination of natural variation of geochemical releases of mercury to the marine environment is not clear.

There is very limited information on temporal trends of other metals in Arctic marine biota. In bivalves from Pangnirtung fiord, lead concentrations in shells of living animals were five-fold higher ($2.5 \pm 0.4\ \mu g \cdot g^{-1}$) than in fossil shells from circa 8200 BP (<0.5-$0.8\ \mu g \cdot g^{-1}$) (Bourgoin and Risk 1987). There are few reliable older measurements of lead in ringed seals with which to make temporal comparisons.

In the terrestrial and freshwater environment there is very limited temporal trend information for OCs or metals. No temporal trend information for OCs or metals appears to be available for caribou or mink. Declining concentrations of toxaphene and other OCs were observed in burbot liver from the Mackenzie River at Fort Good Hope. The largest declines were seen for ΣPCBs and the smallest for ΣDDT similar to the trends in Holman Island ringed seals. Comparison of OC concentrations in round and lake whitefish from Lake Laberge over an 18-year interval showed a significant decline in ΣDDT, dieldrin and ΣHCH, but increases in ΣPCBs, ΣCHL and toxaphene. This study, along with the work on burbot liver, had the advantage of using the same methodology as on the earlier samples. However, the earlier samples were from round whitefish rather than lake whitefish and, even though mean ages were almost identical, dietary and % lipid differences between the two species may account for some of the trends. In summary, the evidence for declining OC concentrations in the Arctic freshwater environment is very limited due to lack of retrospective samples and of past studies of OC concentrations.

The limited data on temporal trends in mercury in freshwater and anadromous fish in the NWT suggests no increase in mercury over a 20-year period. However, this conclusion is based on results from walleye in one lake known for high natural sources of mercury and on very limited numbers of arctic char from a large number of lakes in the Keewatin/Baffin region.

At present the temporal trend data are too limited to be strongly predictive for most OCs and metals because they are based on only two or 3 sampling intervals. By comparison, temporal trend data for contaminants in Lake Ontario lake trout (Borgmann and Whittle 1991) and in various species from the Baltic Sea and from Lake Storinveld in northern Sweden (Bignert *et al.* 1993, 1995) are available yearly for a 15- to 20-year period. There is clearly a need for well-designed temporal trend studies utilizing tissues from tissue banks created by the Northern Contaminants Program at selected, well-characterized locations.

- *Long-term studies (20 years) of OCs in ringed seal blubber at Holman Island and in sea birds at Prince Leopold Island in Lancaster Sound show declining concentrations from the 1970s to 1980s then a more recent leveling-off (mid-1980s to mid-1990s). The decline and leveling-off in concentrations of ΣDDT and PCBs in marine biota are consistent with the general trend of OC concentrations that has been observed in marine mammals and sea birds from eastern Canadian waters and in lake trout in Lake Ontario.*
- *The decline of ΣDDT in peregrine falcons was greater than observed for arctic sea birds. It may reflect declining concentrations in their wintering grounds. Recent reviews have shown a clear trend of decreasing PCB and OC pesticide contamination of the American peregrine falcon from the late 1960s to the 1990s.*
- *There is much less information on temporal trends of other OCs (such as toxaphene, chlordane, and chlorobenzenes) in marine biota. Limited data for the 1980s to early 1990s suggests that there has been no significant decline in the concentrations of these other OCs in marine mammals or sea birds during this period.*
- *Higher mean concentrations and higher rates of accumulation of mercury were found in ringed seals and beluga in more recent (1993–94) samples than in earlier collections (1981-83 in eastern Arctic, 1972-73 in western Arctic). The results are consistent with observations of increasing anthropogenic mercury deposition in lake and marine sediments (especially in the eastern Arctic).*
- *Cadmium concentrations in the same animals (eastern Arctic only) showed no change over a 10-year period.*
- *No temporal trend data are available for metals in sea birds or polar bears or other Arctic marine biota other than ringed seals and beluga.*
- *There is very limited temporal trend information on OCs or metals in the terrestrial and freshwater environments. For example, no temporal trend information for OCs or metals appears to be available for caribou or mink.*
- *Declining concentrations of toxaphene and other OCs were observed in burbot liver from the Mackenzie River at Fort Good Hope. Comparison of OC concentrations in round and lake whitefish from Lake Laberge over an 18-year interval showed a significant decline in ΣDDT, dieldrin and ΣHCH but increases in ΣPCBs, ΣCHL and toxaphene.*
- *At present, the temporal trend data are too limited to be strongly predictive for most OCs and metals because they are based on two or at most three sampling times. By comparison, temporal trend data for contaminants in Lake Ontario lake trout and in various species from the Baltic and from Lake Storinveld in northern Sweden are available yearly for a 15- to 20-year period.*
- *There is clearly a need for well-designed temporal trend studies utilizing tissues from tissue banks created by the Northern Contaminants Program at selected, well-characterized locations.*

3.5 Biological Effects

3.5.1 Introduction

Although arctic biota contain a range of organic and inorganic contaminants there has been relatively little study of the biological effects of these chemicals. Unlike the situation in the Baltic Sea or the Great Lakes, there is currently no evidence to support or refute arguments that chemical contaminants are present in sufficient quantities to affect reproduction or survival. A series of projects was developed under the Northern Contaminants Program to address this issue by examining indicators of biological responses (biomarkers or bioindicators) to contaminants. Most of the biomarkers selected had established sensitivities (in laboratory animals) to some of the same contaminants measured in Arctic marine mammals and fish, notably several PCB congeners, PCDD and PCDFs, and PAHs including petroleum oil.

Almost any biological change, from molecular to ecological, can serve as a biomarker or bioindicator. However, the terms most often refer to changes at sub-cellular levels (McCarthy and Shugart 1990, Huggett *et al.* 1992, Peakall 1992). Biomarkers measurable at a molecular level are quickly evident, but are not readily interpreted at the population level, while measures with established ecological relevance, like species diversity or population size become evident too late to have diagnostic or preventative value. Biomarkers typically are measures of normal processes that take on abnormal values as a result of exposure to chemicals of interest. That is, biomarkers often measure a quantitative, rather than qualitative, difference between exposed and unexposed individuals. This raises the statistical problems of determining whether the value from a given individual falls within the normal range for the species and biomarker under investigation, and whether the value exceeds the level at which deleterious effects on the individual are likely. There has been so little work on biomarkers in arctic animals that the range of normal (unexposed, or background) responses for these indicators has not been well defined. Except for those biomarkers used in human or veterinary medicine, non-human biomarkers generally have not yet been calibrated adequately against ecologically meaningful responses; these linkages are under active investigation in many laboratories. The contributions of biomarkers are varied, but one of the most valuable has been that some are quite specific: they respond to only a few types of chemical exposures and so they offer powerful tools to support or refute hypothetical cause/effect relationships. Examination of animals for responses known to be associated with the contaminants found in them is perhaps the only way to make a convincing case either for or against the hypothesis that trace contaminants are acting biologically on the animals. If the contaminants are affecting the animals, then we should expect to see some correlations between biomarker values and contaminant levels.

Biomarker studies in arctic biota have been largely confined to cytochrome P450 (CYP) enzyme induction in liver. Cytochrome P450 mixed function oxygenase (MFO) enzymes are central to the metabolism of a wide array of endogenous compounds (e.g. hormones) and anthropogenic organic pollutants. Members of the P450 subfamilies (e.g. isozymes CYP1A1, CYP2B) are important in the metabolism of many pollutants. CYP1A is induced by planar organics such as PAH, 2,3,7,8 chlorine-substituted PCDDs and PCDFs and 3,4,3',4' chlorine-substituted n PCBs. Concentrations of CYP1A protein in liver are therefore an index of exposure to TCDD-like chemicals, and potentially an indicator of TCDD-like effects. CYP2B is induced in mammals by a wide variety of halogenated compounds, including organochlorine pesticides and ortho-substituted PCBs. In practice, liver microsomes are isolated by centrifugation and the enzyme activity determined spectrophotometrically using standard substrates and by immunochemical techniques. Two of the most widely used substrates used to measure CYP1A1 activity are benzo(a)pyrene, which is used to measure aryl hydrocarbon hydroxylase (AHH) activity, and ethoxyresorufin, which is used for ethoxyresorufin-O-deethylase (EROD) activity. Immunochemical techniques are used to determine levels of the isozymes directly using antibodies raised against the enzymes in test animals.

While CYP induction is a good measure of exposure, and may correlate with toxic effects, there have been very few studies of actual toxic effects at any level of organization. Knowledge of population sizes, fecundity and survival are relatively unknown except for polar bears, so statements cannot be made about the lack or presence of effects at the population level. Even in the case of polar bears, concentrations of organochlorines were relatively high and stable during the last 25 years when population information has been gathered, so it is not possible to speculate on the population-level effect of contaminant exposure. A Norwegian study showed that retinol (vitamin A) concentrations in polar bear plasma were negatively correlated to PCB concentrations (Skaare

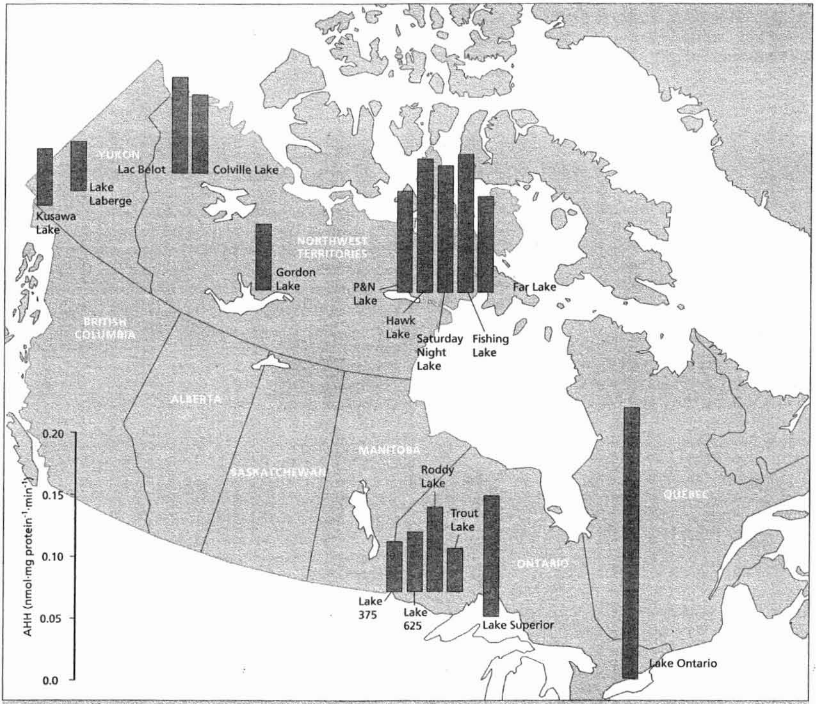


FIGURE 3.5.1

Lake trout liver microsomal AHH (nmol·mg protein^{-1}·min^{-1}) at 25°C.

et al. 1994b). There may also be immune suppression in polar bears, based on studies of marine mammals in Europe and elsewhere. Our understanding of the impact of PCBs and other organochlorines on arctic biota at high trophic levels is poor, and further research in areas such as immunotoxicology is warranted and necessary.

3.5.2. Biochemical Markers in Fish and Marine Mammals

3.5.2.1 Cytochrome P-450 Activities in Fish

The cytochrome P-450 system has probably been one of the most widely used biomarkers to date, with numerous laboratory and field cases of responses established. The preferred field study design has been the comparison of an exposed group of individuals with similar groups not exposed to the same source. For example, Figure 3.5.1 shows liver microsomal cytochrome P450 (CYP1A1) activity (as AHH) in collections of lake trout from several locations in northern Canada. Two collections from the Great Lakes are also included, with that from Lake Ontario showing a clear excess over any of the arctic collections. Collections such as these provide insight into basal ranges of biomarker values and enable the recognition of abnormal values when they are encountered. In this instance, the Lake Ontario lake trout stands out clearly from the other collections. This population is exposed to higher amounts of a variety of chemical contaminants (especially PCBs and PCDD/Fs) and reproductive failure was observed during the 1970s and 1980s (Government of Canada, 1991).

Another design, less commonly encountered, is the comparison of individuals within a group to search

TABLE 3.5.1

Mixed function oxidase enzyme activity (nmol·mg protein^{-1}·min^{-1}) in liver from fish in Lake Laberge and Kusawa Lake.

Location	Species	Sex	N	EROD	AHH	TEQ (pg·g^{-1})[1]
Laberge	Lake trout	M	5	0.0130±0.0040	0.0442±0.0247	47±42
		F	2	0.0085	0.0350	11
Laberge	Burbot	M	10	0.0095±0.0040	0.0077±0.0040	255±130
		F	10	0.0041±0.0020	0.0061±0.0036	83±14
Laberge	Whitefish	F	6	0.0132±0.0061	0.0373±0.0142	
Kusawa	Lake trout	M	12	0.0226±0.0139	0.0547±0.0234	
		F	8	0.0134±0.0107	0.0359±0.0185	

[1] *Results from Muir (1992) based on concentrations of non-ortho- and mono-ortho PCBs.*

for statistical linkages between biomarker values and exposure to contaminants (as indicated by levels of chemical residues). This is a valuable approach for those contaminants stable enough to remain identifiable as residues. For example, Hodson *et al.* (1992) found that catalytic activities varied directly with chemical residues of 2,3,7,8-TCDD and -TCDF in fish taken from near a bleached kraft pulp mill in Québec, although more recent studies have indicated that unidentified components, in addition to PCDD/Fs, can induce the cytochrome P-450 system (Munkittrick *et al.* 1994, Servos *et al.* 1994).

CYP1A1 enzyme activity (as EROD) in lake trout liver from Laberge and Kusawa lakes was similar (Table 3.5.1), despite higher levels of PCBs in the Lake Laberge fish. Higher levels of TEQs were found in burbot liver than in lake trout muscle from Lake Laberge but EROD and AHH activities were lower in burbot. Both burbot and whitefish were in spawning condition, a factor known to influence CYP1A1 enzyme activity. EROD and AHH activity in lake trout is similar to those reported for lake trout from the Hawk Lake (near Chesterfield Inlet, NWT) (Lockhart 1995). ERODs in male lake trout from Laberge are about two-fold higher than that found in Buchanan Lake char but similar to levels observed in char from Amituk Lake (Lockhart 1992). There appeared to be no correlation between total TCDD equivalents in lake trout muscle and burbot liver based on the analysis of a subset of the fish reported in Table 3.5.1.

3.5.2.2 Cytochrome P450 Enzyme Induction in Beluga and Ringed Seals

The principal biomarker being used is the microsomal monooxygenase system since it has been associated both with exposures to planar aromatic compounds and with a cascade of other biological responses. There is a strong correlation between liver microsomal EROD or AHH activities and blubber residues of several PCB congeners in a group of beluga whales from the western Canadian Arctic (Figure 3.5.2) (Lockhart and Stewart 1992, White *et al.* 1994). While this is not proof of a causal relationship between the PCBs and the biomarkers, it is probably the closest we can come to connecting residues and responses in animals that are not available for experimental study. These particular whales entered a freshwater lake system during the open-water season and became trapped there by the onset of winter. Breathing holes became smaller and smaller and native hunters decided to take the whales in mid-winter. They were about 200 kg lower in body weight than whales of their length should have been. The hypothesis suggested by these data is that the whales mobilized blubber fats during the period of poor feeding in freshwater and concomitantly mobilized fat-soluble contaminants which then acted to produce the correlations. For obvious reasons we cannot reproduce this observation on whales subject to known, experimental diets, but we have been able to reproduce the effect in fish in the laboratory. Arctic char received an initial low dose of congener CB126 and were then maintained at different feeding rates for several months. Fish receiving the low rations responded by increased cytochrome-P-450 catalytic activities. These observations on the beluga whales are the first known statistical inference that current body burdens of contaminants in an arctic species can be associated

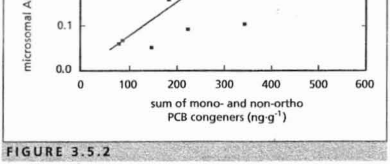


FIGURE 3.5.2

Aryl hydrocarbon hydroxylase (AHH, nmol·mg microsomal protein^{-1}·min^{-1}) activities in liver microsomes of beluga whales taken from the Eskimo Lakes area of the Mackenzie Delta as a function of PCB congener concentrations in blubber (White et al. *1994).*

with subtle effects on the animals. Clearly, they point to a possible interaction between diet and the pharmacology of organochlorines and suggest that existing body burdens may be sequestered effectively when blubber reserves are high but may become significant during a poor feeding season.

In an earlier review of this subject, Lockhart *et al.* (1992) concluded that contaminant levels in arctic animals had probably not reached levels high enough to exert biological responses. Since then, better biomarker data have emerged showing strong statistical correlations with measured residue levels. Consequently, in a subsequent review, Lockhart (1995) could not retain the earlier conclusion. The new data suggest that even the relatively low levels of contaminants present in arctic animals may not cause effects, especially during years of poor feeding. Other studies not shown here have shown weak but suggestive associations between biomarkers and contaminant residues in burbot and ringed seals. As is usually the case with arctic animals, the lack of experimental dosage/response data continues to limit the ability to interpret effects in arctic animals.

3.5.2.3 Hepatic Cytochrome 450 Enzymes in Polar Bears

The Cytochrome P450 enzyme system of the polar bear was characterized by Bandiera *et al.* (1995) by immunochemical techniques. This was the first study of MFO activity in polar bears. There is only one other study on an ursid species. Several families and subfamilies of P450 enzymes, including CYP1A and CYP2B, in polar bears were found to be similar to those in rats based on immunochemical cross-reactivity.

Correlations among immunoquantitated CYP1A and CYP2B protein concentrations, enzyme activities and OC concentrations were made in livers of 16 male polar bears (Letcher *et al.* 1995). Activities of the enzymes EROD, pentoxyresorufin (PROD) and benzyloxyresorufin (BROD) were found to increase with CYP1A content, and did not correlate with CYP2B content. EROD activity was shown to be CYP1A-mediated from inhibition of its activity by antibodies to CYP1A (Letcher *et al.* 1995). EROD activities leveled off, but CYP1A protein concentration continued to increase with increasing organochlorine concentrations. Therefore, immunoquantitated CYP1A protein is a more reliable measure of exposure of polar bears to TCDD-like compounds and their potential effects than EROD activity. CYP1A content in male polar bear liver was highly correlated with total PCBs ($p<0.0001$) as well as TCDD equivalents ($p<0.0004$) (Figure 3.5.3a).

Mean EROD activity in male polar bears was about two times higher than in beluga liver (White *et al.* 1994) and 10-times higher than activity in male

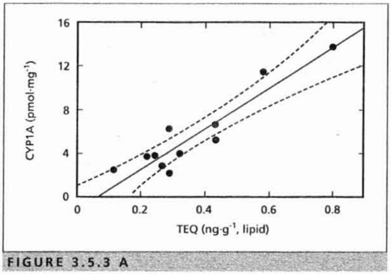


FIGURE 3.5.3 A

Correlation between CYP1A1 and CYP1A2 content in male polar bear hepatic microsomes vs. 2,3,7,8-TCDD toxic equivalents (TEQs) using the TEF indices given by Safe (1990).

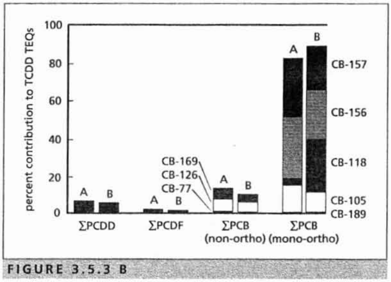


FIGURE 3.5.3 B

Mean contributions of non-ortho and mono-ortho PCBs, PCDDs and PCDFs to the total TEQ composition of 13 polar bear livers using a TEF for CB118 of 0.0001 (A) (Ahlborg et al. 1994) or a TEF of 0.001, (B) (Safe 1990).

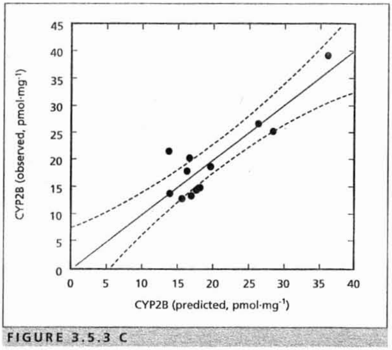


FIGURE 3.5.3 C

Correlation between observed CYP2B content in male polar bear hepatic microsomes vs. levels of CYP2B predicted from ortho-substituted PCBs and total chlordane levels (mainly oxychlordane) by backwards, stepwise, multiple linear regression. The regression was highly significant, $r^2=0.784$, $p<0.0005$. Data are from Letcher et al. *(1995c).*

hooded seals (Goksøyr *et al.* 1992). Assuming that the measurements of EROD activity are comparable among laboratories, this means that the CYP1A-mediated activity was somewhat elevated in polar bears, and raises the possibility of TCDD-like toxic effects. The large contribution of mono-ortho PCBs (mainly CB156 and CB157) to TCDD equivalents in polar bear liver suggests that these compounds are mainly responsible for induction of CYP1A protein and EROD activity (Letcher *et al.* 1995). PCDD/Fs and non-ortho PCBs contributed only a small fraction of the total TCDD equivalents in polar bear liver. Based on comparison of PCB patterns in ringed seal, there is strong evidence that polar bears metabolize TCDD and PCBs with similar toxic action (CB126, CB118 and CB105), more rapidly than do other mammals studied to date, possibly because of high CYP1A induction.

Stepwise, backward multiple regression showed that the CYP2B content in male polar bear liver was only correlated ($p<0.0005$) to a combination of total chlordane and total ortho-substituted PCB concentrations in liver (Figure 3.5.3b). CYP2B activity was much more weakly correlated to PCBs alone ($p<0.03$). Thus, CYP2B activity in polar bears is probably due to the combined inducing effect of PCBs and chlordanes (mainly oxychlordane), which together constitute over 95% of total organochlorine concentration in liver. The toxic significance of this induction is not known, however CYP2B-inducing chemicals are more likely to be implicated in neurobehavioural and some endocrine disruption effects than the TCDD-like compounds.

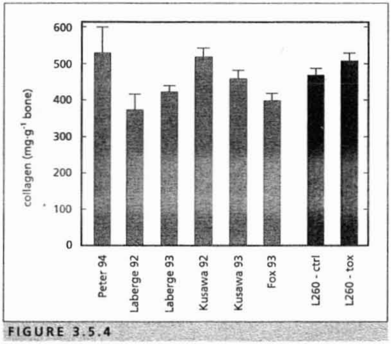


FIGURE 3.5.4

Comparison of bone collagen in lake trout (mean ± S.E.) from Peter Lake, Yukon Lakes, toxaphene-treated fish in ELA L260 (Delorme 1995) and control fish.

3.5.2.4 Bone Collagen and Hydroxyproline in Fish

Bone hydroxy-proline has been shown to be depressed in fish during exposure to toxaphene (Mayer *et al.* 1977). Lake trout collected in 1992 from Lake Laberge had significantly lower levels of hydroxyproline than lake trout from Kusawa or from northwestern Ontario (Lake 260) (Figure 3.5.4). Fish in Lake 260 were treated with a single intraperitoneal dose of 7 $\mu g \cdot g^{-1}$ toxaphene, tagged and returned to the lake (Delorme 1995). No differences were seen in mean levels of hydroxyproline or calcium between treated and control fish from L260. A repeat analysis of trout bone collected in 1993 also showed lower, but not significantly different, bone collagen than Kusawa and very similar levels to those for trout from Fox Lake. Lake trout from Peter lake had higher bone collagen and lower toxaphene concentrations than the trout from Yukon Lakes. No correlation was observed between toxaphene levels and collagen or hydroxy-proline in 19 trout from Peter lake. Thus, bone collagen did not prove to be a strong indicator of toxaphene exposure in lake trout in contrast to results for fathead minnows in the laboratory. Mayer *et al.* (1977) found that adult fathead minnows and channel catfish exposed to 54–173 $ng \cdot L^{-1}$ and 72-630 $ng \cdot L^{-1}$ toxaphene respectively, had no decreases in hydroxyproline levels, but levels in exposed offspring were significantly decreased.

3.5.2.5 Metallothioneins in Arctic Marine Mammals, Fish and Birds

Metallothionein is a low-molecular weight (6.0–6.5 k Da), cytoplasmic protein that occurs naturally at background levels in most organisms. Outstanding features are its naturally high content of zinc, copper and cadmium (depending on availability), and an amino acid composition characterized by a high cysteinyl residue content. Its metal composition is determined primarily by physiological and environmental circumstances, and tissue levels can vary in response to stressors such as heavy metals (notably cadmium), diseases, stress, and some nonmetallic substances. The induction of metallothionein, primarily in response to copper, zinc and cadmium exposure, is a protective cellular response. When this protective capacity is exceeded, particularly for cadmium, toxicity could occur. Cadmium associated with this low-molecular protein is less toxic to organisms than when associated with higher-molecular weight cytosolic proteins. Metallothionein is a potential biomarker for cadmium toxicity and other stressors, and knowledge of its background concentration in animals and the amount of cadmium associated with it is required for an assessment of the adverse influence by cadmium on animals.

TABLE 3.5.2

Metallothionein levels ($\mu g \cdot g^{-1}$) in narwhal liver

	Repulse Bay	Iqaluit	Pond Inlet
N	7	10	9
Mean ± S.D.	418±396	394±140	1021±681
Range	164-1295	200-777	154-2314

Determination of metallothionein in narwhal and other marine mammals from the eastern Arctic was a high priority because of their very high concentrations of cadmium in the liver and kidney compared with terrestrial mammals. Arctic sea birds also have relatively high cadmium levels (Table 3.3.18). It has been shown that cadmium, zinc and copper induce production of metallothionein as a detoxification mechanism in both the liver and kidney of birds (Scheuhammer and Templeton 1990).

Metallothionein in narwhal

Liver tissue samples of narwhal from Pond Inlet, Repulse Bay and Iqaluit were analysed for metallothionein using published methods (Wagemann *et al.* 1984, Wagemann *et al.* 1993) (Table 3.5.2). The ages of the animals were not determined. The mean metallothionein concentrations in narwhal from Resolute Bay and Iqaluit were similar (404 μg·$^{-1}$g for the two groups combined) and considered to be normal. Individual values were mostly within one standard deviation of the mean. In narwhal from Pond Inlet the mean metallothionein concentration was higher (1021 μg·$^{-1}$g) and individual values ranged widely. The narwhal (Pond Inlet), which had the highest metallothionein concentration (2314 μg·$^{-1}$g), also had the highest cadmium concentration. In most other individuals there was no similar correspondence between high metal and high metallothionein concentration in liver cytosol, and vectors other than metals may have induced additional metallothionein synthesis in these individuals. Tissues were not examined histologically for evidence of disease. The distribution of cadmium in the liver between high and low molecular weight proteins was determined in one individual (Iqaluit group) with a normal concentration of metals and metallothionein in the liver (Figure 3.5.5). Liver cytosol was fractionated by high pressure liquid chromatography (HPLC) and fractions were analysed for metallothionein, cadmium, copper and zinc. Cadmium was predominantly associated with metallothionein in the liver of this individual and was considered to be nontoxic.

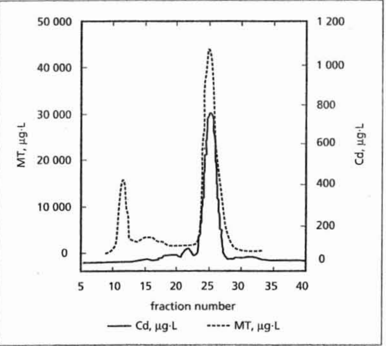


FIGURE 3.5.5

Metallothionen and cadmium in narwhal liver cytosol ($\mu g \cdot L^{-1}$) fractionated by HPLC.

Tissues from other animals in the eastern arctic (walrus, belugas, narwhal from Pond Inlet), which are more heavily loaded with cadmium were not analysed similarly, and it is not known whether or not some cadmium might "spill over" to higher molecular weight proteins in these animals.

Metallothionein in lake trout and arctic char

Liver, kidney, gill, and intestine tissues from lake trout and arctic char at three sites in the Arctic (Saqvaqjuac [63°N], Amituk Lake [75°N], and Hazen lake [82°N]) as well as in northwestern Ontario at the Experimental Lakes Area (ELA) were analysed for metallothionein (Table 3.5.3).

Metallothionein concentrations in liver, kidney and gill appeared to be latitudinally dependent, decreasing from south to north, with the exception of Hazen Lake on Ellesmere Island. Fish from this lake had higher concentrations of metallothionein than projected from the trend. This lake is underlain by coal deposits, and exposure of fish to PAHs found in sediments may have induced the higher metallothionein concentrations in these fish. The data have not been analysed for the influence of age and sex on metallothionein and trend indications are tentative.

Metallothioneins in sea birds

Metallothionein was measured in kidneys of adult sea birds using the silver saturation method

TABLE 3.5.3

Mean metallothionein concentration ($\mu g \cdot g^{-1}$) ± S.D. in lake trout and arctic char

Region	N	Kidney	Liver	Gill	Intestine
ELA (Lake trout)[1]	24	74.0 ± 15.5	475 ± 99	25.5 ± 4.5	—
Saqvaqjuac (Lake trout)	26	21.2 ± 3.4	315 ± 41	10.9 ± 1.1	15.5 ± 2.5
Resolute (Char)	39	8.9 ± 1.5	154 ± 18	7.3 ± 0.5	31.9 ± 3.0
Hazen Lake (Char)	45	25.0 ± 1.7	354 ± 26	—	75.0 ± 4.4

[1] *Fish metallothionein data was provided by J.F. Klaverkamp, DFO, Winnipeg.*

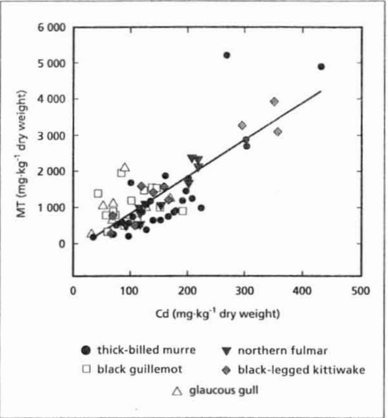


FIGURE 3.5.6

The relationship between Cd concentration and metallothionein (MT) in kidney of arctic sea birds.

(Scheuhammer and Cherian 1991). Regression analyses of metallothionein levels in kidney against cadmium, copper and zinc concentrations were run on data for 71 sea birds representing five species (Figure 3.5.6). The highest correlation was with cadmium ($r=0.81$, $p<0.001$) followed by zinc ($r=0.63$, $p<0.001$). The relationship between metallothionein and copper was not significant ($r=0.23$, $p>0.05$). Regression analyses of metallothionein levels against cadmium in kidney run on the individual species showed significant correlations for thick-billed murres ($r=0.84$, $p<0.001$), northern fulmars ($r=0.96$, $p<0.001$) and black-legged kittiwakes ($r=0.96$, $p<0.001$). There were no significant correlations found for black guillemots ($r=0.22$, $p>0.05$) and glaucous gulls ($r=0.44$, $p>0.05$). The black guillemots and glaucous gulls also had lower mean concentrations of cadmium in the kidney (103 $\mu g \cdot g^{-1}$ dry wt.; 87 $\mu g \cdot g^{-1}$ dry wt., respectively) than the murres, fulmars and kittiwakes (163 $\mu g \cdot g^{-1}$, 157 $\mu g \cdot g^{-1}$, 184 $\mu g \cdot g^{-1}$ respectively). Elliott *et al.* (1992b) also found marked differences in metallothionein levels among Atlantic sea bird species. Of the four species measured in that study, petrels, which are surface-feeders much like fulmars and kittiwakes, had the highest levels of both metallothionein and cadmium in the kidney, followed by puffins, then gulls.

TABLE 3.5.4

PAH-DNA adducts in tissues of male arctic beluga whales.

Location and sample number	Year	Tissue	Length (m)	Adducts (per 10^7 nucleotides)
Mackenzie Delta				
HI-11	1992	liver	4.38	2.4
HI-16-92	1992	liver	4.27	3.8
HI-19-92	1992	liver	4.51	3.8
HI-16-92	1992	brain	4.27	8.4
HI-18-94	1994	brain	4.06	1.2
Pangnirtung				
ARPG-27-84	1984	kidney	4.31	0.6
ARPG-18-84	1984	kidney	3.35	1.5

3.5.2.6 DNA Adducts in Marine Mammals

Covalent bonding of many chemicals or their metabolites to DNA can result in the formation of DNA adducts. Such adducts are believed to be important in mutagenesis and carcinogenesis and the measurement of DNA adducts is currently being used to assess exposure of humans to genotoxic compounds in the work place and general environment (Santella *et al.* 1993, Poirier 1994). DNA adducts have also been used to a limited extent to assess risks of exposure of wildlife to environmental genotoxins (Stein *et al.* 1994). The detection of DNA adducts commonly entails fairly detailed HPLC- fluorimetric, immunochemical or radioactive post-labeling procedures.

Initial studies of DNA adducts in beluga, carried out with the ^{32}P postlabelling procedure (Ray *et al.* 1991), showed similar levels of aromatic DNA adducts in liver of Mackenzie delta and St. Lawrence estuary beluga. This contrasted with evidence of much higher PAH levels in the sediments of the St. Lawrence estuary and Saguenay Fiord relative to the Mackenzie River delta. The ^{32}P technique is constrained by difficulties in resolving PAH adducts or otherwise distinguishing adducts of endogenous origin from those of contaminant origin. The HPLC-fluorimetric assay is specific for adducts produced by benzo(a)pyrene (a common environmental carcinogen) and is based on the measurement of fluorescent tetrols produced from B(a)P diol epoxide adducts upon hydrolysis of DNA (Shugart *et al.* 1983). Brain and liver tissues of beluga from the Gulf of St. Lawrence have been reported to contain specific B(a)P adducts while these adducts were undetectable in Mackenzie delta beluga brain (Martineau *et al.* 1994, Shugart 1990). Recent work by Mathieu *et al.* (1995) using HPLC-fluorimetric procedures, has not confirmed the original findings of Shugart (1990) for specific B(a)P epoxides in liver and brain tissues in St. Lawrence beluga. Mathieu *et al.* (1995), detected "uncharacterized" PAH-DNA adducts in brain, liver and kidney of arctic beluga using a radioimmunoassay technique specific to PAH-DNA adducts (Table 3.5.4). This is the first such evidence for PAH-related adducts in arctic wildlife in general and marine mammals in particular.

3.5.3 Population Level Effects and Comparison with Other Stressed Populations

3.5.3.1 Peregrine Falcon Population at Rankin Inlet, NWT

Perhaps because they were not exposed to such high levels of contaminants year round, populations of tundra peregrines did not suffer the extreme declines seen in southern *anatum* populations (Ratcliffe 1967, Peakall 1976, Newton 1979). However, by the 1970s, it is estimated that *tundrius* populations had declined to only 35% of their numbers prior to the introduction of DDT (Kiff 1988, Schempf 1989). Thomas *et al.* (1992) concluded that total DDT levels in *F. p. tundrius* had peaked in the mid-seventies but decreased by the early 1980s. Subsequently, Court *et al.* (1990) concluded that while remaining a productive population, significant amounts of OC pesticides and PCB residues were still being accumulated. Eggshell thinning, levels of DDE residues in eggs, and the levels of contaminants found in prey items were all close to the critical levels considered to result in decreased production in this species (Court *et al.* 1990, Peakall and Kiff 1988, Peakall *et al.* 1990). Court *et al.* (1990) concluded that about 10% of all breeding attempts between 1981 and 1985 failed due to pesticide contamination.

Peregrine eggshell fragments collected from Rankin Inlet between 1991 and 1994 were 15% thinner (mean 0.306 ± 0.022 mm; n = 54; Johnstone *et al.*, in press) than the average pre-DDT shell thickness for peregrines from the Nearctic (0.360 mm, Berger *et al.* 1970, Anderson and Hickey 1972, Walker *et al.* 1973). Of the 54 clutches sampled, 28% showed thinning equal to or greater than critical levels associated with reproductive failure and population decline in this species (i.e. 17% of average pre-DDT shell thickness) (Peakall and Kiff 1988). There was no difference in mean eggshell thickness between samples collected in the period 1990-1994 (0.306 ± 0.022 Johnstone *et al.* in press) and that collected by Court *et al.* (1990) between 1982-1986 (mean 0.303 ± 0.023 mm, n= 62; Students t-test $t^{114}= -1.018$ $p= 0.3107$). Contrary to predictions, there has not been any improvement in eggshell quality for this population of tundra peregrines since the early 1980s. It has been concluded that eggs with DDE residues of 15 to 20 µg·g^{-1} would experience reproductive failure (Peakall *et al.* 1975). Two clutches (10%) in the 1991 sample contained eggs with residue levels exceeding critical levels.

PCBs and *p,p'*-DDE were found in the blood plasma of nestlings sampled at about 20 days of age (R. Johnstone, Univ. of Saskatchewan, unpublished data). Mean PCB levels (as Aroclor 1254:1260) in nestlings were about one-half of the level in adult males and almost one-tenth the levels found in adult females. It can be argued that this represents an inheritance of contaminants originally present in the egg, since elevated residue levels were found in tissue samples from six peregrine nestlings that were found dead in the nests. Levels of 59 µg·g^{-1} PCBs and 19 µg·g^{-1} *p,p'*- DDE were found in the liver of a 25-day-old chick, which suggests that nestlings may accumulate significant quantities of contaminants from the prey species fed to them at the nest.

TABLE 3.5.5

Mean (geometric) organochlorine residues (µg·g^{-1} wet wt., range in parentheses) in whole bodies of prey species of Peregrine Falcons nesting at Rankin Inlet, NWT (Johnstone *et al.* in press).

Species	N	PCB	*p,p'*-DDE	Dieldrin
Oldsquaw *Clangula hyemalis*	5	6.9 (2.9-19)	1.1 (0.61-3.8)	0.51 (0.02-4.0)
Black Guillemot *Cepphus grylle*	8	0.38 (0.13-0.65)	0.17 (0.10-0.24)	0.004 (<0.01-0.02)
Pintail *(Anas acuta)*	4	2.1 (<0.01-5.4)	0.47 (0.09-0.91)	0.09 (0.03-0.26)
Semipalmated Plover *Charadius semipalmatus*	5	0.40 (<0.01-2.1)	0.50 (0.10-1.1)	0.02 (<0.01-0.11)
Semipalmated Sandpiper *Calidris pusilla*	3	0.03 (<0.01-0.10)	0.25 (0.04-0.70)	<0.01 —
Water Pipit *Anthus spinoletta*	5	0.14 (<0.01-0.79)	0.69 (0.004-9.5)	<0.01
Horned Lark *Eremophila alpestris*	7	<0.01	0.07 (<0.01-0.17)	<0.01 —
Lapland Longspur *Calcarius lapponicus*	10	0.03 (<0.01-0.36)	<0.01 —	<0.01 —
Snow Bunting *Plectrophenax nivalis*	2	<0.01 —	<0.01 —	<0.01 —
Arctic Ground Squirrels *Spermophilus parryi*	7	<0.01	<0.01	<0.01
Collared lemmings *Dicrostonyx groenlandicus*	13	0.06[1] (<0.01- 4.2)	0.01[1] (<0.01- 0.11)	<0.01 —

[1] *Contaminants were only found in one collared lemming that had levels of ΣPCB 4.17, and DDE 0.11*

In Rankin Inlet the larger prey species include oldsquaw, black guillemot, and pintail (Bradley and Oliphant 1991), all species that are relatively high in contaminants (Table 3.5.5). These species, especially oldsquaw, are also likely to have eggs with elevated levels of contaminants, although this was not determined.

Baril *et al.* (1990) concluded that, in prey species of the peregrine falcon, PCBs, DDE and dieldrin were at levels high enough to be of concern. This assessment was based on established toxicological effects from the literature for various species (see Baril *et al.* 1990 for a more thorough discussion). Levels of 1 $\mu g \cdot g^{-1}$ DDE, 5 $\mu g \cdot g^{-1}$ PCBs and 0.1 $\mu g \cdot g^{-1}$ of dieldrin were suggested as threshold levels. OCs in some avian prey collected at Rankin Inlet exceeded these levels (Table 3.5.5). The maximum detected levels in oldsquaw and northern pintail exceeded the suggested threshold level for PCBs, while oldsquaw, water pipit and semi-palmated plover exceeded the DDT threshold level and dieldrin in oldsquaw, northern pintail and semi-palmated plovers exceeded the 0.1 $\mu g \cdot g^{-1}$ level. Water pipit had the highest recorded residue levels of *p,p'*-DDE of all prey species of 9.5 $\mu g \cdot g^{-1}$. There was little evidence of a decline in concentrations in prey species from those found by Court et al. (1990). For example, water pipit had the same *p,p'*-DDE levels as those recorded in the early 1980s (Court *et al.* 1990).

Mammalian species represent a source of relatively clean prey for peregrines at Rankin Inlet (Table 3.5.5). Up to one-third of the total biomass of prey used are mammalian species, namely ground squirrels and microtine rodents (Bradley and Oliphant 1991). These are non-migratory species, so any contamination in their tissues represents contamination of the immediate environment around Rankin Inlet. That mammals represent a relatively "clean" food source is also reflected in residue levels in the blood plasma of rough-legged hawk (*Buteo lagopus*) nestlings. This species primarily utilizes mammalian prey. OC contaminants were below detection limits in the blood plasma of seven rough-legged hawk nestlings.

Although *p,p'*-DDE levels appear to be decreasing in the population of *F. p. tundrius* nesting around Rankin Inlet (see Section 3.4.3.2) the changes are small, as there is no overall improvement in the average thickness of eggshells produced by birds in this population. Levels of PCBs show little change in the same time period. Also, eggshell thickness, residue levels in peregrine eggs, blood plasma, tissue samples, and prey species are within the range that will result in failure of a portion of reproductive attempts each year.

3.5.4 Assessment of Biological Effects Data

There have been major advances in knowledge of specific biomarkers in arctic biota over the past few years. Biomarkers measure chemical changes at the cellular level but are not readily interpreted ecologically.

While studies of ecological relevance, such as species diversity or population size, are ongoing in the Arctic they are not related specifically to the effects of contaminants. With the possible exception of peregrine falcons it is not possible to link contaminant levels or biochemical indicators of effects to effects at the individual or population levels. At the individual level, detailed pathological examinations of the type carried out on St. Lawrence beluga whales (Martineau *et al.* 1994) or on Baltic ringed seal (Bergman and Olsson 1986), which have attempted to link certain pathologies to contaminants, have not been conducted on Canadian Arctic species. Studies of Baltic grey seals have revealed a disease complex including skull bone lesions, thought to be caused by environmental pollutants (Mortensen *et al.* 1992). This bone loss may be indicative of endocrine imbalance caused by hyperadrenocorticism associated with high levels of PCBs and DDT (Bergman *et al.* 1992). Martineau *et al.* (1994) have identified a high frequency of malignant neoplasms along with the presence of benzo(a)pyrene adducts in necropsies of dead St. Lawrence beluga whales and suggest this is related to PAH exposure. These authors have also developed preliminary evidence of a link for immune system dysfunction in beluga (relative to Hudson Bay animals as controls) which may be related to high PCB exposure. Concentrations of organochlorine contaminants in arctic beluga such as PCBs are 10- to 20-fold lower than in St. Lawrence beluga and similar, or even greater differences, are observed between arctic ringed seals and those inhabiting the Baltic Sea (Blomkvist *et al.* 1992). Concentrations of PCBs associated with poor reproductive success of captive harbour seals (~ 25 $\mu g \cdot g^{-1}$ in blood lipids) (Boon *et al.* 1987) are also about 10-to 20-fold higher than concentrations in blubber lipids of ringed seals. Thus, as far as organochlorines are concerned, Arctic marine mammals are regarded as controls for much more contaminated members of their populations or related species in temperate regions.

Although body burdens of PCBs in arctic seals and cetaceans are much lower than in highly contaminated populations, the threshold for effects of the contaminants is not well known. Immunotoxic effects are thought to be one of the most sensitive and environmentally relevant effects of planar halogenated aromatic compounds such as PCBs and PCDD/Fs (Vos and Luster 1989). Harbour seals fed Baltic Sea herring highly contaminated with PCBs and other organochlorines had impaired natural killer (NK) cell activity, *in vitro* T-lymphocyte function, antigen-specific *in vitro* lymphocyte proliferative responses and *in vivo* delayed-type hypersensitivity (DTH) responses compared with seals fed relatively uncontaminated Atlantic Ocean herring (Ross *et al.* 1996, Ross *et al.* 1995, De Swart *et al.* 1995, De Swart *et al.* 1994). Concentrations of TCDD TEQs associated with impaired immune function (depressed leucocyte activity) in harbour seals (mean of 204 ng TCDD TEQ·kg^{-1} mainly due to mono- and di-ortho PCBs) (Ross *et al.* 1995) are three to five times greater than in arctic ringed seal or beluga blubber (Ford *et al.* 1993), but similar to observations for walrus blubber from northern Quebec (Muir *et al.* 1995c).

The impairment of immune function is thought to have played a role in the morbillivirus-induced mass mortalities of harbour seals, grey seals and striped dolphin populations in Europe in the 1987-1991 period (Hall *et al.* 1992, Aguilar *et al.* 1994). Lymphocyte proliferation was highly negatively correlated to PCB and DDT concentrations in blood in bottlenose dolphins, which also experienced morbillivirus mortality in North America (Lahvis *et al.* 1995). Good progress has been made in characterizing the immune system of beluga and its response to exposure to organochlorines (De Guise *et al.* 1995, Béland *et al.* 1995). Comparison of responses of the highly contaminated St. Lawrence River population with those in arctic beluga will provide information about the immunocompetence of both populations.

The species with the most significant risk of exposure to PCBs and OC pesticides may be the polar bear which, based on comparison with EROD activity in other marine mammals, appears to have elevated CYP1A-mediated activity. The strong positive correlation of CYP1A with non-ortho and mono-ortho PCBs in polar bear liver suggests that these compounds may be responsible for the activity. There were also indications that elevated CYP2B activity in polar bear was due to the induction by chlordane components (mainly oxychlordane and nonachlor) and ortho substituted PCBs. Similar positive correlations of EROD activity with non-ortho PCB levels were observed in beluga whales that were starving due to ice entrapment. This leads to the conclusion that arctic animals with relatively low levels of contaminants compared with temperate species may be vulnerable if they have to mobilize lipid deposits during fasting or starvation. Polar bears in Hudson Bay, which lose up to 80% of their adipose tissue while fasting for four to five months on land until the Bay re-freezes may be particularly at risk. Concentrations of PCBs and chlordane in female polar bears during their eight-month long fast have been found to increase by approximately a factor of 2, resulting in high concentrations in polar bear milk (Polischuk *et al.* 1995, 1994). Mean concentrations of TCDD TEQs in polar bear liver on a lipid weight basis are two times higher (Letcher *et al.* 1996) than that in blubber of harbour seals showing immunosuppression (Ross *et al.* 1995). The high concentrations of TEQs in polar bears, and the highly induced CYP1A and CYP2B hepatic enzymes in some individuals suggest the potential for immunosuppression in polar bears.

Females and their offspring may be most vulnerable during mobilization of these lipophilic contaminants for lactation. Lactation provides a mechanism whereby large quantities of organochlorines can be transferred from one generation to the next and the transfer of OC contaminants in milk from mother to young occurs at a crucial point in the growth and development of the young. Overall, the MFO enzyme data for polar bear and beluga suggest that even the relatively low levels of contaminants present in arctic animals may not be without biological effects, especially during years of poor feeding.

Other studies on fish and ringed seals have not shown the strong correlations between MFO enzyme activity and PCB levels, nor have they shown the presence of effects related to toxaphene exposure. As is usually the case with arctic animals, the lack of experimental dosage/response data continues to limit the ability to interpret concentrations observed in the animals.

Although body burdens of PCBs in arctic seals and cetaceans are much lower than in highly contaminated populations the threshold for effects of the contaminants is not well known. Immunotoxic effects are thought to be one of the most sensitive and environmentally relevant effects of planar halogenated aromatic compounds such as PCBs and PCDD/Fs (Vos and Luster 1989). Concentrations of TCDD TEQs associated with impaired immune function (depressed leucocyte activity) in harbour seals (mean of 204 ng TCDD TEQ·kg^{-1} mainly due to mono- and di-ortho PCBs) (Ross *et al.* 1995) are three to five times greater than in arctic ringed seal or beluga blubber (Ford *et al.* 1993) but similar to observations for walrus blubber from northern Québec (Muir *et al.* 1995c). The impairment of immune function is thought to have played a role in the morbillivirus-induced mass mortalities of harbour seals in the North Sea in 1988 (Ross *et al.* 1995).

In the case of heavy metals and radionuclides, arctic animals have relatively high body burdens compared to similar or related species in temperate regions. But they also may be adapted to relatively high exposure. Macdonald *et al.* (1996) concluded that caribou receive relatively high doses of natural radiation (mainly to ^{210}Po) relative to other mammals in the Canadian environment. The animals have presumably adapted to the radiation stress from these natural sources but potential effects of the high doses on the animals are not clear. Cadmium levels are particularly elevated in narwhal and beluga kidney and liver, relative to terrestrial mammals. Cadmium was found to be predominantly associated with metallothionein in narwhal liver, suggesting it was in a relatively nontoxic form. The strong correlations of metallothionein in sea birds and marine mammals show that they are responding to heavy metal exposure. But it is not clear what, if any, negative consequences are resulting from this exposure.

The tundra peregrines represent one of the only cases in the Arctic where population size, biochemical effects (egg shell thinning) and chemical concentrations (PCB, DDE) can be linked. It has been concluded that eggs with DDE residues of 15 to 20 $\mu g \cdot g^{-1}$ would experience reproductive failure (Peakall *et al.* 1975). While no eggs exceeded this level, two clutches (10%) in the 1991 sample contained eggs with residue levels exceeding the "critical" level of 5 $\mu g \cdot g^{-1}$. This population was estimated to have declined to only 35% of their numbers prior to the introduction of DDT of tundra peregrines by the 1970s. Although the peregrine is assumed to be exposed to elevated DDT in tropical countries during overwintering results show significant exposure of adult and hatchlings because of feeding by peregrines on fish-eating birds.

The information available on concentrations of PCBs and other persistent organochlorines in fish can also be used to assess risks of exposure of fish-eating birds and mammals. The draft Canadian Environmental Quality Guidelines (EQG) for PCBs for protection of animals that consume aquatic biota is 7.6 $ng \cdot g^{-1}$ for freshwater wildlife and 6.0 $ng \cdot g^{-1}$ for estuarine and marine wildlife (Environment Canada 1996). The draft Canadian EQG for DDE is 36.6 $ng \cdot g^{-1}$, while a guideline of 1.1 $ng \cdot kg^{-1}$ for PCDD/Fs has been proposed (Environment Canada 1996). Concentrations of PCBs and DDE in muscle of non-piscivorous arctic fish (e.g. broad and lake whitefish, char, arctic cod) approach and in a few cases exceed these guidelines (See Tables 3.3.1 and 3.3.2). If nPCBs are included in TCDD TEQs, most arctic fish would exceed the Canadian WQG for TCDD (Table 3.3.3). The United States' Environmental Protection Act (US EPA) guideline values for assessment of hazards to fish-eating wildlife are in some cases higher than for Canada (US EPA 1995): 160 $ng \cdot g^{-1}$ for PCBs, 39 $ng \cdot g^{-1}$ for ΣDDT, 57 $ng \cdot g^{-1}$ for mercury, and 0.5 $ng \cdot kg^{-1}$ for TCDD, in fish flesh (US EPA 1995). Concentrations in muscle of most non-piscivorous Arctic freshwater and marine fish sampled (see Table 3.3.1, 3.3.2 and 3.3.12) are lower than these levels by a factor of at least 2- to greater than 10-fold. But the US EPA wildlife criteria for 2,3,7,8-TCDD of 0.5 $ng \cdot kg^{-1}$ is exceeded in most fish if TCDD TEQs based on nPCBs are used (see Table 3.3.3), although the data are limited. Both the US EPA and Canadian EQG values are based on an evaluation of all available toxicological data related to establishing a No Observable Adverse Effects level concentration in the most sensitive species.

In summary, an evaluation using criteria established to protect fish-eating wildlife suggests that there is not a large margin of safety for Arctic marine or freshwater piscivores especially if TCDD TEQs are used for the assessment. Using these criteria, carnivores such as polar bears would be at risk due to consumption of ringed seal tissues.

- *With the possible exception of peregrine falcons, contaminant levels or biochemical indicators of effects have not been linked to effects on arctic animals at the individual or population level.*
- *The species with the most significant risk of exposure to PCBs and OC pesticides may be the polar bear which, based on comparison with EROD activity in other marine mammals, appears to have elevated CYP1A-mediated activity. The strong positive correlation of CYP1A with non-ortho and mono-ortho PCBs in polar bear liver suggests that these compounds may be responsible for the activity.*
- *Similar positive correlations of EROD activity with non-ortho PCBs levels were observed in beluga that were starving due to ice entrapment. Arctic animals with relatively low levels of contaminants compared with temperate species may be vulnerable if they have to mobilize lipid deposits during fasting or starvation.*
- *Concentrations of TCDD TEQs in arctic ringed seal or beluga blubber are three to five times lower than those associated with impaired immune function (depressed leucocyte activity) in harbour seals (mean of 204 ng TCDD* $TEQ \cdot kg^{-1}$*).*
- *Concentrations of organochlorine contaminants in arctic beluga such as PCBs are 10- to 20-fold lower than in St. Lawrence beluga where preliminary evidence of a link for immune system dysfunction due to high PCB exposure (relative to Hudson Bay animals as controls) has been developed.*
- *Concentrations of PCBs associated with poor reproductive success of captive harbour seals (~ 25* $\mu g \cdot g^{-1}$ *in blood lipids) (Boon* et al. *1987) are also about 10- to 20-fold higher than concentrations in blubber lipids of ringed seals.*
- *Females and their offspring may be most vulnerable during mobilization of these lipophilic contaminants for lactation because it occurs at a crucial point in the growth and development of the young. Overall, the MFO enzyme data for polar bear and beluga suggest that even the relatively low levels of contaminants present in arctic animals may not be without biological effects, especially during years of poor feeding.*

- *Other studies on fish and ringed seals have not shown the strong correlations between MFO enzyme activity and PCB levels, nor have they shown the presence of effects related to toxaphene exposure. As is usually the case with arctic animals, the lack of experimental dosage/response data continues to limit the ability to interpret concentrations observed in the animals.*
- *Arctic animals have relatively high body burdens of heavy metals and radionuclides compared to similar or related species in temperate regions. But they also may be adapted to relatively high exposure because of the importance of natural sources of these contaminants.*
- *Potential effects of the high doses of metals such as cadmium on caribou, beluga and narwhal are not clear. Cadmium was found to be predominantly associated with metallothionein in narwhal liver, suggesting it was in a relatively nontoxic form. The strong correlations of metallothionein in sea birds and marine mammals show that they are responding to heavy metal exposure.*
- *An evaluation using criteria established to protect fish-eating wildlife suggests that there is not a large margin of safety for Arctic marine or freshwater piscivores, especially if TCDD TEQs are used for the assessment. Using these criteria, carnivores such as polar bears would be at risk due to consumption of ringed seal tissues.*

3.6 Knowledge Gaps

The state of knowledge of contaminants in Canadian Arctic biota has advanced enormously since the publication of the first major reviews in *The Science of the Total Environment* in 1992 (Thomas *et al.* 1992, Lockhart *et al.* 1992, Muir *et al.* 1992a). The most significant gains are new knowledge of spatial trends of organochlorines and heavy metal contaminants in terrestrial animals, such as caribou and mink, and in waterfowl, where no information was previously available. Spatial trends in freshwater fish have been broadened, especially in the Yukon, where contaminant measurements of, for example, OCs were previously nonexistent. In the marine environment, the circumpolar survey of OCs in polar bear fat and the broadening of geographic coverage of heavy metals in ringed seals are major advances in knowledge.

3.6.1 Priority Chemicals and Their Sources

The list of priority substances monitored in biota in the Canadian Arctic the past five years is relatively long, about 100 OCs (including PCB congeners, isomers of HCH, numerous components of technical chlordane and DDT) and 25 metals in many samples. There are still a number of chemical contaminant groups for which information is quite limited. These groups include PCDD/Fs, as well as non-ortho PCBs, which have not been included in most surveys due to high costs. Non-ortho PCBs were found in all samples that were analysed (fish and marine mammals). The nPCBs, along with mono-ortho PCBs, accounted for most of the TCDD TEQs and therefore deserve priority for future monitoring. No measurements have been made of many other planar OCs such as chlorinated naphthalenes, chlorinated diphenyl ethers or their brominated analogs in Arctic marine mammals or fish although measurements in Svalbard have shown these contaminants are present in arctic biota.

Toxaphene is a major OC contaminant in arctic air, seawater, fish and marine mammals. Yet there is no information available on toxaphene levels in terrestrial animals and in waterfowl and sea birds. Current methods of quantifying toxaphene may overestimate levels in some species, such as marine mammals. There is a need to broaden the dataset for toxaphene because of its predominance in the abiotic environment and importance in risk assessment for human dietary exposure.

Perhaps the largest group of potential arctic contaminants that has not received attention, in terms of monitoring in biota, is the current use pesticides. There is recent evidence that this diverse group of compounds — which includes less persistent chlorinated organics such as endosulfan, methoxychlor, pentachlorophenol, as well as other weed and insect control products widely used in North America such as trifluralin, atrazine, chlorpyrifos and chlorthalonil — are present in arctic air and snow (see Chapter 2) but measurements of these compounds in biota are very limited. These compounds were not considered a priority for monitoring in biota because they do not biomagnify in food webs and therefore do not threaten higher predators directly. However, they may be accumulated by plants and phytoplankton at the base of the food web (France *et al.* 1996, Chernyak *et al.* 1996). Some of these compounds (e.g. atrazine) are potent inhibitors of photosynthesis while others (e.g. methoxychlor) are known to have estrogenic activity. Degradation in soils and revolatilization from surfaces, which are important removal processes for current use pesticides in temperate climates, may be less rapid in the Arctic environment. Unlike the persistent OCs, which are declining in use worldwide and are subject to bans in most countries bordering on the Arctic Ocean, current use pesticides continue to be used in increasing amounts in agriculture and for controlling biting flies.

While a wide range of metals have been determined in arctic biota samples, there has been little work on speciation and on isotope ratios of these metals in biota. An exception is the determination of methylmercury and of metallothionein or bound cadmium fractions. Major questions that need to be answered concern the relative contributions of metals, from natural geological sources and pollution sources, and the biological implications of those different sources. Where isotope ratios can be determined (e.g. for lead and other metals), this information can be beneficial in determining sources of metals to the food web.

- *Although the list of priority substances monitored in biota in the Canadian Arctic over the past five years is relatively long (including PCB congeners, isomers of HCH, numerous components of technical chlordane and DDT and 25 metals in many samples) there are still a number of chemical contaminant groups for which information is quite limited or nonexistent. These groups include PCDD/Fs and non-ortho PCBs, chlorinated naphthalenes, chlorinated diphenyl ethers or their brominated analogs (all of which are cytochrome P4501A1 enzyme inducers).*

- *Toxaphene is a major OC contaminant in arctic air, seawater, fishes and marine mammals. Surprisingly, there is no information available on toxaphene in terrestrial animals and in waterfowl and sea birds. Current methods of quantifying toxaphene may overestimate levels in some species such as marine mammals.*
- *Current use pesticides (a diverse group of less persistent organics) have not received attention, in terms of monitoring in biota, although recent work indicates they are present in arctic air and snow and terrestrial plants.*

3.6.2 Gaps in Spatial and Temporal Information

3.6.2.1 Modelling

Knowledge of the regional and circumpolar distribution, as well as temporal trends in concentrations of OCs and heavy metals is important in determining the sources and potential significance of these contaminants to Arctic marine and maritime wildlife and the humans that consume them. Although progress has been made, the state of knowledge of the dynamics of these chemicals in the Arctic environment is inadequate for predictions of future trends in bioaccumulation from water or air to biologically significant levels in species at the top of the food web, including humans. In regions such as the Great Lakes, the North Sea and the Baltic, contaminant monitoring data have been used to calibrate and validate computer models of distribution and food chain bioaccumulation of priority pollutants (e.g. Gobas 1993). The goal of these models is to predict temporal and, in some cases, spatial trends of contaminants in water, sediments and food webs. No comparable modelling has been done in the Canadian Arctic although models have been used to examine input/output of HCH in the Arctic Ocean (Chapter 2), fate and bioaccumulation of OCs in lakes (Diamond 1994) and accumulation with age in beluga and seals (Hickie 1996). The lack of integration of chemical measurements with the development and validation of chemical fate and bioaccumulation models presents a major gap in the present program. A future goal should be to use the existing database, along with further studies, to calibrate models and to link chemical fate/distribution model output to contaminant bioaccumulation. Examples are:

1. the prediction of mercury and OC levels and trends in marine and freshwater fish based on water concentrations, food chain characteristics and atmospheric inputs
2. prediction of spatial and temporal trends in contaminant levels in marine mammals including important tissues such as liver/kidney
3. linkage of food chain models for freshwater and marine biota to models of human dietary exposure and biological effects in top predators.

3.6.2.2 Temporal Trends

The lack of temporal trend information for most contaminants is perhaps the most significant knowledge gap at the present time. Temporal trend data are very limited for most OCs and metals because they are based on only two or three sampling intervals. Long-term (>10 year) temporal trend data are only available for PCBs and DDT-related compounds and comparisons are limited because of changes in methodology. By comparison, temporal trend data for contaminants in Lake Ontario lake trout and in various species from the Baltic Sea and from Lake Storinveld in northern Sweden are available yearly for a 15- to 20-year period. The Lake Ontario and Swedish studies have been possible in part because tissue banks enabled a retrospective analysis of samples using current methodology. There is clearly a need for well-designed temporal trend studies utilizing tissues from tissue banks created by the Northern Contaminants Program at selected, well-characterized locations. The creation of tissue banks by the Northern Contaminants Program has made possible future retrospective analysis beginning with samples from the 1980s and earlier in some cases.

Temporal trend information for OCs and metals is particularly limited for the terrestrial and freshwater environment. No temporal trend information for OCs or metals is available for caribou or mink while reliable trends of OCs in fish are available for only species at one site (burbot liver at Fort Good Hope). There is limited data on temporal trends in mercury in freshwater and anadromous fish in the NWT which indicates no increase in mercury over a 20-year period. However, this conclusion is based on results from walleye in one lake known for high natural sources of mercury (Lac Ste Therese), and on very limited numbers of arctic char from a large number of lakes in the Keewatin/Baffin region. The results are not in agreement with observations for marine mammals or trends in lake sediments.

The temporal trend information in the case of mercury is particularly important because of the observation that levels are increasing in beluga and ringed seal. The increase is very significant — 1.5- to 2.5-fold — for both mean concentrations and rates of accumulation, in both species in the western Arctic. High mercury concentrations are also seen in freshwater fish, especially lake trout. While this information is not new, the possibility that concentrations are increasing puts the issue in a new light. Whether this increase is entirely due to anthropogenic inputs, which have been shown to be increasing slowly all this century in dated sediment cores, or whether it is due to some other environmental change which is mobilizing mercury, is not known. But it clearly is a major knowledge gap that deserves high priority for further investigation.

- *While a wide range of metals have been determined in arctic biota samples there has been little work on speciation and isotope ratios of these metals in biota. Where isotope ratios can be determined (e.g. for lead), this information can be beneficial in determining sources of metals to the food web.*
- *The lack of integration of chemical measurements with the development and validation of chemical fate and bioaccumulation models presents a major gap in the present program. A future goal should be to use the existing database, along with further studies, to calibrate models and to link chemical fate/distribution model output to contaminant bioaccumulation.*
- *The lack of temporal trend information for most contaminants is perhaps the most significant knowledge gap at the present time. Temporal trend data are very limited in the Canadian Arctic for most OCs and metals because they are based on two or at most three sampling times and because changes in methodology have made it difficult to compare recent and older data.*
- *The need for temporal trend information in the case of mercury is particularly important because of the observation that levels are increasing significantly in beluga and ringed seal. Whether this increase is entirely due to anthropogenic inputs, which have been shown to be increasing slowly all this century in dated sediment cores, or is due to some other environmental change which is mobilizing mercury, is not known. But it clearly is a major knowledge gap that deserves high priority for further investigation.*

3.6.2.3 Spatial Trends and Geographic Coverage of Various Species/Tissues

The geographical coverage of contaminant measurements in fish is very good in the Yukon, but is much less detailed in the NWT and northern Québec. In the NWT, most measurements have been carried out in fish muscle samples (or burbot liver) from Great Slave Lake and nearby smaller lakes, as well as in the Mackenzie River and delta. Contaminant analysis of freshwater fish in the Nunavut region of the NWT and the Nunavik area of northern Québec is limited mainly to arctic char and is insufficient to assess spatial trends. Further sampling of other subsistence fish in Nunavut and Nunavik should be a priority in the future.

The high lake to lake variability with a species (e.g. lake trout and burbot) of both mercury and persistent OCs has not yet been adequately explained. For example, toxaphene concentrations in Great Slave Lake burbot liver were substantially lower, averaging 244 ng·g^{-1} for the Slave River Delta and 762 ng·g^{-1} for East Arm burbot, than the 2,820 ng·g^{-1} observed for Lake Laberge, although burbot appear to be piscivorous in both lakes (based on stable isotopes) and fluxes of toxaphene to lake sediments are similar. Similar considerations apply to mercury in lake trout in the NWT lakes. It is difficult to assess whether these inter-lake differences in contaminant concentration reflect true differences between the systems or are more a reflection of the inherent variability in such measurements. The resolution of this is important because the concentrations of organic contaminants in fish appear to be a function not only of trophic level but of other aspects of the lake ecosystem.

Geographic coverage of major species such as caribou and mink (within the NWT), as well as waterfowl and game birds, is quite complete in terms of measurements of OCs, radionuclides and metals. Contaminant data are available for a greater number of terrestrial mammals in the Yukon compared with the NWT or Northern Québec. Very limited data is available on OCs in mammals in northern Québec.

Geographic coverage of contaminant measurements in marine mammal and sea bird populations is very good. All major beluga, ringed seal and polar bear stocks along with several major sea bird colonies have been sampled for OC and heavy metal contaminants within the past six years. Studies on contaminants in walrus are limited to Foxe Basin and northern Québec stocks, while migratory harp seals have only been studied recently at one location. Contaminant measurements in bearded seal, harbour seal, bowhead whale and killer whale tissues from the Canadian Arctic are very limited or non-existent.

While concentrations of most OCs are quite similar, spatially, in most species in the Canadian Arctic an exception is the polar bears from Region 3 (M'Clure Strait). This population showed elevated levels in samples collected in 1990-91 and in 1982-84. These bears may be influenced by a different food chain structure in a permanent ice environment, or by some as yet unknown feature of atmospheric or oceanic transport. There is anecdotal information that bearded seals may be more abundant in this area, and there is the possibility that the base of the ringed seal food chain is more epontic than pelagic compared with areas further east (Welch *et al.* 1992). Polar bears from M'Clure Strait also had exceptionally high levels of mercury in liver compared with polar bears from nearby regions. Unfortunately there is no seal or lower food web contaminant data from this area for corroboration.

There is much less information on contaminant levels and very limited geographic coverage for marine fish and invertebrates. Contaminant data from the marine food web is important because, combined with information of trophic levels of each organism, it leads to an understanding of the pathways of bioaccumulation of the contaminants. The marine food web data is also important because some animals such as mussels and amphipods may be harvested by Northerners or may be fished commercially (turbot, prawns).

At some locations in the Canadian Arctic there is evidence of PCB contamination of terrestrial plants, soils and nearshore sediments and biota due to pollution from military radar facilities and other activity. The detailed study of PCBs in nearshore areas of Cambridge Bay illustrates how PCBs can be transported from dump sites to the immediate marine environment. While these studies also show that the PCB contamination is quite localized, when considered on a broad regional scale, there is a need to determine whether marine mammals frequenting the waters within the general area of these sites as well as terrestrial mammals, such a caribou and arctic fox feeding within the impacted zones, have elevated PCB and lead contamination. No specific studies on large mammals have been carried out to examine this question.

With the completion of work on beluga muktuk, information is now available on levels of metals for most marine mammal tissues that may be used in traditional native diets. Additional studies of metal contaminants in muktuk need to be made in eastern Canadian marine mammal populations where fewer samples have been analysed. Information is more limited on OC levels in muktuk and other tissues of marine mammals and additional measurements should be made. In the case of muktuk, further studies should take into account any variations, between individuals or communities, in methods of preparing muktuk, which might affect fat content of this food. Levels clearly vary with fat content of muktuk, thus OC concentrations in blubber may be used to estimate concentrations in muktuk with good accuracy. Whether OC levels in other marine mammal tissues can be estimated from blubber levels has not been throughly investigated.

There now exists a relatively large dataset on chlorinated dioxins and -furans in arctic biota (fish from the Slave River, Great Slave Lake and Yukon lakes, caribou, ringed seals and polar bears). TCDD levels are low (typically <1 pg·g^{-1}). Fewer measurements have been made of the toxic non-ortho substituted PCBs in arctic fish, but in general concentrations are an order of magnitude higher than those of PCDD/Fs. Where nPCBs have been measured along with PCDD/Fs, calculation of TCDD TEQs shows that nPCBs, especially CB126 account for most of the TEQs. Future measurements of toxic planar OCs can therefore be limited to nPCBs unless specific sources of PCDD/Fs (such as waste PCB oils or pentachlorophenol use) are suspected.

Additional measurements of PCDD/Fs can be justified in the case of seals, where PCDD/Fs account for up to 50% of TCDD TEQs (Ford *et al.* 1993). Initial surveys of ringed seals and polar bears, which showed higher PCDD/Fs in the high Arctic (Norstrom *et al.* 1990), have not been followed up.

- *Contaminant analysis of freshwater fishes in the Nunavut region of NWT and the Nunavik area of northern Québec is limited mainly to arctic char and is insufficient to assess spatial trends. Further sampling of other subsistence fishes in Nunavut and Nunavik should be a priority in the future.*
- *The high lake-to-lake variability with a fish species (e.g. lake trout and burbot) of both mercury and persistent OCs has not yet been adequately explained.*
- *Contaminant data are available for a greater number of terrestrial mammals in the Yukon compared with NWT or northern Québec. Very limited data is available on OCs in mammals in northern Québec.*
- *Studies on contaminants in walrus are limited to Foxe Basin and northern Québec stocks, while migratory harp seals have only been studied recently at one location. Contaminant measurements in bearded seal, harbour seal, bowhead whale and killer whale tissues from the Canadian Arctic are very limited or non-existent.*
- *There is much less information on contaminant levels and very limited geographic coverage for marine fish and invertebrates.*
- *While studies also show that the PCB contamination of terrestrial plants, soils and nearshore sediments and biota due to pollution from military radar facilities is quite localized when considered on a broad regional scale, there is a need to determine whether marine mammals frequenting the waters within the general area of these sites as well as terrestrial mammals, such as caribou and arctic fox feeding within the impacted zones, have elevated PCB and lead contamination.*
- *Information is more limited on OC levels in muktuk and other tissues of marine mammals and additional measurements should be made. In the case of muktuk, further studies should take into account any variations, between individuals or communities, in methods of preparing muktuk that might affect fat content of this food.*
- *Initial surveys of ringed seals and polar bear which showed higher PCDD/Fs in the high Arctic have not been followed up.*

3.6.3 Biological Effects

The observation that PCB concentrations in polar bear liver and some Northern Québec walrus are within or exceed the range of 200 ng·kg^{-1} (as TCDD TEQs) in blubber associated with immunotoxic effects in harbour seal, and that concentrations of PCBs, DDE, mercury, and especially TCDD TEQs, in fish tissue are at or near Canadian and US EPA guidelines for protection of fish-eating wildlife, suggest that we should not be complacent about the contaminant exposure levels of higher trophic level animals in the

Arctic. A large suite of biological effects indicators are available but only a few have been determined in samples from arctic animals. The limited data on biological effects indicators, especially immunosuppression in mammals at high trophic levels, is a major gap that should be addressed in future. It should be noted that moderate immunosuppression may have little effect under normal conditions, but can be critical if a population is involved in an epizootic such as that caused by the morbillivirus in seals and dolphins. A research program directed at polar bear and beluga immunology and immunosuppression is logically the top priority given the fact that these species have the highest levels of PCBs and other OCs such as chlordane and toxaphene.

At present, measurements of PCB congeners and other persistent OCs, are for the most part not linked to biomarkers. Linkage of the results for planar OCs to bioassays of CYP1A1 and CYP2B activity on sample extracts, a technique widely used in contaminant studies in the Great Lakes, should be considered in order to confirm that the biological activity associated with the measured contaminants is accounted for. Other planar compounds such as chlorinated naphthalenes and brominated and chlorinated diphenyl ethers, which have not been measured in Canadian Arctic samples, could account for some of the MFO activity observed in belugas and polar bears.

The strong positive correlation of CYP1A with non-ortho and mono-ortho PCBs in polar bear liver suggests that these compounds may be responsible for the activity. There were also indications that elevated CYP2B activity in polar bears was due to the induction by chlordane components (mainly oxychlordane and nonachlor) and ortho-substituted PCBs. Similar positive correlations of EROD activity with nPCBs levels were observed in beluga whales that were starving due to ice entrapment. This leads to the conclusion that arctic animals with relatively low levels of contaminants compared with temperate species may be vulnerable if they have to mobilize lipid deposits during fasting or starvation. Only limited work has been carried out to confirm these results in other species and to combine MFO measurements with other biochemical indicators of effects of PCBs such as retinoid and tocopherol levels. Given that some of the persistent OCs such as *p,p'*-DDE and *o,p'*-DDT have antiandrogenic activity, and that *p,p'*-DDE has been found to be inversely correlated with testosterone in porpoises (Subramanian *et al.* 1987), information is also needed on steroid hormone levels in polar bears and beluga (Colborn and Smolen 1996)

The very high levels of mercury and cadmium in sea birds and marine mammals have been known for some time but the biological implications for the animals themselves are unknown. Positive correlations of metallothionein and cadmium in sea birds demonstrate that the animals are responding to the metal exposure but they do not provide direct evidence for adverse impacts at the cellular or individual levels. As is usually the case with arctic animals, the lack of experimental dosage/response data continues to limit the ability to interpret concentrations observed in the animals. Given that the levels of, for example, cadmium, are among the highest ever reported in marine mammal tissues, further efforts are needed to examine possible physiological effects. For example, mercury is known to have neurotoxic effects at relatively high doses, but whether such effects are being observed in beluga or narwhal is unknown.

- *A large suite of biological effects indicators are available but only a few have been determined in samples from arctic animals. The limited data on biological effects indicators, especially immunosuppression in mammals at high trophic levels, is a major gap that should be addressed in future.*
- *Only limited work has been carried out to confirm observed correlations of non-ortho and mono-ortho PCB concentrations with CYP1A1 activity in polar bear and beluga livers and to combine MFO measurements with other biochemical indicators of effects of PCBs such as retinol levels.*
- *Given that some of the persistent OCs such as* o,p'-*DDE,* p,p'-*DDE and* o,p'-*DDT have estrogenic activity, information is needed on steroid and thyroid hormone levels in polar bears and beluga.*
- *The very high levels of mercury and cadmium in sea birds and marine mammals have been known for some time but the biological implications for the animals themselves are unknown. Given that the levels of for example, cadmium, are among the highest ever reported in marine mammal tissues, further efforts are needed to examine possible physiological effects.*

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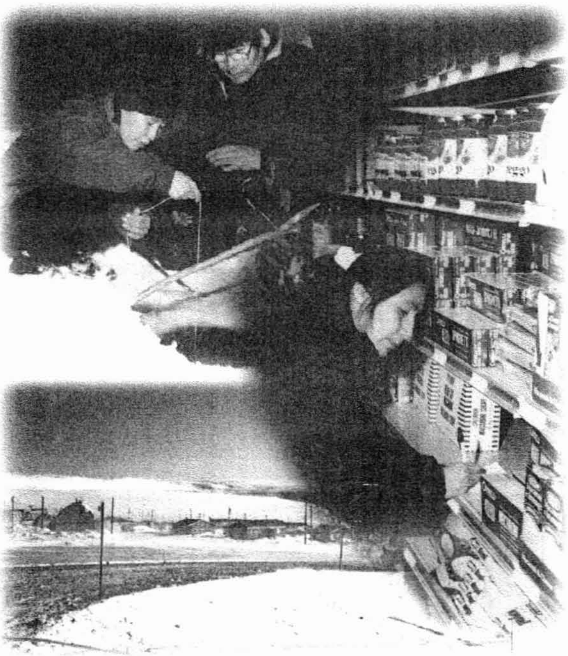

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4.1 Introduction

4.1.1 Objectives

This chapter reviews what is known about the human health implications of contaminants (particularly organochlorines, heavy metals, and radionuclides) in the Canadian Arctic and identifies the important knowledge gaps. Specifically, it addresses: the distinct susceptibility of Aboriginal Northerners to exposure to contaminants through their consumption of traditional/country food, and the significance and consequences of this exposure; the degree of exposure to and body burdens of contaminants; the human health implications, taking into account both the risks and benefits of country food consumption; and the distinctive requirements for risk management in the Arctic.

This chapter draws on existing knowledge and incorporates new information from the research and communication activities funded under the human health and communications subprograms of the Northern Contaminants Program of the Arctic Environmental Strategy (AES), which began in 1991. The AES human health research subprogram was not intended to address all of the questions relating to health implications of contaminants. As a result, many gaps remain. Some of these gaps will be filled as current research activities are completed; others will be filled with the initiation of new studies.

In this chapter, the term "contaminants" refers to metals, persistent chlorinated organics and radionuclides that are taken up in the food chain and pose a risk to consumers of country foods. Clearly, these contaminants are only some of many factors affecting human health and well-being in the Arctic. Many other hazards exist, some locally created (e.g. community garbage dumps) and some not (e.g. oil spills), some voluntarily assumed (e.g. cigarette smoking) and some not (e.g. exposure to arctic haze). Their assessment will need to be addressed elsewhere.

- *The key objectives of this report are to assess the impact on human health of exposure to current levels of environmental contaminants in the Canadian Arctic, and to identify the data gaps that need to be filled by future human health research and monitoring.*

4.1.2 Aboriginal Peoples of Canada

The Aboriginal peoples of Canada as defined in s.35 of the *Constitution Act*, 1982, include Indians, Inuit, and Métis. All of these peoples are present in the North. Six geo-political regions in Canada are identified as Inuit regions: Labrador, Nunavik (northern Québec) and in the Northwest Territories (NWT), the western Arctic (also known as the Inuvialuit Settlement Area) Baffin, Keewatin and Kitikmeot

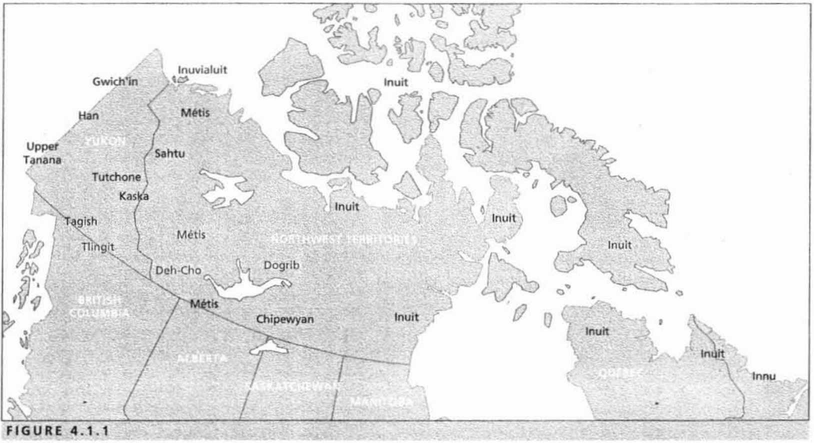


FIGURE 4.1.1

General locations of Arctic cultural groups.

TABLE 4.1.1

Proportion of Aborginal people in the total population of the Arctic region, 1991 (Statistics Canada 1995a).

Region	Total Population	% Aboriginal People
Yukon	27 655	17.1
Northwest Territories	57 430	62.6
Northern Québec	7 690	88.8
Labrador (Arctic only)	2 519	73.8
Total Arctic region	92 985	50.1

TABLE 4.1.2

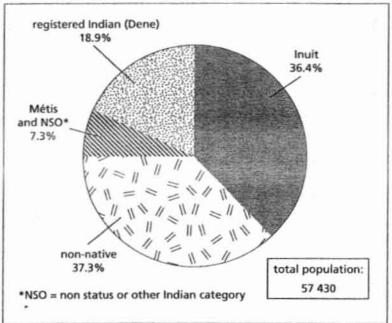
Life expectancy (at birth) for Canada and the total population (Aboriginal and non-Aboriginal people) of Yukon and the Northwest Territories.

Region	Male	Female
Canada	74.6	80.9
Yukon and NWT	71.9	75.9

(Young 1994, Statistics Canada 1995b)

(the latter three fall within the new territory of Nunavut). A small proportion of Inuit also reside in Yellowknife, Whitehorse, or southern Canadian cities. The Indian, or First Nation, peoples who reside in the NWT and the Yukon generally identify themselves as Dene, but there are also some Tlingit people in the southern Yukon. Métis are present in the Mackenzie Valley and southern Yukon (Figure 4.1.1). While "Aboriginal peoples" is the inclusive term normally used to describe the descendants of Canada's original inhabitants, the term "Indigenous peoples" is often used internationally. The term "Aboriginal northerner" is also used in this report.

Approximately 7% of Canada's Aboriginal people live in the Arctic region (Yukon, NWT, northern Québec and Labrador), where they comprise about half of the total population living in this region (Table 4.1.1). A population breakdown by ethnicity in the NWT shows the Inuit and non-native populations as the largest in the area (Figure 4.1.2). An average 81% of people living in those areas defined as Inuit regions identified themselves as Inuit (Figure 4.1.3).



*NSO = non status or other Indian category

FIGURE 4.1.2

Population estimates by ethnicity, NWT 1991. (Statistics Canada, 1995a)

A breakdown of demographic data for the NWT, the largest region in the Canadian Arctic (by area and population), indicates that there is a very high proportion of the total population between the ages of 0 to 14 years (32%) and 25 to 44 years (35%) (Figure 4.1.4). Similarly, in the Canadian Inuit population, 39% of the population are in the 0 to 14 age range and 27% are between 25 and 44 years of age (Figure 4.1.4). In comparison, the age breakdown for the total Canadian population shows a more even age distribution with a smaller young population and larger population in the 65 and over age class. However, as with both populations described above, the largest segment of the population is in the 25 to 44 age group (33.5%).

Life expectancies in the Yukon and NWT populations (including Aboriginal and non-Aboriginal populations) are lower than the Canadian national values (Table 4.1.2). According to Young (1994), North American Aboriginal people in general have a reduced life expectancy at birth, primarily due to their higher rates of infant mortality. For the period 1961–1966 in the NWT, Inuit infant mortality rates were 144/1000, compared with 26/1000 for the total Canadian population. For 1981–1985 this rate was 28/1000 for the Inuit, compared with 9/1000 for all of Canada (Young 1994).

- *Approximately 7% of all Aboriginal people in Canada live in the arctic region, which includes the Yukon, the*

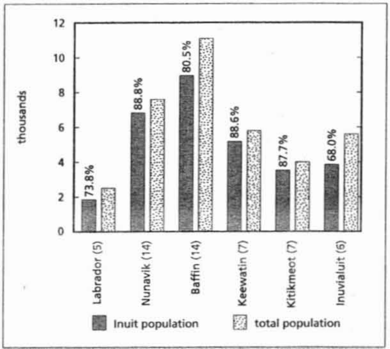


FIGURE 4.1.3

Population of Inuit regions of Canada (includes only residents of organized communities within Inuit region). Numbers in brackets beside regions represent the number of Inuit communities in the region. Percentages represent the percentage of Inuit in the total population. (Adapted from data from Statistics Canada 1995a)

NWT, northern Québec, and Labrador. Aboriginal people make up about 50% of the population living in the arctic region. Comparing the age breakdown of the Canadian with that of the Arctic illustrates the fact that Arctic populations are younger than the Canadian population, which has a more even age distribution.

4.1.3 Health

This assessment report focuses on environmental contaminants and their effects on the northern ecosystem and on human health. There are, however, many factors that affect the health of individuals and communities in the North, as elsewhere. While a complete presentation of these factors is beyond the scope of this report, the following provides a framework for addressing environmental contaminants in an overall context of human health. This is relevant for a more complete understanding of Northerners' health, as well as for communication concerning environmental contaminants and health.

According to the World Health Organisation's (WHO) definition, health is:

> a state of complete physical, mental, and social well-being and not merely an absence of disease or infirmity...a resource for everyday life, not the objective of living...a positive concept emphasizing social and personal resources as well as physical capacity.
>
> (WHO 1967)

This holistic definition recognizes that health, the ideal state, is socially and culturally defined. Various researchers note that similar holistic views are held by Indigenous groups of the Arctic but are expanded to take into account the spiritual dimensions (Figure 4.1.5).

The Government of the NWT (GNWT) recognizes five "determinants of health," all of which contribute to the health status of the individual and that of the community (Department of Health and Social Services 1995). To enhance the health and well-being of the overall population, the following health determinants must be addressed in a comprehensive and interrelated way:

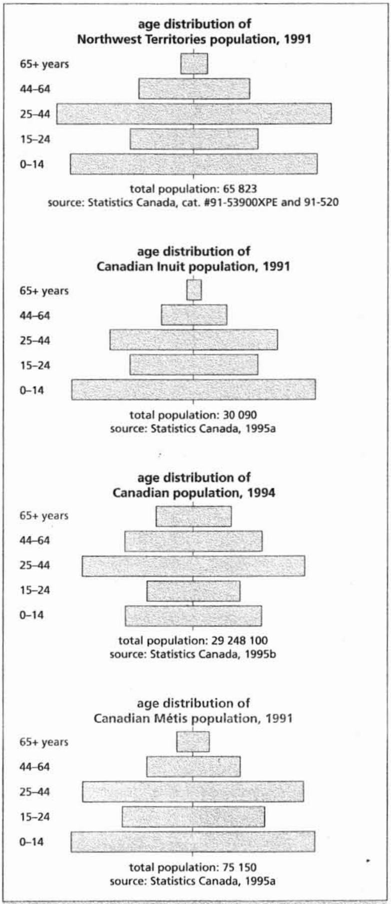
i. ***The social and economic environment*** includes income and social status, social support networks (e.g., support from families, friends and communities), education, employment and working conditions.

ii. ***The physical environment*** includes the natural environment (e.g., air, water, food, soil quality) and the human-built environment (e.g., housing, workplace safety, community and road design). Ecosystem health includes human health.

iii. ***Personal health practices*** are behaviours that can enhance health or increase personal susceptibility to risk factors. They include physical activity, food preparation and consumption activities, smoking, use of alcohol and other drugs, and the adoption of protective behaviour in regard to environmental factors.

iv. ***Individual capacity and coping skills*** include both biological and genetic characteristics as well as psychological characteristics (e.g., personal competence, coping skills, sense of control).

v. ***Health services*** include the quantity, quality, arrangement, nature and relationships of people



age distribution of
Northwest Territories population, 1991

total population: 65 823
source: Statistics Canada, cat. #91-53900XPE and 91-520

age distribution of
Canadian Inuit population, 1991

total population: 30 090
source: Statistics Canada, 1995a

age distribution of
Canadian population, 1994

total population: 29 248 100
source: Statistics Canada, 1995b

age distribution of
Canadian Métis population, 1991

total population: 75 150
source: Statistics Canada, 1995a

FIGURE 4.1.4

and resources in the provision of health care. Health Centres (formerly known as Nursing Stations in some regions) and nurses, along with Community Health Representatives (CHRs) play a key role in Northern communities, compared to the roles of hospitals and physicians elsewhere in Canada.

Healthy child development is not included as a separate category in the framework, in spite of its crucial importance as a determinant of adult health. Rather, each of the above categories includes factors known to contribute to healthy child development.

Northern Aboriginal people perceive "health" as a synthesis of the state of being and the individual well-being of the entire community (Borré 1990, 1994, Young 1994, Johnson 1992). Clearly, and in contrast to the situation that prevails in the Western medical/health framework, it is the community and the environment — not the individual as separate from the community — that form a critical component for understanding good health (Wenzel 1981, Stairs and Wenzel 1992).

Aboriginal interpretation of health focuses on wellness rather than on sickness. The concept of being healthy is expressed in many native languages by a word or phrase which, roughly translated, means "feeling good all over" (Wheatley 1995). Among Yukon First Nations, for example, little distinction is made between food and medicine (the latter mostly from plants), because both come from the land and both are required for health and healing (Wheatley 1994). Health represents wholeness and balance among all the interrelated aspects of well being (Figure 4.1.5). Balance is perceived by Aboriginal people in their close relationship with the environment, with appropriate behaviour ensuring the maintenance of good health. Cultural identity is expressed in traditional values such as respect and responsibility for the environment and its resources, which promote physical, cultural and spiritual health (Wheatley 1996). Environmental degradation is therefore seen as a direct threat to the maintenance of human health.

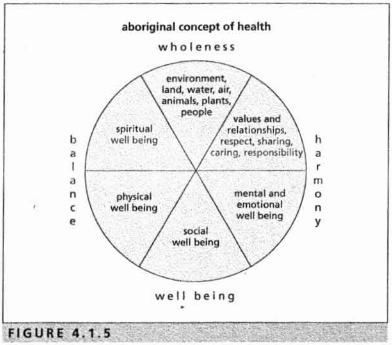


FIGURE 4.1.5

source: Wheatley, 1996

- *The concept of "health" in Indigenous groups of the Arctic is holistic in that it recognizes that health, the ideal state, is socially and culturally defined and has spiritual dimensions. Cultural identity is expressed in traditional values such as respect and responsibility for the environment and its resources which promote physical, cultural and spiritual health. Environmental degradation is a threat to health.*

4.1.4 Aboriginal Perspectives on Food and Health

In Aboriginal populations, food is perceived as an integral component of being healthy. While this report cannot discuss this issue in full detail, it attempts to illustrate how Aboriginal perspectives on food and health differ from those in Western societies.

Aboriginal people describe their food quite specifically: as Inuit food, or Dene food, as the case may be. However, in this chapter we have used the terms "country food" and "traditional food" interchangeably to refer to the mammals, fish, and birds harvested from local stocks (i.e. those foods also constituting the possible vector for contaminant exposure). There is no unanimous preference among Aboriginal Northerners for either term, both of which are generic and have no direct translation into Inuktitut or the Dene languages. "Imported" or "market" foods are terms used interchangeably to refer to all other foods which are mainly bought from the store.

For Inuit, country foods are directly associated with physical health and well-being. At Sanikiluaq, for example, people regard certain foods such as seal meat, as capable of generating bodily warmth and strength in a way that imported foods cannot. Such foods are therefore essential in the diet for Inuit activities such as hunting (Usher *et al.* 1995). However, as Egede (1995) notes, country foods represent not only health in the most obvious, corporeal sense, but they also constitute an essential basis of personal and community well-being for Inuit and Dene.

> Inuit foods give us health, well-being, and identity. Inuit foods are our way of life...Total health includes spiritual well-being. For us to be fully healthy, we must have our foods, recognising the benefits they bring. Contaminants do not affect our souls. Avoiding our food from fear does.
>
> (Egede 1995)

Among Inuit, individual life is itself seen as a synthesis of two elements that can be roughly translated as: the body (the physical actuality and functioning of the human body) and the soul (spirit, mind, immediate emotional state, or even the expression of consciousness) (Borré 1994). Traditional food is essentially important in contributing to this synthesis (Borré 1994), and thus to the way individuals conceptualize their own well-being (Ridington 1988). Among Inuit, this integration is accomplished through the capturing, sharing, and consuming of country food. The cultural aspects related to the harvesting of traditional foods such as sharing and communal processing of food are important to individual and community health.

- *Traditional foods are an integral component to good health among Aboriginal people.*

4.1.5 Social, Cultural and Spiritual Importance of Traditional Food

For Aboriginal peoples, the way in which individuals and entire communities participate in subsistence depends as much on social aspects — giving, receiving and consuming traditional food — as it does the actual harvesting of wild game. Harvesting, sharing, processing, and consuming traditional food is important to cultural identity. To stop hunting or fishing (or to stop eating country food) is for many also to stop participating in subsistence in its broadest sense. Consequently, the contamination of country food raises problems that transcend the usual confines of public health, and that cannot be resolved simply by health advisories or food substitution. This concept is well illustrated in the following citation:

> Let me mention the upbringing to hunting, the hunt itself, the upbringing to be a hunter's wife, the processing of food, the sharing of meat — all are to be seen as a social glue that shapes our minds, our feeling of belonging to a society.
>
> When many other things are changing, our foods remain the same, and they make us feel the same as they have for generations. Maybe that is even more true today, since we see so many influences from outside, and we think more often about what it means to be an Inuk.
>
> What we do know is that avoiding Inuit foods today will affect our society, our communities, and our culture. Foreign foods do not bring joy when we share them. They do not tie families and communities together and to one another. Only Inuit food sustains the Inuit way of life.
>
> (Egede 1995)

The Inuit's emphasis on the importance of traditional food is further illustrated by an Inuk hunter from Nunavut's response to the question "If you were told by some researcher with whom you were familiar that you had to stop eating ringed seals, how would you react?" (Poirier *et al.* 1996):

> Ah, I would be sceptical of this, and would not do as advised. I am an Inuk and my way is to eat this. I cannot do without it, because it is intrinsic to who I am. Even if I am told this is unsafe to eat, I think I could not refrain from doing so.

The act of harvesting and consuming animals is just one component of a socio-cultural process of sharing and receiving that extends, on the one hand, from animal to human and, on the other, from person to person. The connection between animals and humans is accomplished through the action of harvesting (Fienup–Riordan 1983, Ridington 1988, Brody 1987). Sharp (1988) notes the intensive connection between Dene and the animals they use as food. This is embodied in the concept of *inkoze*: that a game animal cannot be killed without its consent. *Inkoze* embodies the integral relationship between humans and animals and the connection between those living and the cultural past. This process of transformation is continued through the normative allocation of country food (Stairs and Wenzel 1992) within Dene and Inuit societies. In all aspects of these exchanges, as Egede (1995) notes above, traditional food is as much the substance of Aboriginal Northerners' social well-being as it is that of their physical health.

Exchanges of country foods within northern Aboriginal societies also involve a complex set of socio-economic rules and procedures that correlate strongly with the structural organization of these societies. Food is given and received within a socially constructed framework of reciprocity; the generosity that underlies the relationship between animals and humans also directs the process of human sharing. The tenets of this system of sharing emphasize the relation of an individual to his or her kindred, and of kin groups to community through a web of practices that ensure that food is available to all who are in need (Wenzel 1995). In many communities, a relatively small proportion of the population produces country food well in excess of the harvester groups' own immediate needs (Wenzel 1991).

A recent survey in 16 Dene/Métis communities addressed the social and cultural functions of traditional food. Preliminary results indicate that ≥90% of 1012 respondents believed that harvesting and using traditional food by the family:

- keeps people "in tune with" nature
- promotes sharing in the community
- is an essential part of community culture
- contributes to childrens' education by teaching skills in survival, food preparation at home, and providing opportunities for developing patience and other desirable attributes

The survey also reported that respondents believe that a successful harvest brings respect from others and builds one's pride and confidence.

More than 70% of respondents also believed that harvesting traditional food presents an opportunity to practice and teach humility and spirituality (Kuhnlein *et al.* 1996b).

Finally, as noted by Egede, country food, because it is the point of articulation between sociocultural, socioeconomic, and pragmatic decision making, provides the focus upon which northern Aboriginal identity, whether Dene/Métis or Inuit, is founded. In the most definite terms, the act of gathering, processing, sharing and consuming traditional food defines "what it means to be an Inuk."

- *Traditional food is as much the substance of Aboriginal Northerners' social well-being as it is that of their physical health. Harvesting, sharing, processing and consuming traditional food is important to cultural identity. Harvesting traditional foods is an opportunity to practice and teach humility and spirituality. Sharing of foods, quite common in many communities where a small proportion of the population produces food well in excess of the harvester groups' own needs, is also an important sociocultural process. Consequently, the contamination of country food raises problems which go far beyond the usual confines of public health and which cannot be resolved by health advisories or food substitutions.*

4.1.6 Factors that Contribute to Aboriginal Northerners' Exposure to Country Food Contamination

Food is a significant vector for contaminant exposure in all populations (with few exceptions, water, air and occupational exposure are limited and relatively insignificant). Because a substantial proportion of the Aboriginal diet consists of country food, Aboriginal Northerners have a higher risk of contaminant exposure than the non-Aboriginal population in the North specifically or in the rest of Canada.

Most of the traditionally harvested fish and land and marine animals taken are long-lived and are from the higher trophic levels of the food chain thereby providing an opportunity for considerable bioaccumulation and biomagnification of persistent contaminants (Chapter 3). Aboriginal Northerners are especially likely to be exposed to metals such as mercury and cadmium through the consumption of fish and mammalian organ meats respectively, and to organochlorines (OCs) through the consumption of fats, especially those of marine mammals (Kinloch *et al.* 1992, Kuhnlein 1995, Kuhnlein *et al.* 1995c).

The health impacts of country food consumption cannot be fully assessed by estimating exposure to contaminants alone. They must also be considered in terms of the distinctive significance of traditional food to Aboriginal Northerners. Hazard and risk, as social scientific studies of these phenomena have noted, are not merely "objective" categories, they are socially constructed. Vulnerability to a hazard is a function not only of the intrinsic nature of the hazard but also of the geographic and socio-economic location of people exposed to it, as well as their cultural perspective on the hazard and its vectors (Hewitt 1983, Douglas 1992).

Country food is an economic necessity for most Aboriginal northerners. In many northern communities, employment and incomes are low, and country food makes a significant addition to effective household income. A recent survey in sixteen Dene/Métis communities calculated the cost of a standard basket of market food to provide a nutritionally adequate diet for a family of four persons for one week. This basket, which cost $110 in Edmonton, ranged in price in the survey communities from $133 at the Hay River Dene Reserve to $254 in Kahbamitue (Colville Lake) (Kuhnlein *et al.* 1996b). Calculations of the cost to purchase equivalent amounts of imported meats in local stores have estimated the value of traditional food production in the NWT to be $55 million, or well over $10,000 per Aboriginal household per year (Usher and Wenzel 1989). Such calculations apply only to the product itself, as a food commodity. They do not include any other values that Aboriginal people commonly attach to traditional foods such as the activity of hunting itself, or the collective cultural and social values of sharing and teaching (section 4.1.5).

There are also health costs, which are borne both by Aboriginal people themselves, and by public health programs, associated with not eating country food. For example, reduced country food consumption in northern Native populations, coupled with decreasing physical activity, has been associated with obesity, dental caries, anemia, lowered resistance to infection, and diabetes (Szathmary *et al.* 1987, Thouez *et al.* 1989).

- *The geographic and socio-economic location of Aboriginal Northerners makes them particularly susceptible to the negative implications of country food contamination. Country food is economically essential for most Aboriginal Northerners. Calculations of the cost to purchase equivalent amounts of imported meats in local stores have estimated the value of traditional food production in the NWT at well over $10,000 per Aboriginal household per year.*

- *Reduced country food consumption in northern Native populations has also been associated with decreased physical activity, obesity, dental caries, anemia, lowered resistance to infection, and diabetes.*

4.2 Traditional Food Consumption in the Arctic

This section reviews data derived from harvest and diet surveys on the quality and quantity of country food consumed, and the proportional contribution of country food to the diet. Harvest data exist in some form for virtually every Inuit community, much of it obtained in the last 20 years in preparation for, or implementation of, comprehensive land claim agreements. These freely available data can be used to estimate potential gross dietary exposure to contaminants on a community basis, and in some cases to monitor trends in consumption rates and species consumed. Few such data exist for Dene communities in the Yukon or the NWT although there have recently been numerous diet surveys in these regions, many conducted under the AES/NCP research program (Figure 4.2.1). While there are problems of intercomparison of food consumption studies, harvest and diet survey data can provide the basis for provisional food intake estimates. The nutritional benefits of country food including their vitamin, mineral, lipid, and protein contents are also considered because of their applicability to risk-benefit evaluations.

4.2.1 Harvest Survey Results

Regional harvest survey data from across Canada (Table 4.2.1) indicate that country food is widely available to, and widely harvested by, Aboriginal Northerners. The amount available for human consumption ranges from 87 to 284 kg·capita^{-1}·year^{-1}, with the lowest amounts in more urbanized, road accessible areas, and the highest in more isolated areas. Regional aggregates mask the tendency for community variation, nonetheless the great majority of the people of the nearly 100 communities across the Canadian Arctic fall within this per capita range. Similar regional aggregate averages (170–276 kg·capita^{-1}year^{-1}) are reported in comparable regions of Alaska (Wolfe and Walker 1987). Dietary survey results are consistent with these findings, and indicate that traditional foods are widely consumed within communities. Consequently, potential exposure of Aboriginal Northerners to contaminants in country food is not an isolated or local problem, but it is widespread across the Canadian Arctic. Actual levels of exposure, as measured by contaminant levels in local food supplies and by more precise dietary surveys, are discussed in section 4.4.5.

Harvest statistics estimate the number of animals harvested (i.e. struck and retrieved) and are usually presented as annual totals by community (Usher and Wenzel 1987). Applying known parameters of edible weight per animal, estimates of the quantity of country food available for consumption can be

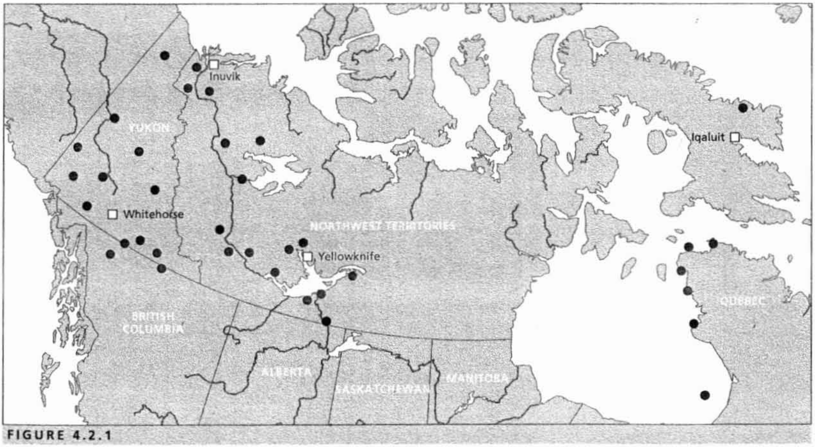


FIGURE 4.2.1

Locations of dietary surveys conducted and reported since 1985. (Kuhnlein and Soueida 1992, Kuhnlein et al. 1995a, Receveur et al., in progress, Wein 1994a, Wein 1995c) derived by species. These data are standardized in Table 4.2.1 as kg edible weight·capita^{-1}·yr^{-1}. Harvest survey data have been standardized by Usher and Wenzel (1989), Coad (1994), and Weihs *et al.* (1993). In the partial or complete absence of comprehensive harvest surveys, the Yukon and Labrador regional estimates rely heavily on administrative data and speculative estimates (see Usher and Weinstein 1991 p.15 for definition of these terms).

While Table 4.2.1 presents harvest survey data, actual consumption is somewhat less. Reasons include loss or non-consumption of harvest, cooking (which may account for up to 25% weight loss), and plate waste (Wein 1995b). Formerly, a substantial quantity of the harvest was used to feed dogs. This proportion has become negligible in recent years, with the near-universal adoption of snowmobiles for winter transport, and can be ignored in interpreting Table 4.2.1. First Nation community estimates, standardized by Coad (1994), are the only ones to exclude dog food.

There are no regional harvest data for the Dene communities of the Mackenzie Valley, although Berger (1977) estimated an apparent harvest rate of 108 kg·capita^{-1}·yr^{-1} for the Mackenzie Valley, Beaufort Sea, and north Yukon areas as a whole. Table 4.2.1 also includes two community harvest estimates derived independently from the regional surveys cited.

By weight, the key food resources for Aboriginal Northerners are large ungulates (caribou, and in the southern Yukon and NWT, moose), and fish. Marine mammals are an important food source in all Inuit regions, although seal is less frequently consumed in the Inuvialuit region. Fish is at least a seasonal staple throughout the north, with a few exceptions. A Canada-wide review of 93 community and 10 regional Aboriginal domestic fish catch estimates (Berkes 1990) suggest a median value of 45 kg edible fish·capita^{-1}·yr^{-1}. Northern Aboriginal communities are probably close to this value. The amount of country food harvested per capita exceeds the Canadian average consumption of meat and fish (75.8 kg in 1988 — Weihs *et al.* 1993) in every region of the North.

- *Traditional foods are widely consumed within communities, the key food resources being large ungulates and fish. Marine mammals are important food sources in Inuit regions. Consequently, potential exposure of Aboriginal Northerners to contaminants in country food is widespread and typical across the Canadian Arctic, including northern Québec.*

4.2.2 Dietary Survey Results

Dietary surveys can provide more accurate estimations of potential dietary exposure to contaminants than harvest surveys. This is partly because they measure consumption directly. However, some diet surveys intended to estimate dietary exposure to contaminants focus on particular at-risk groups, particular parts of animals (e.g. organ meats and animal fats that are known to contain substantial amounts of both contaminants and nutrients) and the form of food preparation. As a result, it is not always possible to derive a directly comparable community-wide estimate of total country food consumption, expressed as kg·capita^{-1}·yr^{-1}, from the survey data. Some dietary survey information, coupled with human body weight by age and gender, can be used to estimate probable weekly intakes of contaminants that are used in "risk determination" (section 4.4.1).

Dietary surveys usually employ some combination of periodic 24-hour recall, frequency of food

TABLE 4.2.1.

Country food available for consumption in northern Canada, as measured by harvest surveys

	Edible weight[1] (kg·capita^{-1}·yr^{-1})	Leading foods[2]	Source
FIRST NATIONS			
Regional estimates[3]			
Yukon	87	moose, caribou, fish	Usher and Staples 1988
Québec Cree	123	fish, moose, waterfowl	JBNQ 1982
Community estimates			
Deline (Fort Franklin), NWT	131	fish, caribou, moose	Rushforth 1977
Fort Resolution, NWT	69	muskrat, rabbit, fish	Bodden 1981
INUIT			
Regional estimates[3]			
Inuvialuit	164	caribou, fish, muskox	Fabijan 1991
Kitikmeot	270	caribou, fish, seal	Jingfors 1984
Keewatin	214	caribou, fish, seal	Gamble 1988
Baffin	275	(not indicated)	Pattimore 1985
Nunavik	284	fish, caribou, seal	JBNQ 1988
Labrador	131	fish, caribou, seal	Usher 1982
Community estimates			
Makkovik, Labrador	96	caribou, fish, seal	Mackey and Orr 1987

[1] *Edible weight normally includes eviscerated, partially bled carcass, bone out, plus normally consumed viscera (see, for example, Usher 1971, JBNQ 1976)*

[2] *In order of contribution by weight to the total harvest*

[3] *Includes all communities in the region*

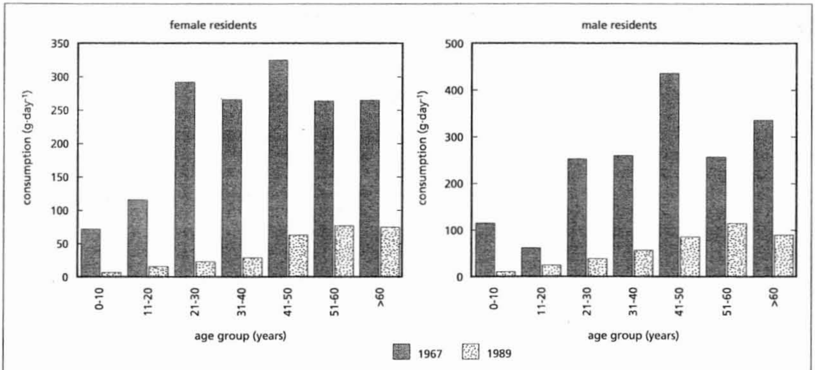


FIGURE 4.2.2

Caribou consumption among female and male residents of Baker Lake in 1967 and 1989. (Tracy et al. *1996)*

consumption, and food preference, each obtained by survey methods (Gibson 1990, Wein 1995a). A key methodological issue is the precise estimation of portion size by respondents. In general, it is recognized that methodologies used for frequency data result in overestimation of intakes, and that methodologies used for 24-hour recall data result in underestimation of intakes. One way to increase confidence in recall data is to compare total reported energy intakes based on reported consumption with accepted intake standards, which range from 1800 to 3000 kcal·day^{-1} (arithmetic mean) for healthy adults. Differences in seasonal consumption, typical of traditional food consumers versus market food consumers, renders the calculation of annual consumption rates problematic, unless a sufficient number of 24-hour recall periods and sufficiently precise recall periods for frequency surveys are used.

Nonetheless, per capita estimates of annual country food consumption which can be compared with those based on harvest data in Table 4.2.1, can be derived from some recent dietary surveys. Table 4.2.2 shows that while a few species typically account for the bulk of country food consumption, many animal and plant species are utilized.

- *Dietary surveys indicate that while many animal and plant species are used as country food, a few species typically account for the bulk of country food consumption.*

4.2.3 Traditional Food as a Proportion of Aboriginal Diet

The diet of Aboriginal Northerners consists of a mix of country food and imported, store-bought food. The trend began decades (and in some cases a century or more) ago when staples such as flour, sugar, and tea were introduced to the northern diet, chiefly through the fur trade. Imported foods have become increasingly available, and to a degree more affordable, in the north. In every community, there are a variety of groceries in local stores, and in some, there are also fast food outlets and restaurants. Television and travel have created a demand for a wider variety of foods, and improved transportation has enhanced the supply. At the same time, the everyday realities of contemporary community life, such as employment, education, and housing, require that food be readily obtained, stored, and prepared. "Delocalization" of the food supply in remote and rural communities has been widely observed (Pelto and Pelto 1983, Waldram 1985) and is associated with industrialization, improved transport, and modernization.

In some communities, dietary changes have been quite recent. For example, based on whole body measurements of radiocesium at Baker Lake, Tracy *et al.* (1996) found that caribou consumption rates among adults in 1989 were about one-quarter the rate in 1967, and that the decline in consumption was especially notable among young adults and among women of all ages (Figure 4.2.2). This trend appears to be supported by existing harvest survey data. ·Country food harvests at Baker Lake were estimated at 320 kg·capita^{-1}·yr^{-1} in 1977 (IDS 1978), and 154

$\text{kg}\cdot\text{capita}^{-1}\cdot\text{yr}^{-1}$ nine years later (Gamble 1988), although the survey methods were not identical.

A less physically strenuous lifestyle, especially in cold weather, results in reduced total food needs. As a result of the mechanization of hunting, the effort (as measured by human energy or time) required per unit of food production has declined substantially, although there are no comprehensive quantitative estimates available. Nonetheless, as Table 4.2.1 indicates, country food continues to be an important part of the contemporary diet, and it may be that contemporary consumption rates have stabilized.

- *Traditional diets consist of both traditional and imported foods. Due to many factors associated primarily with industrialization, improved transport, and modernization, imported foods have become much more available in the North. Nonetheless, country food remains an important part of the contemporary diet.*

4.2.4 Variation and Trends in Food Use

Estimations of food intake from diet surveys — which often only measure consumption for two or three 24-hour periods over one year — are complicated by the variations in food use that are common in Aboriginal communities. Variation in food use is due to many factors including natural sources of variation (seasonal or geographic), economic sources, access to urban centres, and variations relating to age or gender.

The variation in abundance and availability of different species regionally, seasonally, and from year to year are natural sources of variation in traditional food consumption. In addition to a few staple food species, a wide variety of country foods may be consumed on a seasonal or occasional basis, sometimes at very high daily rates over short periods. Seasonal intakes of traditional foods by Inuit and Sahtu Dene/Métis were considerable, with daily average portion sizes ranging as high as 562g (Tables 4.2.2 and 4.2.3). Local economic conditions affecting income and employment can also affect harvesting opportunities and therefore the specific components of diet. Some of this variability is modified by the growing phenomenon of intersettlement trade, that is, the exchange (by means of barter or sale) of surpluses between communities and between families.

A study of per capita harvests in 98 Alaskan communities showed that harvest levels tend to be inversely related to proximity to urban centres, particularly the degree of road access from them, and to personal incomes. Road access tends to be associated with increased competition for local resources by non-local residents, increased habitat alteration, and to some extent to an increase in wage-based employment (Wolfe and Walker 1987). Similar tendencies have been noted in the Yukon (Usher and Staples 1988), but no comprehensive analysis of these relationships exists for the Canadian North.

Country food use has also been shown to vary by age and gender. Twenty-four hour recall diet surveys in Baffin Inuit and Sahtu Dene/Métis communities have shown that more traditional food is consumed by older adults than by younger adults and children, and by men than by women (Kuhnlein 1995, Kuhnlein *et al.* 1995a, 1995b, Kuhnlein *et al.* 1996a, Morrison *et al.* 1995). However, when the data are controlled for moisture and for intake per unit energy, intake was equal for adults of both genders (Kuhnlein 1995). Similar generational differences were also reported by Wein *et al.* (1991) for Aboriginal people living near Wood Buffalo National Park.

There are few data on food use by age and gender prior to the 1970s. Earlier ethnographic data focused mainly on the methods of country food preparation

TABLE 4.2.2

Dietary survey results — country food consumption in northern communities

Location	Country Food consumption (kg·yr^{-1})	# species consumed	Proportion of country food consumed by species	References
Sanikiluaq, NWT[1]	292	39[2]	marine mammals (39%) reindeer (34%) fish (13%)	Wein 1995c
Broughton Island, NWT	110[4]	25[5]	—	[4]Kinloch *et al.* 1992 [5]Kuhnlein and Soueida 1992
Wood Buffalo Park area, NWT and Alaska	27	—	large mammals (40%) berries (20%) fish (19%)	Wein *et al.* 1991
Sahtu Dene/Métis communities	—	30	—	Kuhnlein *et al.* 1994
Four Yukon communities	—	80	—	Wein and Freeman 1995
16 Dene/Métis communities in NWT and 10 Yukon First Nations communities	—	> 35 animal and 33 plant	—	Receveur 1996a[3]

[1] *Country food was consumed more than three times a day per household*

[2] *Inuit communities only*

[3] *Preliminary results*

and consumption. The general assumption in the literature is that all parts of all food species were consumed similarly by males and females of all ages, except infants, for whom the broth of boiled meats was the primary food. More recent ethnographic and dietary research notes a tendency among younger Dene and Inuit to consume a diet with a higher proportion of imported foods. Reduced consumption of country food by youth is often attributed to the fact that school age children and adults who do not hunt themselves are less likely to consume country food in the community (Borré 1990). However this observation is moderated by mention that more country food is consumed when these same groups are resident in "camp" (Borré 1990). Other work (Condon *et al.* 1995, Wein and Freeman 1995), however, suggests that even households identified as only occasional participants in hunting can partake of substantial amounts of country food, as calculated by number of meals eaten.

It is uncertain whether or not the reported tendency of young people to eat less country food is a recent phenomenon. Baker Lake data for 1967 (Tracy *et al.* 1996) show that even then, caribou consumption by those under 20 years of age was only about one-third the level of persons 21 years and older (Figure 4.2.2).

The data on country food use by younger Dene and Inuit suggest that youth do consume these foods, but that residence, location, and degree of harvesting may affect their total consumption relative to older adults. It appears that increased formal education and employment among youth can delay, but not necessarily eliminate, their becoming active harvesters and consumers of country food. Consequently, it should not be assumed that low country food consumption rates in young cohorts will persist as they age. A 1992 diet survey of Nunavik Inuit indicated that among women, there was no significant variation in the quantity of country food consumption by age group (Begin and Parent 1995).

In the study "Variance in Food Use in Dene/Métis Communities" conducted by the Centre for Nutrition and the Environment of Indigenous Peoples (CINE), a total of 1012 individual interviews were collected in 16 communities (Receveur 1996a). There was variation among communities in the types and amounts of traditional food species used, and in seasonal use. The contribution of traditional food to dietary energy also varied. Table 4.2.3 provides some preliminary results from this study. From the data analysed to date, it can be noted that the amounts of food consumed per person ranged considerably from 82 g·person^{-1}·day^{-1}, to 562 g·person^{-1}·day^{-1}.

- *A complication in estimating food intake from diet surveys is that while the surveys may only measure consumption for two or three 24-hour periods, country food consumption varies significantly seasonally, and annually. Country food consumption also varies regionally, according to income and access to urban centres, and as a factor of age and gender. Recent dietary research indicates that the diets of younger Dene and Inuit may consist of a higher proportion of imported foods.*

4.2.5 Nutrient Benefits of Aboriginal Diets

The benefits of country food extend beyond their importance in maintaining cultural, spiritual and physical integrity. They are nutritionally vital. They are an important source of lipids, vitamins and minerals and protein and in many cases are the primary source of many important nutrients. Market foods, however, are more significant sources of dietary energy. The type of lipid derived from marine fish and mammals is considered to be responsible for

TABLE 4.2.3

Variation in traditional food use in Dene/Métis communities: intake and percent of total dietary energy[1] (Receveur 1996)

	Fall		Late Winter	
Community	Intake gms·person^{-1}·day^{-1} (mean)	% of total dietary intake (mean±SE)	Intake gms·person^{-1}·day^{-1} (mean)	% of total dietary intake (mean±SE)
Aklavik	200	17 ± 3	197	15 ± 2
Tsiigehtchic (Arctic Red River)	227	22 ± 3	246	17 ± 4
Fort McPherson	562	31 ± 3		
K'ásho Gotine (Ft. Good Hope) + K áhbamitúé (Colville Lake)	455	27 ± 5	383	29 ± 3
Déline (Ft. Franklin)	285	24 ± 3	376	28 ± 4
Rae/Edzo	315	26 ± 3	307	25 ± 3
Pedzehki (Wrigley)	188	14 ± 2		
Lidlii koe (Ft. Simpson)	87	8 ± 2	104	7 ± 1
Jean Marie River	97	8 ± 3		
Fort Providence	155	13 ± 3		
Lutsel k'e (Snowdrift)	336	24 ± 3	386	30 ± 3
Fort Resolution	193	9 ± 3	205	11 ± 2
Hay River Reserve	202	12 ± 2		
Fort Smith	82	6 ± 1		

[1] *Data include both consumers and non-consumers of traditional food*

TABLE 4.2.4

Average daily intake of energy and selected nutrients for arctic women 20–40 years of age: traditional vs. market food

	Energy (kcal)	Carbohydrate (g)	Protein (g)	Fat (g)	Vitamin A (RE)	Iron (mg)	Zinc (mg)
BAFFIN INUIT[4]							
Traditional	661	2	85[1]	32	455[1]	43[1]	12[1]
Market	1636[1]	218[1]	52	63[1]	343	10	6
Both	**2297**	**220**	**137**	**95**	**798**	**53**	**18**
QUÉBEC INUIT[2,5]							
Both	**1829**	**200**	**89**	**76**	**792**	**16**	—
SAHTÚ DENE-MÉTIS[6]							
Traditional	421	0	80[1]	9	20	12[1]	12[1]
Market	1750[1]	190[1]	63	77[1]	477[1]	10	8
Both	**2171**	**190**	**143**	**86**	**497**	**22**	**20**
CANADA MARKET[7]	2001	227	72	89	1292	12	9
NOVA SCOTIA MARKET[8]	1571–1721[2]	188–206[2]	67–69[2]	62–67[2]	845–979[2]	11[2]	—
QUÉBEC MARKET[3,9]	1727–1867	206–224	73–76	65–73	1100–1148	12	10
RECOMMENDED NUTRIENT INTAKES[10]	1900		51	63	800	13	9

[1] *Significantly higher (Wilcoxon paired-sample t-test)*

[2] *Data are reported as ranges representing age groups in the 18–49 year range*

[3] *Québec districts: Matagami, Lebel-sur-Quévillon, Baie Jaimes, Fermont, Schefferville, Saint-Jean Eudes, Basse-Cote-Nord, Territoire Inuit, Territoire Cree*

[4] *Kuhnlein H.V.* et al. *1996a*

[5] *Department of Indian Affairs and Northern Development 1994*

[6] *Kuhnlein HV* et al. *1995b*

[7] *Health and Welfare Canada, Nutrition Canada 1976*

[8] *Nova Scotia Department of Health and Health and Welfare Canada 1993*

[9] *Dewailly* et al. *1994b*

[10] *Health and Welfare Canada 1990*

the low rate of heart disease in Arctic Aboriginal peoples. Certain mineral nutrients are believed to interact with some environmental contaminants so as to reduce their negative effects. This section will discuss the nutritional importance of traditional foods with some comparisons to imported foods, and will discuss added benefits from the nutrients common to traditional diets. All of these factors contribute to a better understanding of the benefits of country food and are important considerations in the risk determination process.

4.2.5.1 Daily Energy Intakes

The contribution of traditional food to the total daily energy requirement varies among studies. In Sanikiluaq on the Belcher Islands (which are isolated/remote), Wein (1995c) found that country food provided 47% of daily energy compared to only 29% in Baffin Inuit women and 19.4% among Sahtu Dene-Métis women (Table 4.2.4). Imported food contributes more to the daily energy intakes for both groups because of the greater fat and carbohydrate content of imported foods (Table 4.2.4). The contribution of fat to the total energy content of the diets of both Baffin Inuit and Sahtu Dene-Métis women was 35% when both traditional foods and market foods were considered (Table 4.2.5). The contribution of fat to the total daily energy intake in these groups (35%) is similar to recent provincial data.

- *Imported foods contribute more to daily energy intakes because they have greater fat and carbohydrate content. Fat from all foods contributes about 35% to the dietary energy of both Baffin Inuit and Sahtu Dene/Métis women.*

TABLE 4.2.5

Percent of dietary energy as fat for Arctic Indigenous women 20–40 years of age (Kuhnlein *et al.* 1995b, 1996a)

	Baffin Inuit (n=217)[1]	Sahtú Dene/Métis (n=142)
% energy as total fat in diet	35.0	34.9
% energy as fat derived from:		
– traditional food	28.5	12.1
– market food	71.5	87.2

[1] *Number of dietary recalls*

4.2.5.2 Lipids

Dietary lipids have a number of nutritional functions; they serve as a concentrated source of energy, act as carriers of fat-soluble vitamins, and are a source of essential fatty acids. Fish and marine mammals, which are a rich lipid source, form a significant portion of the diet of Aboriginal Northerners.

Fat obtained from traditional Inuit food is associated with the consumption of sea mammals. Ringed seal blubber and the muktuk available from small whales are the largest sources of fat; polar bear and walrus, while also high in fat, are generally less significant sources. For Dene and Métis, fats in traditional food are obtained primarily from fish, waterfowl, and small mammals, such as beaver. In the market portion of the diet of both areas, fat is provided in bannock, as a medium for frying, in desserts and other baked goods (Kuhnlein *et al.* 1995b, 1996a), and in infant foods and snack foods. Table 4.2.6 summarizes some of the major sources of lipids (fat) in certain country foods.

Saturated fatty acids and monounsaturated fatty acids are present in high amounts in animal fats, including those found in milk. Although these two classes of fatty acids are important sources of energy, they are not considered as essential fatty acids, because both plants and animals including humans have the ability to synthesize them endogenously. Many health regulatory agencies, including Health Canada, have recommended that intake of saturated fatty acids should not exceed 10% of total dietary energy (11g/1000 kcal) because of the recognition that an excess of saturated fatty acids in the diet is an important risk factor related to cardiovascular disease.

Foods of marine origin including fish, mammals, shellfish and crustaceans provide the highest concentration of docosahexaenoic acid (DHA) and eicosapentaenoic acid (EPA), both members of the long-chain n-3 polyunsaturated fatty acids (PUFA), commonly referred to as omega-3 fatty acids. DHA and EPA are not easily found in imported foods (Bang *et al.* 1980, Appavoo *et al.* 1991, Kuhnlein *et al.* 1991). High concentrations of omega-3 fatty acids and low concentrations of saturated fatty acids were found in the blood phospholipid of Québec Inuit underscoring the significant proportion of the marine species in their diet (Dewailly *et al.* 1994b). For this group, the sum of the various omega-3 fatty acids amounted to about 10% of the total blood phospholipid (Table 4.2.7). Levels among the Québec Inuit population surveyed did not vary according to coastal region, but did vary according to sex and age group, with higher levels of omega-3 fatty acids in females and the elderly. This may indicate that females and older adults are still following a traditional Inuit diet, but that the diets of the younger generation are composed of an increasing proportion of imported food, which is low in omega-3 fatty acids.

Although no Canadian study has been conducted with regard to the general population, the blood phospholipid EPA and DHA levels reported for some United States subjects (Yamori *et al.* 1985) are less than one tenth of the levels reported in Inuit. For Japanese fishermen, EPA and DHA levels (Yamori *et al.* 1985) are comparable to the levels found in the Canadian Inuit population. For Québec's North Shore fishermen the levels are at the half-way point between that of urban North Americans and the Inuit (Table 4.2.7) (Dewailly *et al.* 1991).

A diet rich in fish and marine mammals has been linked to a lower incidence of ischaemic disease (i.e. lack of blood flow due to occlusion of an artery) in Greenland (Dyerberg *et al.* 1975) and in Japan (Yamori *et al.* 1985). Indeed for Inuit living along the shore of Hudson Bay, death owing to ischaemic illnesses is very rare, if not absent. The high ratio of EPA/AA (AA: arachidonic acid, a n-6 fatty acid), is associated with a of low risk of ischaemic disease (Hirai *et al.* 1987). The absence of thrombotic disease (angina, heart attack) despite other risk factors present (smoking, stress, obesity) merits further investigation with a view to clarifying the exact role of eating habits. Omega-3 type fatty acids measured in the bloodstream were associated with a significant increase in high density lipoprotein cholesterol and a decrease in triglycerides. These two factors are extremely important in protecting against heart disease and thrombotic disorders. Essential fatty acids, particularly DHA and AA, are also especially important for normal fetal and infant growth and development, and for brain and visual acuity. Birth weight of babies and gestational age may both be increased by omega-3 fatty acids (Olsen *et al.* 1992). In well-nourished women, an average of approximately 2.2 g of long-chain polyunsaturated fatty acids are deposited in maternal and fetal tissues each day throughout pregnancy.

- *Dietary lipids are a concentrated source of energy, act as carriers of fat-soluble vitamins, and are a source of essential fatty acids (polyunsaturated fatty acids that are essential to health but cannot be synthesized by the human body). Fish and marine mammals which form a significant portion of the diet of Aboriginal Northerners contain many n-3 polyunsaturated fatty acids which are not easily found in imported foods. Omega-3 fatty acids are found at high levels in fish and marine mammal tissues and have been associated with a decreased incidence of thrombotic and ischaemic disease.*

4.2.5.3 Minerals

Nutrient composition studies on arctic traditional foods are few, but minerals routinely reported are iron (Fe), sodium (Na), and calcium (Ca) (Health and Welfare Canada 1988, Helms 1985, and Nobmann *et al.* 1992). Original research on nutrient composition of arctic food is reported in Farmer and Neilson (1967, 1970), Heller and Scott (1967), Hoppner *et al.* (1978), and Mann *et al.* (1962). Recently, publications on nutrient composition of Baffin Inuit food and Sahtú Dene/Métis food have included phosphorus (P), potassium (K), zinc (Zn), magnesium (Mg), and manganese (Mn) as well as calcium (Ca), iron (Fe) and sodium (Na). Values for chromium (Cr), cobalt (Co), barium (Ba), molybdenum (Mo), tin (Sn), strontium (Sr), titanium (Ti), nickel (Ni), silicon (Si), and vanadium (V) are also given (Kuhnlein and Soueida 1992, Kuhnlein *et al.* 1994). Recent reports on the mineral intake from diets of Arctic peoples are given in Doolan *et al.* (1991), Lawn and Langner (1994), Wein (1994a, 1994b), Kuhnlein *et al.* (1995b), and Morrison *et al.* (1995). Table 4.2.6 summarizes qualitative information regarding the mineral content

TABLE 4.2.6

Sources of nutrients in various traditional foods

LEGEND +++Excellent source ++Good source +Poor source Blank space indicates data not available

Food source	Protein	Iron	Vitamin A	Vitamin B	Vitamin C	Fat	Calcium	Omega-3 fatty acids
Dene Traditional Foods[7]								
Caribou								
meat	+++	++		++		+	+	
blood	++	+++		+				
liver	+++	+++	+++			+	+	
intestine	++	++		+		+++	+	
bone marrow	+	++	+	+		++		
stomach contents	+	++	++	+		+	+	
Moose								
meat	+++	++	+	++		+	+	
liver	++	+++	+++			+	+	
blood	++	+++		+				
Fish								
meat	+++	+	+	+++		+	+	
head	+++	+	+	+++		+	++	
liver	+++	++	+++		+++			
eggs	+++	+++		+++	+++	+++	+	
Muskrat								
meat	+++	+++		+++	++	+		
Beaver								
meat	+++	+++		+++[1]				
liver	+++	+++	+++					
tail	+++	+				+++		
feet	+++	++				+++		
Rabbit/Hare								
meat	+++	+++		+++[2]			+	
Duck								
meat	+++	+++		+++[3]				
eggs	++	+++		++		+		
Ptarmigan/Grouse								
meat	+++	+++		+++				
Wild plant greens[4]		++	+++	+	+++		+++	
Wild berries[5]		+			+++			
Inuit Traditional Foods[8]								
Beluga								
meat	+++	+++				+		
blubber	++	+				+++		++
skin	+++		+			+		++
Seal								
intestine	+++	++				+		
liver	+++	+++	+++		++	+		
flippers		+++						
blubber	+	++	+++		++	+++		+++
meat	+++	+++		+++		+		
brain	+++	++				+		
eyes	+++	+++	+++			+		
Walrus								
meat	+++	+++		+++		+		
blubber	+++	+	+++			+++		++
skin	+++	+	+			+		
flippers		+[6]						
Narwhal								
meat	+++	+++				++		
blubber	+	++	+++			+++		++
flippers	+++		++					
skin	+++	+	+++					
Polar bear								
meat	+++	+++						
fat	+	+	+++			+++		++
Muskox								
meat	+++	+++		++	+			
Ground squirrel								
meat	+++	+++	+++	++		+		
Grizzly bear								
meat	+++	+++		++		++		

(continued next page)

TABLE 4.2.6 *(continued)*

Sources of nutrients in various traditional foods

LEGEND +++Excellent source ++Good source +Poor source Blank space indicates data not available

Food source	Protein	Iron	Vitamin A	Vitamin B	Vitamin C	Fat	Calcium	Omega-3 fatty acids
Seaweed		++	+++	++	+++		++	
Seafood								
scallop	+++		+	+++	+			
shrimp	+++	++	+	+++				
sea cucumber	+++		+	+++	+		+	
Mussels	+++	+++	+	+++	+++		+	
Clams	+++	+++	+	++	+++		+	
Crabs	+++	+					+	
Arctic char								
meat	+++	+	+	+++		+		++
skin	+++	+++				+++	+++	
head	+++	+	+	+++		+	++	
eggs	+++	+++	+	+++	+++	+++	+	

[1] *Excellent source of niacin; good source of riboflavin; poor source of thiamin*

[2] *Excellent source of niacin; poor source of thiamin and riboflavin*

[3] *Raw meat*

[4] *For example, lamb's quarter, dock, dandelion, fireweed. Wild plants are also an excellent source of fibre*

[5] *For example, currents, strawberries, blueberries, gooseberries and cranberries. Good source of carbohydrates and excellent source of fibre*

[6] *Measured in skin; blubber is a fair source of iron*

[7] *Mackenzie Regional Health Service and the Dene Nation. GNWT 1996a*

[8] *Baffin, Inuvik, Keewatin Health Boards. GNWT 1996b*

of various country foods and shows that many foods are a significant source of iron in particular. Table 4.2.8 provides a quantitative summary of some essential mineral elements in selected arctic food items.

In comparing the traditional portion of contemporary diets of Arctic Indigenous peoples to the portion derived from market foods, it is evident that the majority of essential minerals in the total diets of all age groups is from traditional animal species. The nutrient density (nutrients per caloric intake: $g \cdot kcal^{-1}$) of minerals is significantly greater in the traditional portion of the diet for Arctic men and women (Kuhnlein 1995, Kuhnlein *et al.* 1995b, Kuhnlein *et al.* 1996a). Table 4.2.4 illustrates the results of a study comparing Baffin Inuit women with Sahtu Dene/Métis women. Traditional diets provide significantly more (and nutritionally adequate) iron and zinc in both groups. Similarly, in Sanikiluaq, country food provided over 80% of daily iron and zinc in 1992–93 although residents had low intakes of calcium (Wein 1995c). Wein (1995b) has also shown that traditional food contributes a substantial portion of overall nutrients for three First Nations in the Yukon.

Because of extensive use of traditionally harvested fish and game, the intake of mineral nutrients is sufficient, with the possible exception of calcium which has been shown to be below recommended dietary intakes for some Inuit, Dene/Métis and Yukon First Nation communities (Wein 1994a, 1995b, Kuhnlein *et al.* 1995b, 1996a). Deficiency symptoms for calcium include poor bone and teeth integrity in adults and children; substandard intakes during development result in poor growth. It is reasonable to conclude that if consumption of traditional food resources, particularly fish and wildlife, were discontinued, the mineral nutrition of most Arctic populations would be compromised to such an extent that nutritional deficiencies could occur.

Foods of marine and freshwater origin, which make up a significant portion of many Aboriginal diets, also provide the highest concentrations of selenium, an essential trace mineral (Lunde 1970, Thorn *et al.* 1978, Lane *et al.* 1983). Muktuk is a particularly rich source of selenium (Dewailly *et al.* 1994b). High concentrations of selenium have been measured in groups consuming these foods (Dewailly *et al.* 1994b, Svensson 1992). For example, the average selenium concentration in the bloodstream of Inuit between 18 and 74 years of age was $2.0\ \mu mol \cdot L^{-1}$ versus 0.4 to $1.5\ \mu mol \cdot L^{-1}$ in the non-Aboriginal southern Québec population. The concentration of selenium in the bloodstream of individuals who consumed muktuk less than once a month was found to be $1.76\ \mu mol \cdot L^{-1}$, whereas for those individuals who consumed muktuk every week, the figure was $2.25\ \mu mol \cdot L^{-1}$ (Dewailly *et al.* 1994B).

TABLE 4.2.7.

Sum of omega-3 fatty acids and EPA/AA ratios in Nunavik (Inuit), southern Québec (Caucasian) and lower North Shore (Caucasian).

Area	Sample Size	Sum of omega-3 (%)	EPA/AA ratio	Reference
S.Québec–Caucasian	40	3.4	0.07	Dewailly 1995
Lower North Shore–Caucasian	156	5.6	0.14	Dewailly *et al.* 1991
Nunavik–Inuit	492	9.8	0.53	Dewailly *et al.* 1994

TABLE 4.2.8.

Some essential elements in selected traditional arctic food (per 100g fresh wt.)[1] (Kuhnlein and Soueida. 1992, Kuhnlein *et al.* 1994).

Food	Calcium (mg)	Phosphorus (mg)	Sodium (mg)	Iron (mg)	Zinc (mg)	Copper (mg)	Magnesium (mg)	Manganese (mg)
SEA MAMMALS								
Ringed seal meat	12	251	114	22	3	0.2	27	0.03
Narwhal muktuk	5.5	172	102	0.4	7	0.1	14	0.01
Walrus blubber	3.7	31	67	1.5	0.4	0.2	2.7	0.01
OTHER MAMMALS								
Caribou meat	9	397	88	8	5.7	0.7	45	0.10
Moose meat	18.6	412	97	9	7.5	0.49	51	0.05
Muskrat meat	20	180	110	11	2.6	0.15	26	0.20
Beaver meat	12	233	133	6.9	5.4	0.18	28	0.13
FISH								
Arctic char flesh	15.6	281	72	0.7	0.6	0.1	29	0.01
Loche (burbot) flesh	38	195	85	0.5	0.7	0.04	21	0.03
Whitefish flesh	39	444	89	1.4	0.8	0.08	48	0.05
Whitefish head flesh	544	365	116	4.8	5.0	0.07	24	0.18
BIRDS								
Black scoter flesh	6.1	270	72	8	2.5	0.74	29	0.07
Canada goose flesh	23	570	200	12	5	0.7	60	0.14
Ptarmigan flesh	6.1	318	51	9	1	0.6	36	0.10
PLANTS								
Kelp	160	60	610	0.9	0.5	16	120	0.06
Blueberries	13	14	0.3	0.2	0.3	0.08	9.5	0.32

[1] *Values are approximate means of food as consumed following various preparations*

Selenium and other anti-oxidants such as retinol are now recognized anticarcinogens. Experimentally, selenium and vitamin E (alpha-tocopherol) have both been shown to reduce the toxic response in the nervous system associated with exposure to mercury (WHO 1990). However, the efficacy of selenium as an antidote against mercury toxicity in humans remains controversial (Magos 1991).

Other nutrients also interact in different ways with environmental pollutants. Zinc, iron, calcium, selenium, cobalt, manganese, pyridoxine (B_6 complex vitamin), ascorbic acid and dietary protein all interact with cadmium while zinc, iron, calcium, selenium, cobalt and chromium can all influence the toxicity of lead (dietary deficiencies increase lead toxicity) (Chowdury and Chandra 1987). Calcium intake through the diet has also been inversely associated with blood lead levels in children 1–11 years old. Vitamin D_3 (cholecalciferol), while stimulating the absorption of calcium, phosphate and other essential elements, also increases the absorption of some toxic metals (cadmium, lead) and radioactive isotopes (strontium, cesium) (Moon 1994).

- *If consumption of traditional food resources — particularly fish and wildlife — were discontinued, the mineral nutrition of most Arctic populations would be compromised to such an extent that nutritional deficiencies could occur.*

4.2.5.4 Vitamins

Traditionally harvested fish and game are also rich in many vitamins, particularly the fat-soluble vitamins and the vitamin B-complex. Dietary surveys have been conducted recently among several Indigenous groups in the north. Data from these surveys, together with information from the Nutrition Canada Survey (Health and Welfare Canada 1976), indicate the importance of northern traditional foods as sources of certain vitamins for the Inuit, the Dene, and probably the Cree.

Reports on vitamin contents of certain traditionally harvested fish and game are contained in Helms (1985), Health and Welfare Canada (1988) and Nobmann (1992). However, these references depend on published databases of southern food, and often have not reported values in species harvested from the Arctic. Table 4.2.6 summarizes qualitative information on the vitamin contributions of various country foods.

In all populations, vitamin A is consumed preformed as retinol derivatives, which are found in liver, selected fats, and dairy products, and also as the provitamin carotene, which occurs in certain yellow and green vegetables. Retinol contents of arctic traditional food are reported in Kuhnlein and Soueida (1992) and Morrison and Kuhnlein (1993). As vegetables and dairy products are consumed infrequently by many Northerners, the intakes of retinol and carotene from these sources are far below those in southern Canadians. Dietary recall surveys indicate that fats, organ meats and certain fish consumed as components of traditional diets are major sources of retinol. Food containing vitamin A as retinol also probably provides substantial amounts of vitamin D. Fish are major sources of vitamin E in the traditional diet.

Some surveys reveal that low intakes of vegetables and fruit are associated with low intakes of vitamin C, vitamin B_6 and folic acid; however complete composition data for these items in traditional arctic food resources do not exist. Intakes of thiamin, riboflavin, and niacin are reasonably adequate in the north. Fish and game provide substantial amounts of vitamin B_{12} and pantothenic acid, and total intakes of these vitamins are probably higher than in the general Canadian population. In Sanikiluaq, Wein (1995c) found that country food provided over 80% of daily niacin although residents had low intakes of vitamins A and C.

Clinical evidence of deficiencies for individual vitamins and/or minerals have not been documented in Arctic populations, although average dietary intakes for some population groups have been shown to be below dietary standards established by Health Canada. For example, vitamin A has been shown to be below recommended dietary intakes for some Inuit, Dene/Métis and Yukon First Nation communities (Wein 1994a, 1995b, Kuhnlein *et al.* 1995b) although in other studies of Inuit populations, vitamin A intakes have been found to be sufficient to meet Health Canada's dietary standards (Kuhnlein *et al.* 1996a). Evidence of vitamin A deficiency includes a variety of symptoms related to infection resistance and poor vision; substandard intakes of vitamin A during development could result in poor growth.

- *Thiamin, riboflavin and niacin intake in the North are reasonably adequate due to the major contributions of these vitamins from traditional meats. Fish and game contribute substantial amounts of vitamin B_{12} and pantothenic acid. Total intakes of these vitamins are likely higher than in the general Canadian population. Some reports indicate that vitamin A, calcium, and vitamin C may be below recommended intakes.*

4.2.5.5 Protein

Protein is an essential nutrient that contributes amino acids and energy needed to build cells. Protein is also needed to make and maintain blood and other body fluids, some hormones, enzymes, and milk. Inadequate dietary protein results in poor growth and development, low infant birthweight, poor resistance to infection, and generally poor health (Hegarty 1995). In practice, if protein is limited in diets, many other nutrients are also in short supply. For arctic peoples, limiting the supply of traditional meats and fish would drastically reduce their protein intake. In a study comparing the relative contribution of traditional and market foods with the total diet, it was found that traditional food contributed adequate amounts of protein to fulfill the recommended nutrient intakes for both Baffin Inuit and Sahtu Dene/Métis women (Table 4.2.4). Wein (1995c) has shown that in Sanikiluaq in 1992–1993, over 80% of the daily protein requirements were fulfilled by traditional foods. Table 4.2.6 also shows that many country foods are excellent sources of protein.

- *In arctic communities, a significant portion of the protein requirements are fulfilled by traditional foods such that limiting the supply of traditionally harvested meats and fish would drastically reduce protein intake.*

4.3 Environmental Contaminants

Traditional foods are the primary route of human exposure to most persistent environmental contaminants in the Arctic. Food is also the primary non-occupational route of exposure to these contaminants elsewhere in the world (Government of Canada 1991). Earlier chapters of this report have provided details of sources and pathways for these contaminants and the effects on animal populations. The specific concentrations of contaminants are provided for a variety of wildlife food species in Chapter 3. Several examples of specific contaminants in traditional foods are given in the case studies (section 4.4.6). This section provides information on 18 specific contaminants of concern from a human health perspective: their toxic effects, exposure levels, biological effect levels and concentration in human tissues. Whenever possible, probable daily intakes (PDI) are calculated and the major sources and routes of exposure proposed for various subregions of the Arctic. Exposure guidelines, reported as acceptable daily intakes (ADI) or tolerable daily intakes (TDI) are given when available. The process by which ADIs and TDIs are developed is explained in more detail in section 4.4.1.1.

This section emphasizes contaminant exposure of the fetus (through placental transfer) and the newborn (through mother's milk). These two early life stages are generally far more sensitive to the effects of most OCs and metals discussed below. Their sensitivity to low level background radiation is uncertain. For the fetus and newborn, new tissues are being developed and organized into organs, and growth is at its maximum. Intake and absorption of contaminants per kilogram of body weight are also more than double that of adults. Mothers can do little to reduce fetal and infant exposure other than to reduce their own lifetime contaminant intake. The level of concern for the health of the fetus and newborn is evaluated in section 4.4.5.

4.3.1 Persistent Organic Pollutants (POPs)

Nine POPs are reviewed in this section — eight organochlorines (OCs); dichlorodiphenyltrichlorethane (DDT) and its metabolites, toxaphene, dioxins and furans, mirex, chlordane and its metabolites, hexachlorocyclohexane (HCH), polychlorinated biphenyls (PCBs), hexachlorobenzene; and polyaromatic hydrocarbons (PAHs).

Information on contaminant levels in the tissues of northern residents is very recent and rapidly increasing in quantity. Much of the information on arctic population contaminant levels has been developed through and supported by the AES/NCP.

To simplify the comparisons and to focus on intergenerational transfer of contaminants, human milk data for residents of Arctic Canada and southern Canada are presented for all of the contaminants for which results are currently available. When inadequate human milk data is available for a specific contaminant, other tissue data (i.e. adipose) may be provided. Cord blood is also a good indicator of intergenerational transfer of contaminants. Data has recently become available on levels of various OCs in cord blood of newborns from Arctic Canada and is discussed in 4.3.1.11.

In southern Canada, levels of most OCs measured in mothers' milk have declined significantly over the last 15–20 years (Mes 1994). There are currently insufficient data to determine temporal or spatial trends in many human populations living north of 60°N latitude because sampling in these regions has been sporadic or non-existent. Though this discussion will focus on the increased levels of various OCs in mothers' milk, the significant benefits of nursing to both the mother and the infant must also be considered. These benefits and risks are discussed in section 4.4.5.1.

4.3.1.1 DDT and its Metabolites

Regulatory restrictions on the use of DDT in Canada were introduced in the 1960s although it was not deregistered for all uses in Canada until 1985. Its persistence in the environment and its continued entry into Canada via long-range airborne transport has meant that it is detectable in almost all compartments of the ecosystem and in all human tissues.

TABLE 4.3.1

Comparative concentrations of DDE in human milk fat and Greenland omental fat in non-Aboriginal and Aboriginal populations

Location	Year of Sampling	Mean ($ng \cdot g^{-1}$ lipid)	Reference
Southern Canada	1992	222	Newsome *et al.* 1995
Southern Québec	1989/90	340	Dewailly *et al.* 1996a
Lower North Shore Québec	1991	823	Dewailly *et al.* 1991
Nunavik, Northern Québec (Inuit)	1989/90	1212	Dewailly *et al.* 1993a
Greenland[1] (Inuit)	1993	3844	Dewailly, 1995

[1] *omental fat, not milk fat*

The toxic effects of DDT and its two major metabolites DDE and DDD in experimental animals are primarily seen in the nervous system, the reproductive system, and the liver (ATSDR 1994a). DDT has been classified as a "probable" human carcinogen by the US EPA and is a weak estrogen mimic (Soto *et al.* 1992). DDT and its metabolites are stored in fatty tissue and are excreted very slowly and primarily via urine and feces. Because DDT and its metabolites are found in human milk, 'excretion' also occurs during nursing.

For the population in southern Canada the dietary intake of total DDT (including metabolites) has been declining since the 1960s (Figure 4.3.1). This decline has been reflected by a concomitant decline in total DDT in human tissues. Mes (1994) has reported an approximate 10-fold decline in total DDT from 1967 to 1986 in Canadian human milk samples collected in southern Canada (Figure 4.3.1). Samples collected in 1992 have slightly lower total DDT levels (Newsome *et al.* 1995). The TDI for DDT and all of its metabolites is 20 $\mu g \cdot kg\ bw^{-1} \cdot d^{-1}$ (Table 4.4.1).

Levels of total DDT in human tissue in the Arctic are considerably higher than those in southern Canadians, reflecting the greater consumption of high trophic level species of food. Concentrations of DDE (usually referred to as *p,p'*-DDE), are four- to five-fold higher in mothers' milk from Inuit in northern Québec than populations from southern Canada (Table 4.3.1). Nursing women from the north shore of the St. Lawrence who consume large amounts of fish and gull eggs have intermediate levels of DDE in their milk (Table 4.3.1). There is no indication that these levels are declining in arctic populations; however tissue sampling in the north has only occurred relatively recently and there are not sufficient data for trend analyses. Even higher concentrations of DDE are seen in omental fat tissue (i.e. membrane covering and supporting abdominal organs) from Greenlandic Inuit (Table 4.3.1). This difference may be due to increased consumption of contaminated country foods, older age groups in the Greenland study versus the Québec study or tissue specific concentration differences (omental fat vs human milk fat).

Although Québec Inuit mothers have higher levels of DDE in their milk than mothers from southern Canada (Table 4.3.1), infant exposures do not exceed the TDI. A 5 kg Inuit infant consuming 750 mL of human milk per day with 3% fat would take in approximately 5 $\mu g \cdot kg\ bw^{-1} \cdot d^{-1}$. Suppression of reflexes in neonates appears to be associated with levels of DDE in human milk exceeding 4000 $ng \cdot g^{-1}$ lipid however, it has not been substantiated whether DDE is the causative factor (Rogan *et al.* 1986). Elevated levels of DDE in human milk (≥3000 ng/g lipid) have been correlated with a shortening of breast-feeding duration; inhibition of lactation was hypothesized (Rogan *et al.* 1987).

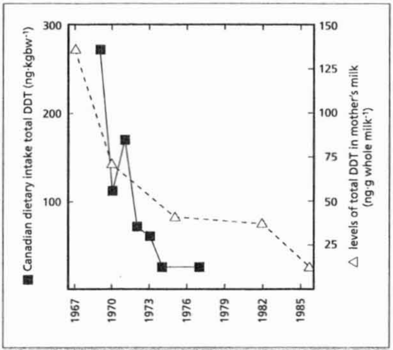


FIGURE 4.3.1

DDT levels in Canadian dietary intake (1969–1977) and in mothers' milk (1967–1986). (Adapted from Conacher and Mes 1995, Mes 1994)

- *Levels of total DDT in human tissue in the Arctic are considerably higher than those in southern Canadians, reflecting the greater consumption of high trophic level species for food. Concentrations of DDE, the major metabolite of DDT are four- to five-fold higher in human milk from Inuit in northern Québec than populations from southern Canada.*
- *Québec Inuit mothers have higher levels of DDE (1200 ng/g lipid) in their milk than mothers from southern Canada but their children do not exceed the tolerable daily intake (TDI).*

4.3.1.2 Toxaphene

Toxaphene (or Camphechlor) is a generic name for a mixture of polychlorinated terpenes and bornanes. It has had only limited use in Canada, however, in the US, toxaphene has been one of the most heavily applied insecticides. Toxaphene enters Canada mainly via long-range airborne transport. Although it is found in human diet and tissue, its historical quantification has been compromised by difficulties in analysis and in the estimation of amounts of the various chemicals that make up toxaphene. As a result, comparisons of published data on toxaphene are difficult.

The research base on toxaphene is limited to the technical product and is difficult to apply to human environmental exposure. Toxaphene accumulates readily in fatty tissues and many of its chemical constituents can be metabolized. Some congeners are retained (Stern *et al.* 1992), primarily an octachlorocamphene (T_2) and a nonochlorcamphene (T_{12}) (corresponds with Parlar standards 26 and 50 respectively). Toxaphene is likely to affect the nervous system and liver (ATSDR 1994b). It is a 'possible' human carcinogen (IARC 1987) based on animal evidence. Little is known about the effects of long-term low-level exposure to toxaphene and the likelihood of effects if the TDI of 0.2 μg·kg bw^{-1}·d^{-1} is exceeded (Table 4.4.1).

In southern North America, toxaphene intakes have been estimated from total diet analyses in the US. Intakes are remarkably uniform across a number of age groups ranging from 2.4 to 5.9 ng·kg bw^{-1}·d^{-1} in 1991 and appear to be declining (FDA 1990, 1991, 1992).

Toxaphene intakes in arctic populations are entirely dependent on the type and amount of country food consumed (toxaphene levels are highest in narwhal and beluga blubber in the Eastern Arctic — see Chapter 3). While seasonal intakes may occasionally exceed the current TDI for toxaphene, the known nutritional, social and cultural benefits of consumption of traditional foods are believed to outweigh the known risks (see section 4.4.6, Case Studies 1, 2, 3). There are few data available on toxaphene in the tissues of Arctic human populations. A study of three Inuit women from northern Québec reported levels of toxaphene in milk of 221 ng/g lipid (Stern *et al.* 1992). This is a very small sample size for any evaluation and may not be representative of the larger population. Infant intakes based on this level in human milk would be 1 μg·kg bw^{-1}·d^{-1}.

- *A study of three Inuit women from northern Québec reported levels of toxaphene in human milk very similar to the levels of DDE. Because of the large uncertainty factor used to derive the TDI for toxaphene there is considerable public concern over toxaphene intakes.*

4.3.1.3 Polychlorinated Dibenzodioxins (PCDDs) and Polychlorinated Dibenzofurans (PCDFs)

Dioxins and furans are two structurally similar families of compounds that include 75 and 135 congeners, respectively. These compounds enter the arctic ecosystem almost exclusively via long-range airborne transport. Seventeen members of these two families of chemicals are highly toxic and contribute most of the toxicity to complex mixtures of dioxins and furans. The seventeen most toxic congeners in both families have chlorine substitutions in the 2,3,7, and 8 positions. These congeners cause a variable and wide range of deleterious effects in laboratory animals. The most toxic dioxin (2,3,7,8–TCDD) is carcinogenic, teratogenic, and capable of causing reproductive impairment in laboratory animals (Environment Canada and Health Canada 1990).

Due to the different relative toxicity of the dioxin and furan congeners a set of international toxicity equivalency factors (TEFs) have been adopted, ranging from 1, for 2,3,7,8–TCDD (the most toxic congener), to 0.001 for the least toxic congener (NATO 1988). The TEF value for each congener is multiplied by the respective congener's concentration and the resulting values summed and expressed as International Toxic Equivalents (2,3,7,8–TCDD TEQs).

The general population is exposed to small amounts of complex mixtures of PCDDs and PCDFs and other organochlorines. An extensive analysis of adipose tissue samples from a number of countries has concluded that almost all humans contain TCDD at concentrations up to, and occasionally greater than, 3 pg·g^{-1} lipid (Travis and Hattemer–Frey 1991). These levels in the general population have not been associated with disease. In a few incidents, humans who have been exposed to very large amounts of a limited number of these compounds have developed chloracne, a skin disorder. There is also evidence that high level exposure to dioxins and furans can cause variations in serum lipid levels; other dermatological effects related to chloracne; microsomal enzyme induction and gastrointestinal alterations (Schulz *et al.* 1990). Other studies of high level occupational exposure have found associations with some types of cancer (Fingerhut *et al.* 1991). Pluim *et al.* (1993) concluded that *in utero* and lactational exposures to PCDDs/PCDFs are capable of affecting the hypothalamic/pituitary/thyroid regulatory system in human infants.

The best documented poisonings by PCDFs in humans are the Yusho and Yu-cheng incidents in Japan and Taiwan, respectively, when rice oil was accidentally contaminated with PCBs from electrical transformer oil and subsequently consumed by local communities. The PCBs were heavily contaminated

TABLE 4.3.2

Comparative concentrations of dioxins, furans, and dioxin-like PCBs in human milk fat in non-Aboriginal and Aboriginal populations

Location	Year of Sampling	Mean[1] (pg·g^{-1} lipid)	Reference
Dioxins and Furans			
Southern Canada	1986–87	15.6	Ryan *et al.* 1993
Netherlands	1990–92	30.2	Koopman-Esseboom *et al.* 1994a
Southern Québec	1989–90	9.6	Dewailly *et al.* 1992
Nunavik, Northern Québec (Inuit)	1989–90	19.0	Dewailly *et al.* 1992
Dioxins, Furans, and Dioxin-Like PCBs			
Netherlands	1990–92	46.2	Koopman-Esseboom *et al.* 1994a
Southern Québec	1989–90	17.9	Dewailly *et al.* 1992
Nunavik, Northern Québec (Inuit)	1989–90	42.3	Dewailly *et al.* 1992

[1] *expressed as 2, 3, 7, 8–TCDD TEQs*

with PCDFs which were the most likely cause of many of the reported effects (Rappe *et al.* 1983, Rappe *et al.* 1984). The mean total consumption of PCDFs by the Yusho and Yu-cheng patients has been estimated to be 0.9 µg·kg bw^{-1}·day^{-1} (Hayabuchi *et al.* 1979) or 3.3 ng 2,3,7,8–TCDD TEQs·kg bw^{-1}·day^{-1} (Ryan *et al.* 1990). The smallest amount of total PCDFs causing chloracne has been estimated to be 0.16 µg·kg bw^{-1}·day^{-1} (Hayabuchi *et al.* 1979).

In 1987 the average daily southern Canadian intake of PCDDs and PCDFs over a lifetime was estimated to be 2.0–4.2 pg 2,3,7,8–TCDD TEQ·kg bw^{-1}·day^{-1} for a 60 kg person (Birmingham *et al.* 1989). Extrapolation from human tissue levels in Canada confirmed this estimated intake (i.e., intakes were likely 1.9 pg·kg bw^{-1}·day^{-1}; Environment Canada and Health Canada 1990). The Canadian TDI for dioxins and furans is 10 pg·kg bw^{-1}·day^{-1} (Table 4.4.1).

The mean PCDD and PCDF concentration in human milk for southern Canadian women in 1986/87 was 15.6 pg· g lipid^{-1} 2,3,7,8–TCDD TEQs, Table 4.3.2 (Ryan *et al.* 1993). Similar levels have been reported in Sweden (Nygren *et al.* 1986) and the US (Schecter *et al.* 1986). In 40 milk samples collected between July 1989 and July 1990 from Inuit women residing in the Nunavik region of Arctic Québec (Hudson Bay, Hudson Straight and Ungava Bay), 2,3,7,8–TCDD TEQs for PCDDs and PCDFs were moderately elevated when compared to the southern Québec non-Aboriginal population (19.0 pg/g lipid vs. 9.6 pg/g lipid, Table 4.3.2) (Dewailly *et al.* 1992). Computer-based predictions of the maximal PCDD/PCDF concentrations to which Inuit newborns are exposed via human milk indicate that lifetime exposures are well below the levels associated with cancer and reproductive impairment in experimental animals (Ayotte *et al.* 1996). Because of the well known benefits of breast-feeding, it has been recommended that mothers in Nunavik continue to breast feed their newborns.

- *PCDDs and PCDFs measured as 2,3,7,8–TCDD TEQs in human milk from Nunavik Inuit women were lower than levels seen in the Netherlands, but were higher than levels in southern Canadian women. It has been recommended that mothers in Nunavik continue to breast-feed their newborns due to the well known benefits of breast-feeding.*

4.3.1.4 Polychlorinated Biphenyls (PCBs)

PCBs are a group of 209 structurally similar congeners that were commercially produced as mixtures. The primary source of PCBs to the Arctic ecosystem is through airborne transport from other countries. Localized release has also occurred due to improper storage, use or accidents at DEW (Distant Early Warning) line sites in the Arctic. Their manufacture and new use is banned in Canada but they are still present in older electrical transformers and at a number of contaminated industrial sites and waste sites.

The interpretation of the toxicity of PCB mixtures is complicated by the varying amounts of the 209 congeners in the mixture and the microcontaminants present (e.g. PCDFs). Individual congeners and mixtures can affect liver function, reproduction, infant birthweights, neurobehavioural development, the immune system and may cause cancer in laboratory animals (ATSDR 1995). Assessing the human health effects of PCBs is very difficult because PCB mixtures used in animal studies or identified in accidental poisonings may contain microcontaminants and undergo extensive "environmental filtering" prior to human exposure. Many of the effects of exposure to PCBs observed in humans (e.g., Yusho and Yu-cheng) reflect exposure to high levels of both PCBs and PCDFs. PCDFs are believed responsible for many of the observed human health effects. For Yusho and Yu-cheng, the earliest toxicological signs included chloracne. Additional generalized adverse health effects included hepatomegaly (enlarged liver), bronchitis and peripheral neuropathy (nervous system damage) (Safe 1987). In Yu-cheng, increased upper respiratory tract infection rates were associated with changes in various immunoglobulins, (i.e. decreased serum IgA and IgM plus increased IgG levels) (WHO 1988). A number of PCB congeners have dioxin-like activity and will be discussed in section 4.3.1.5.

In southern Canada, the estimated adult daily intake for total PCB from diet is 0.008 µg·kg bw^{-1}·day^{-1} (Conacher *et al.* 1994). Intakes by arctic residents consuming large amounts of traditional foods from the aquatic and marine environment are considerably higher. The TDI for PCBs is 1 µg·kg bw^{-1}·day^{-1}. Some

TABLE 4.3.3.

Comparative concentrations of PCBs in human milk fat in non-Aboriginal and Aboriginal populations

Location	Year of Sampling	Mean (ng·g^{-1} lipid)	Reference
Southern Canada	1992	238[2]	Newsome *et al.* 1995
Southern Québec	1989/90	520[1]	Dewailly *et al.* 1992
Southern Québec	1988	770[1]	Dewailly *et al.* 1989
Lower North Shore Québec	1991	2 160[1]	Dewailly *et al.* 1991
Nunavik, Northern Québec (Inuit)	1988	3 600[1]	Dewailly *et al.* 1989
Nunavik, Northern Québec (Inuit)	1989/90	2 900[1]	Dewailly *et al.* 1992

[1] *PCBs as Aroclor 1260*

[2] *Sum of PCB congeners (PCBs as sum of congeners is often one-half PCBs as Aroclor 1260)*

PCBs are selectively stored in fat due to their lipophilicity and their differing degrees of biodegradability. Excretion is primarily through feces and human milk.

While levels of total PCBs have declined since the 1980s in human milk of southern Canadian women (Newsome *et al.* 1995), there is no similar information yet available for Arctic residents. Nor are historical values easy to compare because of changing analytical techniques and quantitation methods (reported PCB concentrations based on an Aroclor 1260 standard are double those based on a sum of PCB congeners). This may explain some of the differences seen in Table 4.3.3 between the 1992 southern Canada PCB level in human milk of 238 $ng{\cdot}g^{-1}$ lipid (sum of congeners) and the 1989–90 southern Québec PCB level of 520 $ng{\cdot}g^{-1}$ lipid (Aroclor 1260, Table 4.3.3).

In human milk sampled from across Canada in 1992, the sum of PCB congeners was 238 $ng{\cdot}g^{-1}$ fat (Newsome *et al.* 1995). The pattern of PCB congeners observed in 1992 samples is similar to that in earlier surveys in Canada (Mes *et al.* 1993) and in studies on human milk conducted in Sweden (Noren 1991) and the Netherlands (Dagnelie *et al.* 1992). PCB levels in serum (2 $\mu g{\cdot}L^{-1}$, mean value) and adipose tissue (0.4 $\mu g{\cdot}g^{-1}$) in Canadians are comparable to those found in other developed countries. Differences in PCB levels between populations may be due to fish consumption patterns (Fiore *et al.* 1989, Humphrey *et al.* 1983). Mothers consuming large amounts of fish, seafood, marine mammals and seabird eggs have significantly higher PCB levels in their blood and milk (Dewailly *et al.* 1989, 1992).

Several studies suggest that PCB levels in human milk fat are significantly elevated in some Inuit mothers (Dewailly *et al.* 1989, 1992). In human milk samples collected from Inuit women residing in the Nunavik region of Arctic Québec (Hudson Bay, Hudson Straight and Ungava Bay), total PCB levels were elevated 4.7 to 5.6-fold when compared to a southern Québec non-Aboriginal population in the same sampling year (Table 4.3.3). Women from the north shore of the St. Lawrence who consume more North Shore fish and country food than women from southern Québec, but less than Inuit, had intermediate levels of PCBs in their breast milk. Based on the 1989 data, the Inuit infant intake of total PCBs (as Aroclor 1260) would be 13 $\mu g{\cdot}kg\ bw^{-1}{\cdot}d^{-1}$. This intake is well above the adult TDI of 1 $\mu g{\cdot}kg\ bw^{-1}{\cdot}d^{-1}$ (Table 4.4.1).

Data obtained from epidemiological studies on infants from Michigan (Jacobson *et al.* 1990, 1992) and North Carolina, US (Gladen *et al.* 1988, 1991, Rogan *et al.* 1986, 1987) suggest adverse neurobehavioural effects from *in utero* exposure to PCBs (calculated as Aroclor 1260 equivalents). High cord blood concentrations were associated with low birth weight, small head circumstance, and short-term memory loss (Jacobson *et al.* 1990, 1992). Birth size among Inuit male infants was inversely related to PCB concentration in mothers' milk (Dewailly *et al.* 1993b). Perinatal exposure to PCBs/dioxins/furans may impair immune responses to infection as suggested by a higher incidence of infectious disease (e.g. ear infections — otitis media) among 1-year-old Inuit with high PCB exposure than among lesser exposed individuals (Dewailly *et al.* 1993c). This may be confounded by a lower seroconversion rate (successful immunization) among Inuits compared with other populations (Dewailly *et al.* 1993c).

It is not clear if PCB exposure is the sole factor leading to neurodevelopmental deficits in the North Carolina or Michigan studies, or if other contaminants or socio-demographic characteristics might also be associated with these results (Ayotte *et al.* 1996). Caution must be used when examining data from the Lake Michigan study in order to assess the health risk for Inuit newborns in the Arctic. The mixture of contaminants to which Lake Michigan infants were exposed may be very different from that found in Arctic ecosystems (industrial sources vs long-range atmospheric transport). In addition, the Lake Michigan population exposure was through fish consumption, while the diet of northern Inuit also includes species at higher trophic levels (e.g. sea mammals). Studies underway in northern Québec should help to clarify these issues.

- *In human milk samples collected from Inuit women residing in the Nunavik region of Arctic Québec, total PCB levels were elevated approximately 5-fold when compared to a southern Québec non-Aboriginal population.*
- *Perinatal exposure to PCBs may impair immune responses to infection as suggested by a 20-fold higher incidence of infectious diseases and ear infections in Inuit infants with high PCB exposure than among lesser exposed individuals. This may be confounded by a lower seroconversion rate (successful immunization) among Inuit compared with other populations.*

4.3.1.5 Dioxins/Furans/PCBs – 2,3,7,8-TCDD TEQs

Several studies have found that a number of PCBs have dioxin-like activity. Thirteen of the 209 polychlorinated biphenyls (PCBs) share certain toxicological properties with the dioxins and furans and have been assigned dioxin-like TEFs (Ahlborg *et al.* 1994). These PCBs can then be included in the total toxic equivalents (2,3,7,8–TCDD TEQs) of a sample (Ahlborg *et al.* 1994). Levels of dioxin-like PCBs such as PCB-77, PCB-126, and PCB-169 are up to several orders of magnitude higher than the levels of 2,3,7,8–TCDD found in human adipose tissue samples from Japan (Tanabe *et al.* 1987, Kannan *et al.* 1988, Kashimoto *et al.* 1989). Results from human adipose tissue and serum collected in the US show that concentrations of dioxin-like PCBs can be more than an order of magnitude higher than concentrations of 2,3,7,8,–TCDD (Patterson *et al.* 1994). The dioxin-like PCBs are an order of magnitude less toxic than 2,3,7,8–TCDD.

Total PCBs were more than five-fold higher in Québec Inuit mothers' milk than the control population. When TEQs for dioxin-like PCBs (77, 126, 169) were factored in, the 2,3,7,8–TCDD TEQ value increased to 42.3 pg·g^{-1} (from 19.0 pg·g^{-1}) for the Inuit milk samples compared to 17.9 pg·g^{-1} (from 9.6 pg·g^{-1}) for the southern Québec samples (Table 4.3.2). For this Inuit population, but not the southern Québec population, the majority of the 2,3,7,8–TCDD TEQs are due to dioxin-like PCBs. Using the standard calculations for infant intake as shown in section 4.3.1.1, the intake of 2,3,7,8–TCDD TEQs by breast fed Inuit infants is 85.5 pg·kg bw^{-1}·d^{-1} for PCDD/PCDF alone and 190 pg·kg bw^{-1}·d^{-1} if dioxin-like PCBs are included.

A large Netherlands study of the effects of contaminants in human milk on child development found levels of 2,3,7,8–TCDD TEQs (PCDD/PCDF/dioxin-like PCBs) in human milk of 46.2 ng TCDD TEQs/kg lipid — very similar to the levels in Inuit human milk (Table 4.3.2). In contrast to the Inuit results, the majority of the 2,3,7,8–TCDD TEQs in the Netherlands study are from the dioxin and furan component. Subtle clinical, immunologic and neurodevelopmental alterations associated with breast-feeding have been reported in the infants/children of the Netherlands cohort (Koopman–Esseboom *et al.* 1994b, Huisman *et al.* 1995, and Weisglas–Kuperus *et al.* 1995). A preliminary report on a study of Canadian Inuit also shows an association between increasing PCB/dioxin/furan levels and immune system deficiencies (Dewailly *et al.* 1993c).

- *Total PCBs were more than five-fold higher in the Québec Inuit mothers human milk than the same control population. When 2,3,7,8–TCDD TEQs for dioxin-like PCBs were factored in, the 2,3,7,8–TCDD TEQ value showed a more marked difference between Inuit and southern controls. For this Inuit population but not the southern Québec population the majority of the 2,3,7,8–TCDD TEQs are due to dioxin-like PCBs.*
- *A large Netherlands study of human milk and child development found levels of 2,3,7,8–TCDD TEQs (PCDD/PCDF/coplanar PCBs) in milk very similar to the levels in Inuit mothers' milk. Subtle clinical, immunologic and neurodevelopmental alterations associated with breast-feeding have been reported in the infants/children in the Netherlands cohort. These immunological findings are consistent with a preliminary study of Canadian Inuit.*

4.3.1.6 Mirex

Mirex has never been registered for use in Canada, however it was imported into Canada as dechlorane from mid-1960 to mid-1970. It also enters via water currents from the US side of the Great Lakes basin. It has since moved extensively via long-range airborne transport and is now found at low levels in human tissues throughout Canada and around the globe (Government of Canada 1991).

The effects of long-term, low-level exposure to Mirex in humans have not been extensively studied; the primary organs in laboratory species affected by Mirex are the liver, kidneys, eyes and thyroid. It is considered a 'possible' human carcinogen (IARC 1987) and also has fetotoxic and teratogenic effects on laboratory species in the 1–6 mg·kg bw^{-1}·d^{-1} range. The Canadian TDI is 0.07 µg·kg bw^{-1}·d^{-1} (Table 4.4.1). Mirex is readily absorbed and stored in fatty tissues. Metabolism to photomirex (a degradation product) is slow and elimination is mainly via feces and human milk.

Mirex levels in human milk are above average for communities consuming high amounts of fish and sea bird eggs (Dewailly *et al.* 1991). Levels in the milk of Inuit from Nunavik, northern Québec are 10 times higher than those in southern Canadian residents (Table 4.3.4). These levels are equivalent to an intake by nursing infants of 0.07 µg·kg bw^{-1}·d^{-1} which is the current Canadian guideline. Intermediate levels of Mirex can be seen in the lower North Shore St. Lawrence population which consumes more fish and seabird eggs than the general population (Dewailly *et al.* 1991). Even higher concentrations of Mirex are seen in omental fat tissue from Greenland Inuit.

- *Levels of Mirex in mothers' milk of Inuit from Nunavik, northern Québec are 10 times higher than those in southern Canadian residents. Even higher concentrations of Mirex are seen in omental fat tissue from Greenlandic Inuit.*

TABLE 4.3.4

Comparative concentrations of Mirex in human milk fat and Greenland omental fat in non-Aboriginal and Aboriginal populations

Location	Year of Sampling	Mean (ng·g^{-1} lipid)	Reference
Southern Canada	1992	1.9	Newsome *et al.* 1995
Southern Québec	1989/90	1.6	Dewailly *et al.* 1993(a)
Lower North Shore, Québec	1991	12	Dewailly *et al.* 1991
Nunavik, Northern Québec (Inuit)	1989/90	16	Dewailly *et al.* 1993(a)
Greenland[1] (Inuit)	1993	153	Dewailly 1995

[1] *omental fat, not milk fat*

TABLE 4.3.5

Comparative concentrations of chlordane and related compounds in human milk fat and Greenland omental fat in non-Aboriginal and Aboriginal populations (Mean, ng·g^{-1} lipid)

Location	Year	Chlordane (*cis* and *trans*)	Cis-nonachlor	Trans-nonachlor	Oxy-chlordane	Reference
Southern Canada	1992	0.37	2.89	17.5	13.4	Newsome *et al.* 1995
Southern Québec	1989/90	ND[1]	—[2]	—	—	Dewailly et.al.1993a
Lower North Shore, Québec	1991	ND	—	—	—	Dewailly *et al.* 1991
Nunavik, Northern Québec (Inuit)	1989/90	3.7	—	—	—	Dewailly *et al.* 1993a
Greenland[3] (Inuit)	1993	11.6	3.12	1463	862	Dewailly 1995

[1] *ND = not detected*

[2] *not analysed/reported*

[3] *omental fat, not human milk fat*

4.3.1.7 Chlordane, Oxychlordane and *Cis/Trans*nonachlor.

Technical chlordane is a mixture of chlordane, nonachlor, heptachlor and other chlordane isomers. It is not currently registered for use in Canada and enters the Arctic ecosystem primarily via long-range airborne transport. High dose exposures affect the neurological and immune systems. Long-term exposures are likely to affect the liver (WHO 1984). Chlordane has been classified as a probable human carcinogen by the US EPA. Excretion of chlordane is primarily through feces and human milk. The TDI for chlordane is 0.05 µg·kg bw^{-1}·d^{-1} (Table 4.4.1). There are no estimated intakes for Canadians for chlordane and its metabolites however, US estimates for southern continental populations indicate that only very small amounts are consumed, 0.002–0.0051 µg·kg bw^{-1}·d^{-1} (Gunderson 1988).

Chlordane and the compounds related to it accumulate in fat and are found in human tissues. In general, only small amounts of chlordane are found in tissues. However, tissues often contain relatively larger amounts of transnonachlor and the metabolite oxychlordane (Table 4.3.5). Inuit mothers' milk from Nunavik, northern Québec contained 10 times higher levels of chlordane than was seen in southern Canadian residents (Table 4.3.5) however Inuit infant exposures during nursing are 0.016 µg·kg bw^{-1}·d^{-1}, which is below the TDI. Even higher relative levels of transnonachlor (80-fold) and oxychlordane (64-fold) are seen in omental fat from Greenland Inuit but this may be due to several factors as indicated previously for DDE.

- *The milk of Inuit mothers from Nunavik, northern Québec contain ten times higher levels of chlordane than was seen in southern Canadian residents. Inuit infant exposure resulting from nursing is below the TDI. Even higher relative levels of transnonachlor and oxychlordane have been reported in omental fat from Greenland Inuit.*

4.3.1.8 Hexachlorocyclohexanes (HCHs)

HCHs are a group of organochlorine pesticides: α-, β-, γ- (more commonly known as Lindane), delta and epsilon are the most stable isomers. Canada still has some minor registered used for Lindane. Like other OCs, most human exposure to HCHs comes from food consumption. HCHs, especially β-HCH accumulate readily in fatty tissues and are excreted slowly via feces, human milk and urine (WHO 1991a).

γ-HCH is the most toxic of the HCH isomers. Excessive exposure can affect the liver, the nervous system, the kidney, the reproductive system, and perhaps the immune system (WHO 1991a). IARC (1987) classifies it as 'possibly' carcinogenic to humans. No effects have been reported in populations exposed to low level environmental concentrations. Exposure levels for HCH via diet in Canada and several other countries were approximately 0.005 µg·kg bw^{-1}·d^{-1} (α-HCH), 0.0003 µg·kg bw^{-1}·d^{-1} (β-HCH) and 0.03 µg·kg bw^{-1}·d^{-1} (γ-HCH) (Gorchev and Jelinek 1985). The TDI for all HCHs is 0.3 µg·kg bw^{-1}·d^{-1}.

Levels of α-, β- and γ-HCH measured in body fluids and tissues in the general population in Europe, Canada and the US can be found in Table 4.3.6. These more recent "background levels" of HCHs in blood, serum, plasma, milk and adipose tissues are relatively low compared to values reported during the 1960s and 1970s, and much lower than levels reported from countries with extensive current usage.

TABLE 4.3.6

Typical levels of HCHs in body fluids or tissues in the general population (Europe, Canada, US) (WHO 1991a, 1991b, Jensen and Slorach 1991)

	Range of average reported HCH levels[1]		
Body fluid/tissue	**α–HCH**	**β–HCH**	**γ-HCH**
Blood/serum/plasma (µg·L^{-1})	<0.1–0.4	0.3–2.0	0.1–0.9
Adipose tissue (ng·g^{-1} lipid)	5–100	210–600	<10–40
Milk, whole (µg·L^{-1})	<1–5	<1–30	<1
Milk, (ng·g^{-1} lipid)	10–40	10–500	<1–10

[1] *In countries where there is a current high use of HCHs, including the technical grade, reported concentrations are considerably higher (e.g. α–HCH levels of up to 3.5 µg·L^{-1} in blood; α-HCH in the range 7–23 µg·L^{-1} in whole milk in India and Iraq; ß-HCH median milk levels of 1430, 4600, 6600 µg·kg^{-1} lipid in Turkey, India, and China).*

Because of its persistence, β-HCH is found at the highest level of the three isomers reported in the southern Canadian population (Table 4.3.7). There are no data for arctic populations, except one report from Greenland where β-HCH levels in omental fat were five times higher than southern Canadian levels in human milk fat (Table 4.3.7). As previously indicated, any comparison of contaminant levels in omental fat and human milk fat may be explained by differing age/sex, tissue type or country food consumption in the two populations. However, it is reasonable to assume β-HCH values in human milk from the Arctic would be in the 30 ng·g^{-1} lipid range.

Infants may constitute a sub-population at special risk. The β-HCH levels in some human milk samples in other parts of the world exceed the TDI of 0.3 μg·kg bw^{-1}·d^{-1}, occasionally in some communities. The β-HCH concentrations in the blood of babies lie within the same range as those in the mothers. The exposure of babies resulting from present β-HCH concentrations found globally in human milk is a matter of concern but is not a reason to stop promoting breast-feeding (WHO 1991).

- *There are no data for arctic populations except for one report from Greenland where b-HCH levels in omental fat were five times higher than southern Canadian levels in human milk fat.*

TABLE 4.3.7

Comparative concentrations of HCHs in human milk and omental fat in non-Aboriginal and Aboriginal populations (ng·g^{-1} lipid)

Location	Year	α–HCH	β-HCH	γ-HCH
Southern Canada[1]	1992	0.31	22.6	1.03
Greenland[2] (Inuit)	1993[3]	—	108.93	—

[1] *Newsome et al. 1995*

[2] *Dewailly (personal communication)*

[3] *omental fat, not human milk fat*

4.3.1.9 Hexachlorobenzene (HCB)

HCB was used in Canada up to 1972 as an anti-fungal agent for various seed crops. Currently, major releases of HCB in Canada are as by-products of the manufacture and through the application of chlorinated solvents and pesticides (Government of Canada 1993). HCB is still widely used elsewhere in the world and a major source in the Arctic is airborne transport and deposition.

Exposure to HCB causes a wide range of effects in laboratory animals ranging from liver pathology, skin lesions (*porphyria cutanea tarda* in humans), behavioural and neurological changes, reproductive effects in primates and effects on the immune system (Government of Canada 1993). In Turkey, HCB-treated grain, improperly used for food, resulted in poisoning in adults and infants. Perinatal exposure was associated with increased infant mortality. The TDI for HCB is 0.27 μg·kg bw^{-1}·d^{-1} (Table 4.4.1).

HCB accumulates in body tissues and human milk has been found to contribute very significantly to human exposure. Levels of HCB in Inuit mothers' milk from Nunavik, northern Québec are five to nine times higher than levels seen in southern Canadian mothers' milk (Table 4.3.8). Infant intakes of HCB from breast-feeding in the Nunavik region in 1989/90 are estimated at 0.6 μg·kg bw^{-1}·d^{-1}which is greater than the adult TDI for HCB of 0.27 μg·kg bw^{-1}·d^{-1}.

- *Levels of HCB in Inuit mothers' milk from Nunavik, northern Québec are five to nine times higher than levels seen in southern Canadian mothers' milk. Infant intakes of HCB from nursing exceed the TDI.*

4.3.1.10 Polyaromatic Hydrocarbons (PAHs)

PAHs are a family of ring structured compounds that do not include chlorine in their formulae. Several PAHs are carcinogenic to animals. While PAHs can enter the Arctic from remote locations (e.g., via industrial activity, forest fires), the primary sources of human exposure are local. The greatest pathways of exposure to PAHs for the general population are inhalation of tobacco smoke (active or passive), wood smoke and contaminated air, and the ingestion of PAHs through consumption of smoked, fried or broiled food. For nonsmokers, food is the main source of exposure (99% of total benzo[a]pyrene (B[a]P) exposure), with air a much less important source (0.9% of total B[a]P exposure).

Benzo[a]pyrene, one well-studied PAH, is a carcinogen in animals and is probably carcinogenic to humans (IARC 1987). PAHs are readily absorbed and can be metabolized with relative ease in the liver and kidney. They are excreted in bile, feces and urine. The total daily potential exposure of carcinogenic PAHs for adult males is estimated to be 3 μg (median), and may be as high as 15 μg. For smokers, exposure levels may be twice as high. Exposure to PAHs through cigarette smoke has been associated with reduced fecundity and low fetal birth weight

TABLE 4.3.8

Comparative concentrations of HCB in human milk fat in non-Aboriginal and Aboriginal populations

Location	Year of Sampling	Mean (ng·g^{-1} lipid)	Reference
Southern Canada	1992	14.5	Newsome *et al.* 1995
Southern Québec	1989/90	28	Dewailly *et al.* 1993a
Lower North Shore Québec	1991	42.3	Dewailly *et al.* 1991
Nunavik, Northern Québec (Inuit)	1989/90	136	Dewailly *et al.* 1993a

(Weinberg *et al.* 1989) and is thought to be a major factor in lung cancer.

The daily intake of PAHs from food is estimated to be 2–20 μg in southern Canada (NRC 1983, Pucknat 1981). PAH concentration in food depends on both the method of preparation and the origin of the food. Barbecuing foods increases their PAH concentration. Smoked and cooked fish and meats are higher in PAHs than uncooked products. Despite considerable potential dietary exposure to PAHs in smoked food, there is little risk to health from this route of intake. Inhalation of PAHs from tobacco smoke (active or passive) is associated with a significant risk to health.

There are no data available on PAH levels in human milk, fat or blood of Arctic residents.

- *Despite considerable potential dietary exposure to PAHs in smoked food, there is little risk to health from this route of intake.*

4.3.1.11 OCs in Newborn Cord Blood

The exposure of the developing fetus to maternal contaminants through the placenta is an important route of exposure during critical stages of development. Recently, data on contaminants in newborn cord blood from several Canadian Arctic and southern populations has become available (Dewailly *et al.* 1996b, Walker *et al.* in prep.). Comparison of the data in Tables 4.3.9 and 4.3.10 indicates that Inuit populations have higher geometric mean concentrations of five contaminants and approximately equal concentrations for the other six contaminants, as well as a greater proportion of detected analytes compared with the Dene/Métis peoples in the central Arctic or the non-Aboriginal populations. This is expected as Inuit consume more marine mammal fats, which have elevated OC residues (Kuhnlein *et al.* 1995c).

Any comparison of maternal and newborn cord blood concentrations of contaminants needs to consider that the important determinants of contaminant levels are age of the mother, number of previous children, and number of children breast fed as well as amount and type of country food consumed. Even though these comparisons are as yet incomplete it is still possible to compare some of the contaminants which show the highest levels and greatest

TABLE 4.3.9

Levels of contaminants in the cord blood ($\mu g \cdot L^{-1}$) of non-Inuit newborns from Arctic and southern Canada (sampled in 1993–1995)

Contaminant	Population	N	Mean[1] ($mg \cdot L^{-1}$)		Range	Proportion detected (%)
			Arithmetic	Geometric[2]		
Aldrin	Dene/Métis NWT[3]	47	0.01	0.01	0.01–0.01	0
	Non-Aboriginals NWT[3]	125	0.01	0.01	0.01–0.01	0
	Gen. Pop. S.Que[4]	502	0.01	0.01	0.01–0.02	0.8
ß–HCH	Dene/Métis NWT	47	0.02	0.02	0.02–0.04	2
	Non-Aboriginals NWT	125	0.15	0.04	0.02–7.49	30
	Gen. Pop. S.Que	502	0.05	0.02	0.02–10.48	30
α–chlordane	Dene/Métis NWT	47	0.01	0.01	0.01–0.01	0
	Non-Aboriginals NWT	125	0.01	0.01	0.01–0.01	0
	Gen. Pop. S.Que	502	0.01	0.01	0.01–0.07	0.6
γ–chlordane	Dene/Métis NWT	47	0.01	0.01	0.01–0.01	0
	Non-Aboriginals NWT	125	0.01	0.01	0.01–0.02	0
	Gen. Pop. S.Que	498	0.01	0.01	0.01–0.07	1
Cis–nonachlor	Dene/Métis NWT	47	0.01	0.01	0.01–0.01	2
	Non-Aboriginals NWT	125	0.01	0.01	0.01–0.02	0
	Gen. Pop. S.Que	502	0.01	0.01	0.01–0.29	3
p,p'–DDE	Dene/Métis NWT	47	0.32	0.25	0.05–1.12	100
	Non-Aboriginals NWT	125	0.63	0.35	0.06–9.25	100
	Gen. Pop. S.Que	502	0.66	0.44	6.07–14.92	100
p,p'–DDT	Dene/Métis NWT	47	0.02	0.02	0.02–0.02	0
	Non-Aboriginals NWT	125	0.04	0.02	0.02–0.70	11
	Gen. Pop. S.Que	502	0.03	0.02	0.01–0.84	32
Hexachlorobenzene	Dene/Métis NWT	47	0.08	0.06	0.01–0.32	85
	Non-Aboriginals NWT	125	0.05	0.04	0.01–0.12	90
	Gen. Pop. S.Que	502	0.05	0.04	0.01–1.40	99
Mirex	Dene/Métis NWT	47	0.01	0.01	0.01–0.01	0
	Non-Aboriginals NWT	125	0.01	0.01	0.01–0.02	0
	Gen. Pop. S.Que	502	0.01	0.01	0.01–0.03	2
Oxychlordane	Dene/Métis NWT	47	0.01	0.01	0.01–0.03	4
	Non-Aboriginals NWT	125	0.01	0.01	0.01–0.07	10
	Gen. Pop. S.Que	502	0.01	0.01	0.01–0.05	21
Transnonachlor	Dene/Métis NWT	47	0.02	0.01	0.01–0.05	21
	Non-Aboriginals NWT	125	0.02	0.01	0.01–0.10	26
	Gen. Pop. S.Que	502	0.02	0.01	0.01–0.17	66
PCBs (as Aroclor 1260)	Dene/Métis NWT	47	0.46	0.24	0.01–2.32	83
	Non-Aboriginals NWT	125	0.42	0.28	0.01–1.89	90
	Gen. Pop. S.Que	502	0.63	0.54	0.01–3.91	99

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[1] *When the sample contained a nondetectable concentration, the mean was calculated using half of the detection limit*

[2] *Both arithmetic and geometric means are presented for comparative purposes. Geometric means are less effected by a few extreme values.*

[3] *Dene/Métis or Non-Aboriginals, Northwest Territories, Walker* et al. *in prep.*

[4] *General Population, Southern Québec, Dewailly* et al. *1996b*

differences between the groups to see if previous information on country food consumption supports the contaminant patterns seen. Based on the geometric mean, HCB, DDE and PCBs had the highest concentrations in newborn cord blood and are plotted in Figure 4.3.2.

The concentrations of both PCBs and DDE were two to four-fold higher in Inuit than in Dene/Métis or non-Aboriginals from the NWT or southern Québec (Table 4.3.9, Table 4.3.10). HCB levels in newborn cord blood were two to three times higher in Inuit than in the other population groups. Inuit from Nunavik have PCB and DDE levels that are double those seen in north-central NWT Inuit (Figure 4.3.2). HCB levels are very similar between these two groups (Figure 4.3.2). The difference between Inuit from northern Québec and Inuit from the north-central NWT deserves further study and may be due to the amount and type of country food consumed, age of mothers, number of children, length of breast-feeding, or number of children breast-fed.

Currently, there are no guidelines for levels of PCBs or other OC contaminants in newborn cord blood. Health Canada has developed a maternal blood level of concern for PCBs of 5 $\mu g \cdot L^{-1}$. As cord blood has fifty per cent lower level of lipids a level of concern for PCBs in cord blood would be 2.5 $\mu g \cdot L^{-1}$. Based on the mean and range of PCB concentrations (Tables 4.3.9 and 4.3.10), more cord blood samples from Nunavik Inuit newborns (mean 1.94; range 0.19–15.6 $\mu g \cdot L^{-1}$) would exceed this level of concern than NWT Inuit newborns (mean 0.96; range 0.20–5.10 $\mu g \cdot L^{-1}$). Proportionately fewer newborns from the general population of southern Québec would exceed this level of concern (mean 0.54; range 0.01–3.91 $\mu g \cdot L^{-1}$).

Analyses were completed for 14 PCB congeners in newborn cord blood from Nunavik, northern Québec and southern Québec populations. Similar PCB congener patterns can be seen for southern Québec and Nunavik population surveys (Figure 4.3.3), however the levels for each congener in the Nunavik population survey are consistently higher.

- *Cord blood samples from Inuit populations have higher geometric mean contaminant concentrations and a greater proportion of detected analytes of almost all OCs compared with the Dene/Métis or non-Aboriginal populations. This is not unexpected as Inuit consume more marine based traditional foods which contain higher OC levels. A proportion of Inuit newborn cord blood samples exceed the level of concern for PCBs.*
- *The Inuit cord blood samples from Nunavik have PCB and DDE levels that are double those seen in NWT Inuit. HCB levels are very similar between these two groups. The difference between Inuit from northern Québec and Inuit from the western NWT deserves further study and may be due to amount and type of country food consumed, or differences in the sample population (e.g. age of mothers, number of children or number of children breastfed).*

TABLE 4.3.10

Levels of contaminants in the cord blood ($\mu g \cdot L^{-1}$) of Inuit newborns from Arctic Canada (sampled in 1993–1995)

Contaminant	Population	N	Mean[1] ($\mu g \cdot L^{-1}$)		Range	Proportion detected (%)
			Arithmetic	Geometric[2]		
Aldrin	Nunavik[3]	273	0.01	0.01	0.01–0.03	2
	NWT[4]	62	0.01	0.01	0.01–0.01	0
ß–HCH	Nunavik	273	0.03	0.02	0.02–0.12	40
	NWT	62	0.03	0.03	0.02–0.11	24
α-chlordane	Nunavik	273	0.01	0.01	0.01–0.20	0.07
	NWT	62	0.01	0.01	0.01–0.01	0
γ–chlordane	Nunavik	271	0.01	0.01	0.01–0.03	0.04
	NWT	62	0.01	0.01	0.01–0.01	0
Cis–nonachlor	Nunavik	273	0.03	0.02	0.01–0.18	67
	NWT	62	0.02	0.01	0.01–0.07	24
p,p'-DDE	Nunavik	273	1.28	0.97	0.13–14.0	100
	NWT	62	0.47	0.37	0.07–1.55	100
p,p'–DDT	Nunavik	273	0.06	0.04	0.02–0.55	76
	NWT	62	0.03	0.02	0.02–0.09	5
Hexachlorobenzene	Nunavik	273	0.18	0.14	0.02–1.20	100
	NWT	62	0.18	0.14	0.03–0.91	100
Mirex	Nunavik	272	0.02	0.01	0.01–0.13	29
	NWT	62	0.01	0.01	0.01–0.65	7
Oxychlordane	Nunavik	273	0.10	0.07	0.01–0.67	96
	NWT	62	0.07	0.04	0.01–0.35	68
Transnonachlor	Nunavik	273	0.17	0.13	0.01–1.13	99
	NWT	62	0.07	0.05	0.01–0.33	21
PCBs (as Aroclor 1260)	Nunavik	273	2.47	1.94	0.19–15.60	100
	NWT	62	1.32	0.96	0.20–05.10	100

[1] *When the sample contained a nondetectable concentration, the mean was calculated using half of the detection limit*

[2] *Both arithmetic and geometric means are presented for comparative purposes. Geometric means are less affected by a few extreme values*

[3] *Nunavik, Northern Québec, Dewailly* et al. *1996b*

[4] *Northwest Territories, Mackenzie and Kitikmeot Regions, Walker* et al. *in prep*

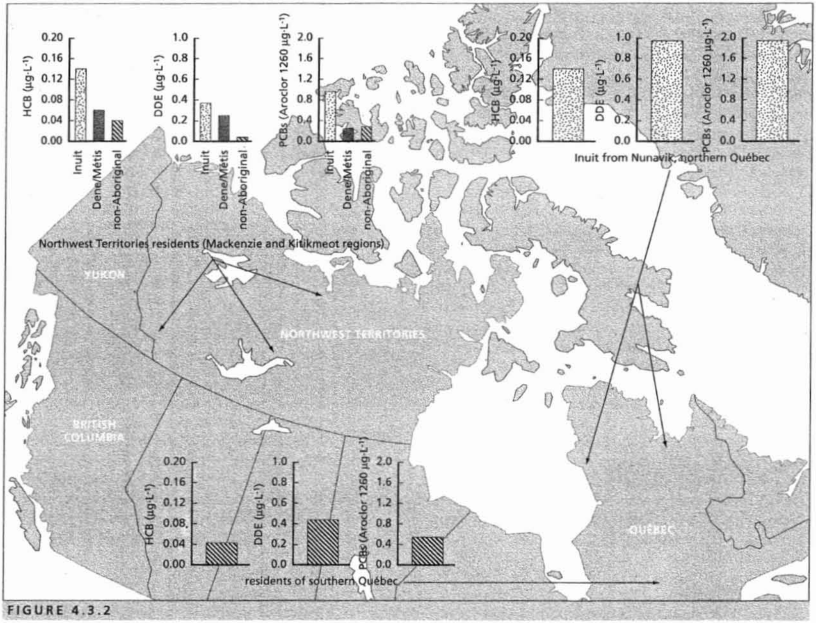

FIGURE 4.3.2

Concentrations of HCB, DDE and PCBs (µg·L-1) in cord blood of newborns in Québec (Dewailly et al. *1996b) and NWT (Walker* et al. *in prep).*

4.3.2 Metals

This section examines three metals: cadmium, mercury, and lead. Considerable uncertainty remains over the extent to which anthropogenic activity contributes to total environmental loadings of these metals, given the significant releases that result from natural mineralization of bedrock. In the Arctic, this is further complicated by a lack of data on the proportion of total deposition that results from atmospheric transport from regions outside the Arctic. However, it is clear that these three metals accumulate in human tissue and that they are harmful to human health.

4.3.2.1 Lead (Pb)

It is estimated that 80% to 90% of human exposure to Pb is from food (Health and Welfare Canada 1989). In the most recent Canadian market basket survey, the estimated mean dietary intake for the average adult in 1986–1988 was 0.4 µg·kg bw^{-1}·d^{-1} (60 kg bw) (Dabeka and McKenzie 1995). The majority of Pb in food originates from atmospheric deposition and to a lesser extent from transportation, packaging and preparation processes. As a result, estimated daily intakes for adults can range world-wide from 15 to 316 µg·day^{-1} (WHO 1995). Infants may have appreciably higher values for estimated Pb intake due to ingestion (pica) of soil and paint chips. Large inter-individual variations also exist in the absorption of Pb from the gastrointestinal tract with the average range estimated at 10% to 20%; infants generally exhibit increased absorption (up to 40% to 50%) (Tsuchiya 1986, Skerving 1988). Low dietary levels of calcium, potassium, zinc, copper and iron have also been associated with increased absorption of Pb from the gastro-intestinal tract (Skerving 1988, Goyer 1988). For infants, both iron deficiency and prolonged breast-feeding by lead-exposed mothers have been associated with elevated levels of blood-Pb. The current WHO TDI for Pb is 3.5 µg·kg bw^{-1}·d^{-1} (Table 4.4.1). The US FDA has developed a specific provisional tolerable daily intake (PTDI) value for children aged 0–6 years old of 6 µg·day^{-1}, which is more stringent than the WHO Provisional Tolerable Weekly Intake (PTWI) for children of 25 µg·kg bw^{-1}·week^{-1}.

Blood-Pb values can reflect relatively recent exposure but are usually stable over time due to equilibration with skeletal stores of Pb, which can account for up to 90% of the total human body burden. In the absence of occupational exposure, the average blood-Pb value in adults is 10 to 20 µg·dL^{-1}, depending on the degree of industrialization (air pollution, automobile exhaust, etc.). This value is far in excess of the estimated blood-Pb level in preindustrialized humans of 0.016 µg·dL^{-1} (Flegal and Smith 1992) (Table 4.3.11). Due to increased absorption of ingested lead through diet and pica, children usually have greater blood-Pb values than adults. In adults, males have greater blood-Pb values than females due to a slightly higher blood hemoglobin content (Skerving 1988). Dietary intake of lead at the WHO PTWI level for infants is estimated to result in a blood lead level of 5.7 µg·dL^{-1} (WHO 1993).

In general, lead poisoning can result in adverse effects associated with the hematological, gastrointestinal and neurological systems. A blood-Pb value of 8 to 10 µg·dL^{-1} has been estimated as the level associated with no observable adverse effect level for neurobehavioral development in children (Ernhart *et al.* 1989, Goyer 1993). In 1991, lead was described as the number one environmental threat to the health of children in the US. The US and Canadian action levels for lead (10 µg·dL blood^{-1}) are similar (FPCEOH 1994).

In a recent investigation of children living in Mexico City (Muñoz *et al.* 1993), a difference of 13 points in full IQ (intelligence quotient) was observed in children with blood-Pb levels of 25 µg·dL^{-1} or more compared with children with blood-Pb of greater than or equal to 15 µg·dL^{-1} or less. Lead levels in cord blood have also been positively associated with lower mental developmental index scores in infants during the first two years of life. Infants from a group with "high" cord blood lead (10–25 µg·dL^{-1}) consistently had lower test scores compared to infants from the groups with cord blood lead of 6 to 7 µg·dL^{-1} or less than 3 µg·dL^{-1} (Bellinger *et al.* 1987). Cord blood-Pb values above 5 µg·dL^{-1} have been associated with premature labour (gestation less than 260 days) (Satin *et al.* 1991). It has been estimated that the risk of preterm delivery (before gestational week

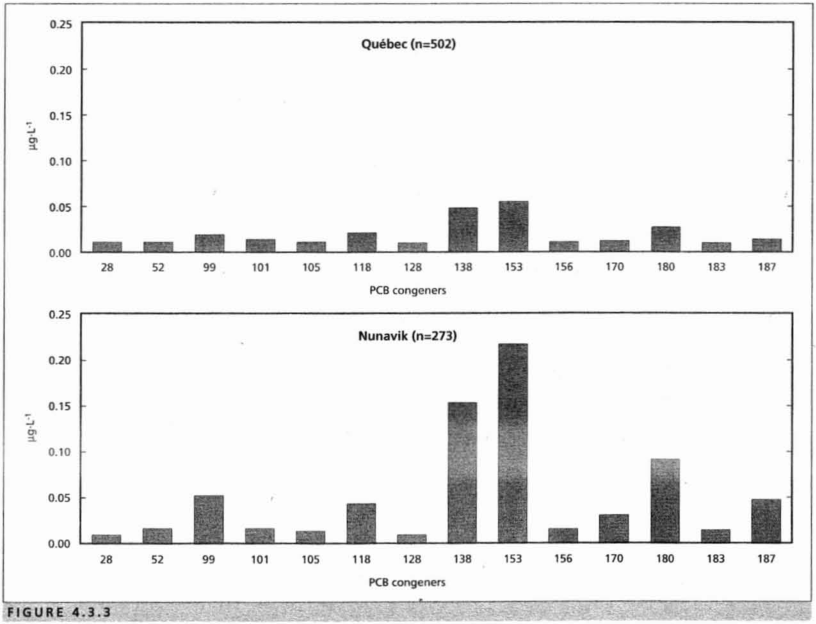

FIGURE 4.3.3

Mean (geometric) concentrations of PCB congeners in cord plasma from newborns in Nunavik (Inuit) and southern Québec (Caucasian). (Adapted from Dewailly et al. *1996b)*

37) increases by almost four-fold for cord blood values greater than 14 µg·dL^{-1} compared to values less than or equal to 8 µg·dL^{-1} (McMichael *et al.* 1986).

Table 4.3.11 compares human blood-Pb levels in Canada, the US and Greenland. Mean blood-Pb concentrations in Nunavik Inuit were approximately 8.6 µg·dL^{-1} (Table 4.3.11) (Dewailly *et al.* 1994b). Blood-Pb levels could be positively correlated to smoking and consumption of marine mammals. In a 1987 survey of northern Greenlandic Inuit hunters and their families, the median blood-Pb value for males was 9.6 µg·dL^{-1} (median age 40 years) and for females, 5.6 µg·dL^{-1} (median age 36) (Milman *et al.* 1994) (Table 4.3.11). The major source of lead exposure was estimated to be food items of marine origin (ringed seal, narwhal, walrus and beluga) and there was a positive correlation between increasing blood-Pb levels and age. In the Faroe Islands, 52 women (20–50 years old) who consumed fish and pilot whale meat had a median blood-Pb level of 2.0 µg·dL^{-1} (Grandjean *et al.* 1992). In a 1989 sample of Michigan fishermen (n=115) who consumed an average of 38 fish meals annually, blood-Pb levels were 5.5 µg·dL^{-1} compared with 3.8 µg·dL^{-1} in a control population (n=95, 4.1 average fish meals·year) (Hovinga *et al.* 1993). Blood-Pb values for both sample sets were directly influenced by the amount of smoking. A study of fish eaters from the Canadian side of the Great Lakes also indicated that women who consumed Great Lakes fish had slightly higher levels of blood lead compared to non-fish eaters (2.4 vs 1.9 µg·dL^{-1}, respectively). (Kearney *et al.* 1995) (Table 4.3.11).

Initial screening results for lead in cord blood samples from Nunavik (Table 4.3.11) have found levels approximately three-fold higher than comparison samples from Toronto and Québec City (5.2 µg·dL^{-1} vs. 1.7 and 1.8 µg·dL^{-1}, respectively) (Dewailly 1994d). A similar study from the NWT found slightly lower lead levels in Inuit, Dene/Métis and non-Aboriginal cord samples (2.9, 2.0 and 1.6 µg·dL^{-1}, respectively) (Walker *et al.* in prep). As the majority of Inuit newborns are well below the 10 µg·dL^{-1} action level they are not considered to be at risk from in utero lead exposure. It is worth noting that the Nunavik value is in the range of cord blood lead associated with the threshold for premature labour (Satin *et al.* 1991). In addition, lead is a cumulative toxin with exposure continuing during lactation, suggesting that biomonitoring in the eastern Arctic should continue during early childhood development.

- *As the majority of Inuit and Dene/Métis newborns are well below the 10 mg·dL^{-1} action level they are not considered to be at risk from in utero lead exposure. It is worth noting that the Nunavik value has been shown to be in the range of cord blood lead associated with the threshold for premature labour.*
- *Because lead is a cumulative toxin and because exposure continues during breast-feeding, blood biomonitoring should continue during early childhood development.*

4.3.2.2 Cadmium (Cd)

The major sources for non-occupational exposure to cadmium in humans are food and tobacco (Dunnick and Fowler 1988, Buchet *et al.* 1990). Based on a market basket survey between 1986 and 1988, the estimated adult intake of cadmium from food in Canada is 0.21 µg·kg bw^{-1}·d^{-1} (Dabeka and McKenzie 1995). This is below the PTWI for adults, as established by the WHO (1989b), of 400–500 µg cadmium (approximately 1 µg·kg bw^{-1}·d^{-1}). Organ meats (liver and kidney) and seafood (shellfish and crustaceans) contain elevated levels of cadmium compared to other food groups. In the Canadian Yukon and NWT, organ meats from terrestrial ungulates (hoofed

TABLE 4.3.11

Lead (Pb) levels in human blood from Canada, US, Greenland and the Faroe Islands.

Location	Year(s) Collected	N	Reported Mean (µg·dL^{-1})	Range (µg·dL^{-1})	Detection Limit (µg·dL^{-1})	Reference	Comments
US Michigan	1989	115 (m)[1]	5.5	1.0–17.0	1.0	Hovinga *et al.* 1993	fish eaters, whole blood
US Michigan	1989	95 (m)	3.8	1.0–10.0	1.0	Hovinga *et al.* 1993	controls, whole blood
CANADA							
Cornwall	1992/93	32 (f)[2]	2.4	<0.5–5.6		Kearney *et al.* 1995	fish eater, whole blood
	1992/93	10 (f)	1.9	<0.5–6.2			non fish eater, whole blood
US California	1984	1728	4.9	0.5–15.0		Satin *et al.* 1991	cord, whole blood
Faroe Islands	1986–87	52 (f)	2.0	0.8–3.6		Grandjean *et al.* 1992	whole blood
Greenland – Inuit	1987	35 (f)	5.6	0.8–32.6		Milman *et al.* 1994	whole blood
Greenland – Inuit	1987	35 (m)	9.6	3.1–24.0		Milman *et al.* 1994	whole blood
Nunavik							
Inuit- adult	1992	492	8.6	0.04–2.28	2	Dewailly *et al.* 1994	whole blood
Inuit-newborn	1994	59	5.2	0.8–26.7	2	Dewailly 1994	cord, whole blood
NWT							
Inuit	1994–95	62	2.9	0.6–15.7		Walker *et al.*	cord, whole blood
Dene/Métis	1994–95	47	2.0	0.2–6.2		(in prep)	
Non-Aboriginal	1994–95	121	1.6	0.2–6.4			

[1] *(m) = males*

[2] *(f) = females*

mammals), particularly caribou and muskoxen, have been identified as the principal dietary cadmium sources. Caribou kidney samples usually contain the highest concentration of cadmium but levels are comparable to Ontario and Québec moose and white-tailed deer kidney samples. Cadmium content of muscle or meat from organs other than kidney or liver has been determined to be up to 100 times lower (Gamberg *et al.* 1994).

After absorption, cadmium is transported in blood, primarily bound to erythrocytes, to the liver. The average blood-Cd concentration for non-smokers with non-occupational exposure is usually less than 1 µg cadmium·litre^{-1} (Ferguson 1990). The major routes of cadmium excretion in humans are through urine and feces. Minor routes of elimination include hair and human milk (Friberg and Elinder 1988). Measures of cadmium exposure include levels in blood, urine, feces and body organs. For the general non-smoking population, blood-Cd values rarely exceed 1 µg·litre^{-1}, while urinary excretion of cadmium is usually less than 0.5–2 µg cadmium·d^{-1} (WHO 1992a). A variety of studies have shown that in non-smoking, non-occupationally exposed subjects, dietary intake of cadmium between 15 and 53 µg·day^{-1} results in a cadmium concentration in whole blood of 0.2–1.6 µg·L^{-1} (Louekari *et al.* 1991).

Chronic human exposure to cadmium has been associated with anaemia, osteomalacia (softening of the bones), cardiovascular diseases and kidney damage. The kidney has been identified as the critical target organ. In humans, when the kidney carrying capacity for cadmium has been exceeded (influenced in part by cadmium concentration and metallothionein-inducing capability of the tubular cells), cadmium-induced nephrotoxicity develops, resulting in decreased glomerular filtration efficiency and proximal tubule dysfunction (Buchet *et al.* 1990). Exposure to cadmium at the recommended WHO PTWI is calculated to result in kidney-cadmium concentrations of less than or equal to 50 mg·kg^{-1} (wet wt.) by age 50. A human kidney concentration of 200 mg·kg^{-1} is associated with a cumulative lifetime intake of 2000 mg or a chronic intake of 140 to 260 µg cadmium·day^{-1}. At this level, it has been estimated that approximately 10% of the population would develop renal tubular proteinuria.

Recent studies indicate that this may be an underestimation of the risk. Elderly people appear to develop renal tubular dysfunction at cadmium concentrations around 50 mg·kg^{-1} (Elinder and Jarup 1996). If the minimal toxic concentration is at this level, some people — especially the elderly and diabetics — living in areas with elevated cadmium levels will be affected. This also implies that the current PTWI of 7 µg·kgbw^{-1} is too high (Elinder and Jarup 1996). This may be a significant problem because cadmium-induced tubular dysfunction is irreversible (Kido *et al.* 1988) and because no effective chelating therapy has been found (Jones and Cherian 1990).

Although metallothionein induction has been detected in placental tissue, cadmium analysis in paired maternal:cord blood samples collected from

TABLE 4.3.12

Cadmium (Cd) levels in human blood from Canada, US and Greenland

Location	Year(s) collected	N	Reported mean (µg·L^{-1})	Range	Detection limit (µg·L^{-1})	Reference	Comments
U.S. Michigan	1989	115 (m)[1]	0.6	0.1–5.0	0.1	Hovinga *et al.* 1993	Fishermen-whole blood
U.S. Michigan	1989	95 (m)	0.41	0.1–2.5	0.1	Hovinga *et al.* 1993	controls–whole blood
CANADA Ontario	1992/93	65	3.94	1.0–14.6	0.2	Kearney *et al.* 1995	smokers, whole blood
CANADA Ontario	1992/93	59	0.14	<0.2–0.9	0.2	Kearney *et al.* 1995	non-smokers, whole blood
Greenland Inuit	1987	32 (f)[2]	2.2	0.8–6.5		Milman *et al.* 1994	whole blood
Greenland Inuit	1987	35 (m)	2.3	0.7–6.9		Milman *et al.* 1994	whole blood
Nunavik Inuit	1988	7	0.27	0.31–0.45		Benedetti *et al.* 1992	non-smokers, whole blood
Nunavik Inuit	1988	117	5.4	4.7–6.2		Benedetti *et al.* 1992	smokers, whole blood
NWT							
Inuit	1994–95	67	1.8	0.07–7.3		Walker *et al.* (in prep)	maternal, whole blood
Dene/Métis	1994–95	51	0.7	0.01–5.4			
Non-Aboriginal	1994–95	121	0.6	0.01–8.6			
Inuit	1994–95	62	0.11	0.011–0.495			cord, whole blood
Dene/Métis	1994–95	47	0.11	0.011–0.641			
Non-Aboriginal	1994–95	121	0.12	0.011–2.383			

[1] *(m) = males*

[2] *(f) = females*

three cities in Taiwan suggests that the placenta acts as only a partial barrier. In 159 samples collected in 1988, the mean cadmium level in cord blood was approximately 60% that of maternal blood (0.78 vs. 1.30 $\mu g \cdot L^{-1}$, respectively) (Soong *et al.* 1991). Similar studies looking at the relationship between cadmium levels in maternal vs. fetal blood are underway in the NWT.

Sampling in Nunavik in 1992 indicated that cadmium blood values for nonsmoking Inuit and non-smoking urban Caucasians (Québec City) are not appreciably different (Table 4.3.12) (Bennedetti *et al.* 1994). However, the mean blood cadmium level in Inuit smokers was 5.4 $\mu g \cdot L^{-1}$, approximately 20-fold higher than the blood cadmium level of a smaller subset of nonsmokers (0.27 $\mu g \cdot L^{-1}$). The influence of dietary and environmental sources of cadmium exposure appear to be minor compared to the contribution from smoking (Benedetti *et al.* 1994). A study of mean Inuit maternal blood-Cd levels from the NWT found similar levels of cadmium (1.8 $\mu g \cdot L^{-1}$) as this group is likely made up of both non-smokers and smokers (Table 4.3.12). Lower levels of cadmium were seen in the Dene/Métis and non-Aboriginal mothers surveyed (0.7 $\mu g \cdot L^{-1}$ and 0.6 $\mu g \cdot L^{-1}$ respectively) (Walker *et al.* in prep). Cadmium cord blood levels of newborns are similar for all three ethnic groups in the NWT study (0.11 to 0.12 $\mu g \cdot L^{-1}$) and are five- to fifteen-fold lower than the mean maternal values (Walker *et al.* in prep).

A study of the diet of Dene people in Fort Resolution, NWT indicated that organ meats (liver and kidney) of caribou and moose contained high concentrations of cadmium. Consumption of organ meats was low in this group so intakes of cadmium were well below the WHO PTWI (Chan *et al.* 1995b). In an evaluation of cadmium exposure of northern Québec Cree (Archibald and Kosatsky 1991), it was estimated that regular consumption of moose and caribou kidney and liver (two moose and three caribou per year equally shared among a family of five) would result in a cadmium intake near the WHO PTWI limit of 1 $\mu g \cdot kg\ bw^{-1} \cdot d^{-1}$. Combining the additional cadmium exposure from one pack of cigarettes per day increased cadmium intake to approximately 50% greater than the PTWI (Archibald and Kosatsky 1991). The Cree Regional Authority were advised that no consumption restriction be applied, but urinary cadmium biomonitoring be implemented. Traditional food use, at least where cadmium is the contaminant of concern, need not be altered if smoking habits can be modified.

- *Cadmium levels in most of the traditional foods are fairly low. Therefore, the major route of cadmium exposure for most people is through cigarette smoking.*
- *Organ meats (liver, kidney) can contain significant amounts of cadmium. Therefore, oral exposure becomes substantial for people who consume organ meats frequently (once a week year round). They may have higher risk of chronic cadmium toxicity, particularly if they also smoke.*
- *Cadmium levels of smokers are elevated and may have an irreversible effect on kidney function. A recent risk assessment for cadmium indicates that some people, especially the elderly and diabetics, may be affected at lower levels than those previously considered safe.*

4.3.2.3 Mercury (Hg)

Of the various species of mercury in the environment, methylmercury (MeHg) has the greatest toxic effect in humans (WHO 1990). It is a more potent neurotoxin than inorganic Hg. Although the majority of Hg released to the environment from natural and human sources is in the inorganic form, certain microorganisms in aquatic systems are able to convert inorganic Hg to MeHg, rendering it available for uptake through the food-web (Lindberg *et al.* 1987). Methylmercury is bio-accumulated and biomagnifies.

In the absence of occupational exposure, food is considered to be the primary environmental pathway of mercury for humans. Methylmercury in the human diet is almost completely absorbed into the blood stream (WHO 1990). Foods containing high levels of MeHg, such as certain fish and marine mammals, can be a very significant source of exposure for humans. Following absorption, MeHg concentrates in kidneys, liver and central nervous system (CNS) tissues, particularly the brain which is the critical target organ. Excretion is primarily via feces. It is also excreted in human milk and deposited in hair. MeHg crosses the blood-brain-barrier and the placenta (WHO 1990).

The WHO PTWI for Hg has been set at 300 $\mu g \cdot person^{-1}$ of which no more that 200 μg may be methylmercury (0.47 $\mu g \cdot kg\ bw^{-1} \cdot d^{-1}$) (WHO 1990). In humans, blood-MeHg and hair-MeHg levels are generally used as indicators of exposure and have been correlated to both total body burden and brain levels. Since hair grows at about 1 cm per month, MeHg levels in hair, if analysed in 1 cm segments, can provide a temporal trend for Hg accumulation. Hair analyses can be correlated to blood-MeHg values at the time of hair growth. In the case of prenatal exposure, a nine-cm hair strand will reflect maternal MeHg-blood values for the entire gestational period and can be related to MeHg levels in maternal milk and in the fetus. The hair:blood Hg concentration ratio in humans has been estimated at approximately 200–300:1 (Clarkson 1992). Among Canadian First Nations peoples the ratio was found to be about 300:1 (Phelps *et al.* 1980).

In populations consuming little or no fish or marine mammals, blood-Hg values are generally less than 2 $\mu g \cdot L^{-1}$, and hair-Hg values are less than 2 $\mu g \cdot g^{-1}$

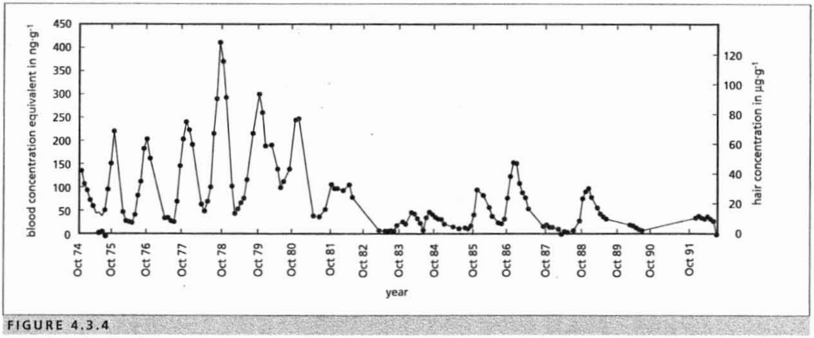

FIGURE 4.3.4

Seasonal variation of methylmercury levels in a fishing guide from Grassy Narrows Reserve, Ontario. Composite from a series of hair samples. Hair samples were analysed cm by cm, and a growth rate of 1 cm·month^{-1} was assumed. (Wheatley and Paradis 1995)

(WHO 1990). Mean hair-Hg levels from North American fish-eating populations range from 1.2 to 2.4 µg·g^{-1}. Among Canadian Aboriginal peoples the blood-Hg range has been found to be 1 to 660 µg·L^{-1} (Wheatley 1979, Wheatley and Paradis 1995) with a strong association with fish eating. In Greenlandic Inuit populations, cord blood methylmercury levels from women who reported consuming marine mammals (beluga and narwhal) more than six times per week were approximately two times greater than levels from women who reported consuming marine foods less than once a week (Hansen *et al.* 1990).

Clinical symptoms and signs of MeHg toxicity are primarily neurological and range from paraesthesia, ataxia and tunnel vision to cerebral palsy and death, depending on the level of exposure. Epidemiological studies among populations in Japan and Iraq have demonstrated that high prenatal exposure to methylmercury (over 400 µg·g^{-1} in maternal hair) may result in severe cerebral palsy and death for the infant. Studies at much lower maternal levels (about 20 µg·g^{-1}) in New Zealand (Kjellstrom *et al.* 1986) and Canada (McKeown–Eyssen and Ruedy 1983) suggested subtle developmental effects only. Extrapolation of the Iraqi data has led the WHO to suggest that there is a 5% risk of neonatal neurological disorders associated with a peak methylmercury level of 10 to 20 µg·g^{-1} in maternal hair (WHO 1990). Further work is currently under way on this issue in the Seychelles and the Faroe Islands and in Canada (Grassy Narrows and Whitedog Reserves) to assess the accuracy of this risk estimate. The WHO (1990) has also suggested that 5% of the adult population may be at risk of developing early signs of mercury toxicity (paraesthesia) at a blood mercury level of 200 µg·L^{-1}. In Canada, despite adult mercury levels up to 660 µg·L^{-1} in some Aboriginal people, possible neurological effects related to methylmercury were found in only 11 individuals, but given numerous confounders, no proven diagnosis of methylmercury poisoning could be made (Wheatley *et al.* 1979, Wheatley and Paradis 1996a). While the data in figure 4.3.4 is only one person's exposure over 15 years, peak mercury concentrations are seen in October/November of each year (Wheatley and Paradis 1995). This seasonal nature of exposure may have protected Aboriginal Canadians exposed to mercury through fish consumption (high exposures in fishing season and lower exposure the rest of the year).

From 1970 to 1995, the Medical Services Branch (MSB) of Health Canada, within its Environmental Contaminants Program, has, as part of clinical risk assessment, carried out analyses of hair and blood

TABLE 4.3.13

Percent of Dene and Inuit individuals in the NWT with methylmercury blood levels within four effect ranges (1972–1989) (Wheatley 1994c).

	DENE		INUIT	
Methylmercury Levels (µg·L^{-1})	**# of individuals**	**Percent (%)**	**# of individuals**	**Percent (%)**
Less than 20	572	80.45	647	42.82
20 to 99	137	19.27	844	55.86
100 to 199	2	0.28	16	1.06
200 to 299	0	0.00	4	0.26
TOTAL	711	100.00	1511	100.00

[1] *Based on WHO guidelines, Medical Services Branch has defined: <20 µg·L^{-1} – normal acceptable range; 20–100 µg·L^{-1} increasing risk; and > 100 µg·L^{-1} at risk.*

TABLE 4.3.14

Current and historical blood mercury levels

Location	Year(s) collected	N	Reported mean (µg·L^{-1})	SD	Range (µg·L^{-1})	DL[1] (µg·L^{-1})	Reference	Comments
CANADA								
(Aboriginal Peoples)	1970–1995	38 571	14.13	22.63	1–660	1	Wheatley and Paradis[3] 1995	Blood, Hair, Cord Blood
QUÉBEC								
Nunavik (Inuit)	1974–1982	1 114	46.76	33.08	2.67–267.33	1	Wheatley and Paradis[3]	Blood, Cord Blood, Hair
	1977–1982	142	48.21	43.98	4.20–254.50	1	1996a	Blood
	1978–1982	125	33.80	20.09	6.00–101.80	1		Cord Blood
	1974–1982	847	48.42	32.10	2.67–267.33			Hair
Nunavik (Inuit)	1992	252	14.06		0.8–96		Dewailly *et al.* 1994	Blood, men
	1992	240	15.97		2.0–112		Dewailly *et al.* 1994	Blood, women
	1993–1995	299	12.09		1.0–99		Dewailly *et al.* 1994	Cord blood
Northern (Cree)	1971–1982	4 670	34.31	40.56	1.50–649.40	1	Wheatley and Paradis[3] 1996a	Blood, Maternal Blood, Cord Blood, Hair
	1971–1982	1 129	42.94	52.32	1.50–649.40	1		Blood
	1971–1982	600	22.69	23.82	1.50–224.00	1		Cord Blood
	1972–1982	2 940	33.38	37.29	1.67–571.00	1		Hair
NWT								
Nunavut (Inuit)	1972–1989	1 339	29.98	23.99	1.00–225.67	1	Wheatley and Paradis[3] 1996a	Blood, Maternal Blood, Cord Blood, Hair
	1972–1989	286	19.52	22.33	1.00–200.00	1		Blood
	1978–1988	61	17.17	15.48	1.50–86.00	1		Maternal Blood
	1978–1986	31	40.53	32.74	6.50–130.40	1		Cord Blood
	1973–1987	961	33.57	23.37	1.00–225.67	1		Hair
Nunavut (Inuit)	1994–1995	67	3.46		00.6–12.84		Walker and Seddon (in prep)	Maternal Blood
	1994–1995	62	5.72		0.6–27.88			Cord Blood
Western (Dene)	1976–1983	980	16.24	16.54	1.00–138.00	1	Wheatley, 1995[3]	Blood, Cord Blood, Hair
	1982–1983	76	10.66	11.13	1.30–66.50	1		Blood
	1978–1983	5	14.36	12.32	4.30–35.00	1		Cord Blood
	1976–1983	899	16.72	16.86	1.00–138.00	1		Hair
Western (Dene)	1994–1995	51	1.74		0.4–5.62		Walker and Seddon (in prep)	Maternal Blood
	1994–1995	47	1.91		0.4–8.83			Cord Blood
Non-Aboriginals	1994–1995	121	1.25		0.20–4.21		Walker and Seddon (in prep)	Maternal Blood
	1994–1995	121	1.66		0.20–12.84			Cord Blood
YUKON	1977–1978	299	7.10	6.15	1.50–67.00	1	Wheatley, 1979[3]	Blood, Cord Blood, Hair
	1977–1978	83	7.04	4.01	2.10–21.70	1		Blood
	1977	31	4.12	1.87	1.50–7.80	1		Cord Blood
	1977	185	7.63	7.19	1.67–67.00	1		Hair
COMPARISON GROUPs								
Ontario	1992–1993	176	2.80	2.30	<2.0–17.0	2	Kearney *et al.* 1995	Whole Blood Fish "eaters"
Greenland	1987	35	12.00		4–23		Milman *et al.* 1994	Serum
	1987	32	10.00		3–24		Milman *et al.* 1994	Serum
	1982–1988	37	44.13	24.07	6.02–96.29	10	Hansen *et al.* 1990	Maternal Blood
	1982–1988	37	88.25	48.14	10.03–178.54	10	Hansen *et al.* 1990	Cord Blood
Faroe Island	1986–1987	53	12.00		2.6–50.0		Grandjean *et al.* 1992	Whole Blood
	1986–1987	53	2.10		<0.1–8.2		Grandjean *et al.* 1992	Serum
	1986–1987	997	24.20		13.0–40.2		Grandjean *et al.* 1992	Cord Blood

continued next page

TABLE 4.3.14 (continued)

Current and historical blood mercury levels

Location	Year(s) collected	N	Reported mean (µg·L⁻¹)	SD	Range (µg·L⁻¹)	DL1 (µg·L⁻¹)	Reference	Comments
HISTORICAL LEVELS FROM MUMMIFIED REMAINS								
Greenland	"1485" AD	1	10.00				HartHansen *et al.* 1991	Hair
N. Baffin (Dorset)	"400" AD	3	8.00		3–12	1	Wheatley and Wheatley 1988	Hair
N. Baffin (Thule)	"1150" AD	6	4.00		2–7	1	Wheatley and Wheatley 1988	Hair

[1] *DL = Detection Limit*

[2] *Measurements of mercury were made in a variety of tissues but all tissue levels are normalized as concentrations in blood*

[3] *Some of this data has been specifically analysed for this report by Wheatley 1996 but is described in Wheatley and Paradis 1995, 1996*

samples obtained from 38,571 residents of 514 native communities across Canada (Wheatley 1994b). Of these, 23% had blood or blood equivalent (extrapolated from hair) MeHg values of greater than 20 $\mu g \cdot L^{-1}$ while 1.6% had levels greater than 100 $\mu g \cdot L^{-1}$. Two thousand four hundred and five (2405) cord blood samples were also obtained — 21.8% of newborns had MeHg levels greater than 20 $\mu g \cdot L^{-1}$ (Wheatley and Paradis 1995). Analysis of samples collected from 59 native communities in the NWT between 1972 and 1989 has shown that 40.6% of the blood-MeHg levels were greater than 20 $\mu g \cdot L^{-1}$ and 1% were greater than 100 $\mu g \cdot L^{-1}$ (for details see Wheatley 1995). Of the 711 Dene and 1511 Inuit who were tested, nearly three times as many Inuit (57.2%) exceeded 20 $\mu g \cdot L^{-1}$ as Dene (19.6%) (Table 4.3.13) (Wheatley 1995, Wheatley and Paradis 1996a).

In a limited number (n=36) of cord blood samples collected from Inuit and Dene from 1978 to 1986 from various regions of the NWT, 69% had methylmercury levels equal to or greater than 20 $\mu g \cdot L^{-1}$ (mean 36.9 $\mu g \cdot L^{-1}$, range 4.3–130.4) (Wheatley and Paradis 1996a). This is in contrast to more recent data (1993–1995) from the NWT and Nunavik which found lower levels in cord blood of Inuit newborns (mean 5.72 and 12.09 $\mu g \cdot L^{-1}$, respectively — Table 4.3.14). The concentration of mercury in maternal blood was also markedly lower (3.46 $\mu g \cdot L^{-1}$ in a 1993–95 study compared with 17.27 $\mu g \cdot L^{-1}$ in the 1978–86 study). (Table 4.3.14) (Wheatley and Paradis 1996a, Walker *et al.* in prep). The lower concentrations seen in 1993–1995 cord and maternal blood samples may be due to dietary differences over time, geographic differences (eastern vs. western NWT), and seasonal differences (Table 4.3.14).

To ascertain which communities showed the most widespread elevated blood mercury over the 20-year review period, the mean methylmercury levels for Canadian Native communities were calculated and divided into quartiles (Figure 4.3.5). Only the fourth quartile reaches levels of increasing risk (Wheatley 1979). The majority of nothern communities are in the fourth quartile (Figure 4.3.5). When the process was repeated for NWT communities alone (Figure 4.3.6), the quartile values change upward, indicating higher exposure levels. The fourth quartile includes primarily the Northern and Eastern Inuit communities where the consumption of fish and sea-mammals is extensive (Wheatley and Paradis 1996a).

Figure 4.3.7 shows comparative MeHg levels found among Aboriginal residents of the Yukon, NWT, and northern Québec. As with Figure 4.3.6, this emphasizes the higher levels found in northern Québec and among the Inuit (Wheatley and Paradis 1996b). The source of mercury (geological or anthropogenic) to the Arctic food chain continues to be a subject of debate (Wheatley and Wheatley 1988, Muir *et al.* 1992). There is also ongoing controversy about

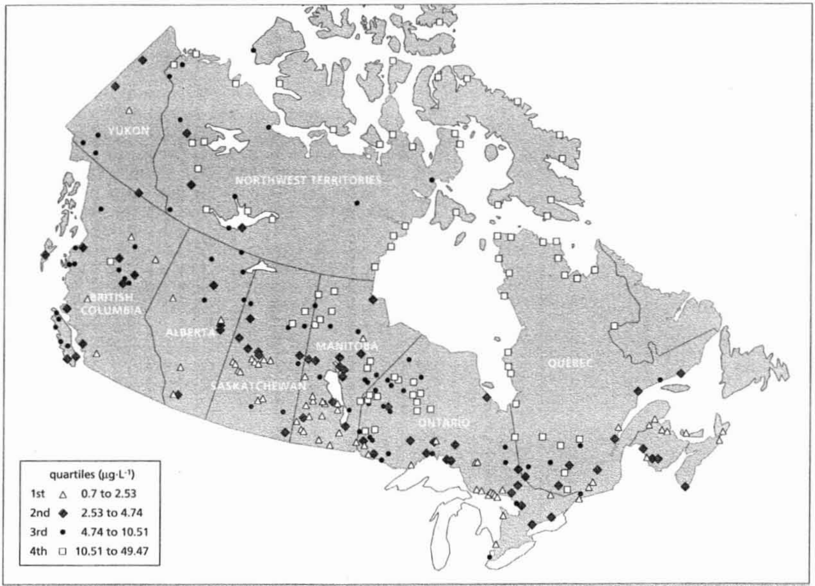

FIGURE 4.3.5

Aboriginal community mean blood methylmercury levels (µg·L^{-1}), 1972–1992. (Wheatley and Paradis 1995b)

possible protective mechanisms involving selenium (Section 4.2.5.3).

The MeHg concentrations in archaeological hair samples from northern Baffin Island (400 and 1150 AD) (Wheatley and Wheatley 1988) and from Greenland (1485 AD) (Hart–Hansen *et al.* 1991) are also included in Table 4.3.14 (converted to blood level equivalents to facilitate comparison). These archaeological samples are generally lower than current human MeHg levels (1972–1989) in the same geographic areas. Figure 4.3.8 provides trend data over 15 to 20 years (1971–1987) for individuals and communities with extensive sampling south of 60°N (First Nations) and in the NWT (Wheatley and Paradis 1996a). The trend in First Nations communities south of 60°N appears to be downward. It has yet to be determined whether this is due to decreasing MeHg levels in fish, or declining fish consumption. However, there is no obvious pattern in the NWT data. More recent data (1992–1995) from the NWT and Nunavik suggest that some Dene and Inuit groups may have lower blood mercury levels (means 1.74 to 15.97 µg·L^{-1} versus 10.66 to 48.42, seen in 1974–1982, Table 4.3.14). Further study needs to be undertaken to assess whether dietary, geographic, regional or seasonal factors are causing this apparent decrease.

- *Methylmercury is a potent neurotoxin and the most toxic form of mercury in the environment. Human exposure in the Arctic is almost exclusively through food consumption, especially fish and marine mammals.*
- *No downward trend is evident in Inuit blood-Hg levels between 1975 and 1987, but more recent data (1992–1995) indicates lower levels of mercury in some groups. Methylmercury concentrations in cord blood taken from Inuit and Dene newborns in the NWT between 1994 and 1995 are seven times lower (4.1 µg·L^{-1} versus 36.9 µg·L^{-1}) than those reported from infants in the same territory (sometimes different communities) in 1978 to 1986. Very few babies exceeded the 20 µg·L^{-1} level in the newer study.*
- *Blood mercury levels are highest in the northern and eastern Inuit communities.*
- *The WHO has concluded that there is a 5% risk of neonatal neurological disorders associated with a peak methylmercury level of 10–20 µg·g^{-1} in maternal hair. Work is underway to assess the accuracy of this risk estimate. Approximately 21% of newborns and 23% of adult native people across Canada had blood mercury levels greater than 20 µg·L^{-1} between the early 1970s and early 1990s. In the NWT, 56% of adult Inuit and*

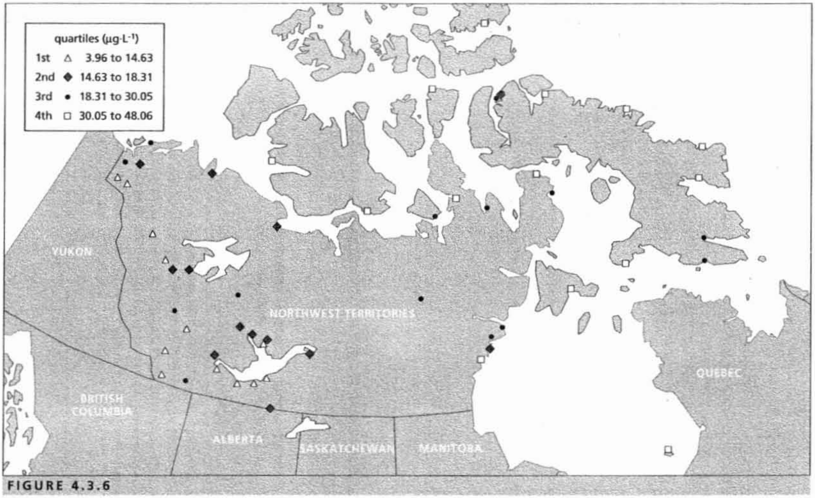

FIGURE 4.3.6

NWT Aboriginal communities mean blood MeHg levels (µg·L^{-1}), 1972–1987. (Wheatley and Paradis 1995b)

19% of adult Dene had blood mercury levels greater than 20 µg·L^{-1}. Blood mercury levels are higher for Inuit than Dene.

4.3.3 Radionuclides

Radionuclides in the environment may be of natural or anthropogenic origin. Natural radionuclides include potassium-40 (^{40}K)and members of the uranium and thorium decay series. Humans have always been exposed to natural background radiation. Environmental levels of these radionuclides may become enhanced as a result of human activities such as uranium mining.

Anthropogenic radionuclides in the environment are mainly products of nuclear fission. During the 1950s and 1960s, radionuclides entered the Arctic environment as fallout from the atmospheric testing of nuclear weapons (UNSCEAR 1982, 1993). The signing of the Limited Test Ban Treaty in 1963 did much to reduce this source of contamination, however, France and China were not parties to the treaty and continued atmospheric testing until 1980. The Chernobyl reactor accident of 1986 was an additional source of radioactive material to the Canadian Arctic via atmospheric transport (EHD 1987, UNSCEAR 1988).

The amounts of radionuclides in Arctic air, water and soil are generally about the same as, or lower than, levels found in temperate zones. Radionuclides bioconcentrate in certain food chains (i.e. the lichens-to-caribou-to-humans pathway, and the fresh water invertebrates-to-fish-to-humans pathway (Figures 4.3.9 and 4.3.10). These pathways are particularly important for anthropogenic radiocesium and for natural lead-210 and polonium-210.

The health effects from radionuclides are related to the fact that they emit ionizing radiation, a known carcinogen. These carcinogenic effects have been well documented by studies of human populations exposed at high levels of radiation, for example, atomic-bomb survivors, occupationally exposed persons, and persons treated medically for other conditions (UNSCEAR 1977, BEIR 1990, ICRP 1991). The activity (disintegrations/second) of any radionuclide is measured in a tissue or sample in Bq·kg^{-1}. The resulting radiation doses or exposures are then measured in units of sieverts (Sv) or millisieverts (mSv) (one Sv = 1000 mSv) which measures the radiation energy absorbed adjusted for the different types of radioactivity (e.g. α-, β- or γ- rays). A relative risk comparison of various doses of radiation is presented in Table 4.3.15.

The following is a brief description of those radionuclides, both anthropogenic and natural, that are of greatest concern in the environment.

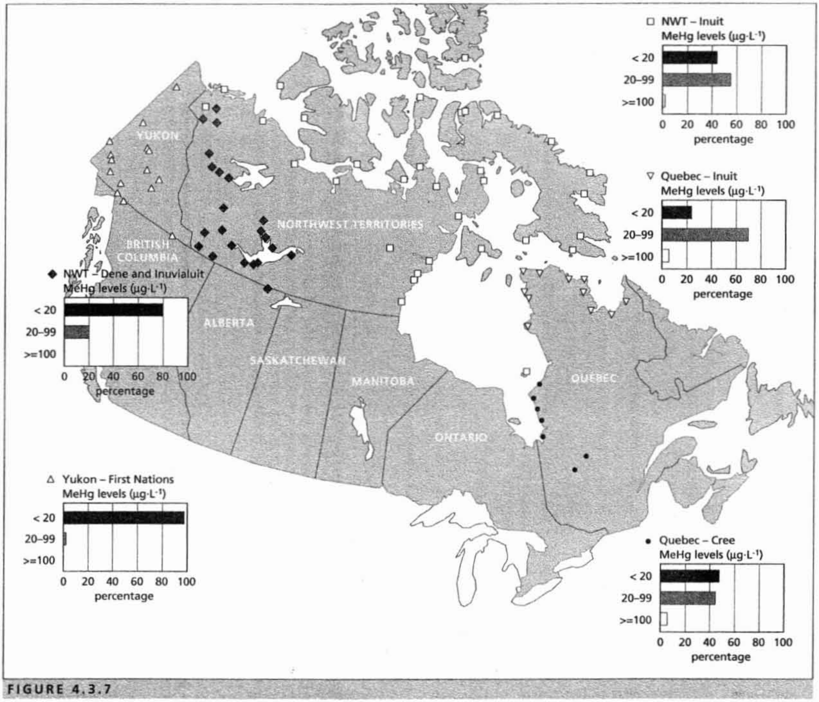

FIGURE 4.3.7

Range of blood mercury levels in northern Aboriginal communities, 1972–1992. (Wheatley and Paradis 1996b)

4.3.3.1 Anthropogenic Radionuclides

Radiocesium

Radiocesium consists primarily of two isotopes, cesium-137 (^{137}Cs)with a half-life of 30 years and cesium-134 (^{134}Cs)with a half-life of about two years. Radiocesium from atomic bomb fallout is predominantly ^{137}Cs. Both ^{137}Cs and ^{134}Cs are present in nuclear reactor wastes in a ratio of about two to one, respectively. The radiocesium isotopes are considered to be the anthropogenic radionuclides of greatest concern in the Arctic environment. The most significant route of cesium exposure is the ingestion of caribou meat (Figure 4.3.10), followed by ingestion of freshwater fish. Levels of ^{137}Cs measured between 1987 and 1988 in caribou of the Canadian Arctic ranged from 50 to 700 Bq·kg^{-1} (EHD 1991, Thomas *et al.* 1992). Fish from arctic freshwater lakes contained up to 20 Bq·kg^{-1} of ^{137}Cs (Lockhart *et al.* 1992). Levels in both fish and caribou are below the maximum acceptable level for ^{137}Cs in meat (i.e. of 1000 Bq·kg^{-1}, Codex Alimentaries Commission 1989). Roughly 25%

TABLE 4.3.15

A relative risk comparison of various radiation doses (UNSCEAR 1988, ICRP 1991)

Dose in millisieverts	Description	Cancer risk
10 000 in a short time (hours)	survival unlikely	—
1 000 in a short time (hours)	onset of acute radiation illness	one in 20
20 in a year	occupational dose limit	one in 1 000
2 in a year	background radiation dose	one in 10 000
1 in a year	public dose limit	one in 20 000
0.1 per exposure	chest x-ray	one in 200 000

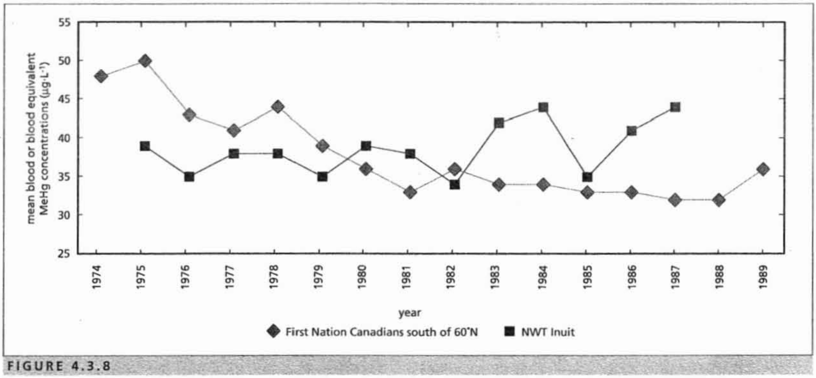

FIGURE 4.3.8

Temporal trends in MeHg levels greater than or equal to 20 µg·L-1 (blood or blood equivalent) in Aboriginal people in northern and southern Canada (i.e. north/south of 60°N). (Based on Wheatley and Paradis 1996a)

of the radiocesium in arctic biota is due to the Chernobyl accident; the remainder is from residual fallout from weapons testing in the 1950s and 1960s.

In biological systems, cesium behaves in a manner similar to potassium, an essential nutrient. Ingested radiocesium is almost completely absorbed by the intestine and becomes distributed throughout the body, mainly in muscle tissue. It is cleared from the body with a half-life of two to three months (Tracy *et al.* 1993a). Because of the radiation it emits, radiocesium can be readily measured in humans by means of a portable whole-body counter. During 1989 and 1990, such measurements were carried out on more than 1100 Inuit and Dene/Métis in five Arctic communities where caribou-hunting is prevalent (Tracy *et al.* 1993b, in press). The highest concentration of ^{137}Cs in human tissue (mainly muscle) was 110 Bq·kg^{-1} corresponding to a radiation dose of 0.40 mSv·y^{-1}. This can be compared with the dose received from natural background radiation which is about 2 mSv·y^{-1}. In the 1960s, some individual levels were as high as 2000 Bq·kg^{-1}, resulting in a dose of up to 5 mSv·y^{-1}. Figure 4.3.11 shows the downward trend in radiocesium body burdens from the 1960s to 1989–90. Each bar represents the average body concentration across all Canadian Arctic communities surveyed for a given year.

Strontium-90

Strontium-90 (^{90}Sr) has a half-life of about 30 years. It is not considered a problem in arctic food chains. In fact the chief route of exposure for Arctic residents would be from food imported from agricultural areas in the south (UNSCEAR 1982).

About 30% of ingested strontium is absorbed by the intestine and, being chemically similar to calcium, it becomes deposited in bone where it may remain for many years (ICRP 1978). It was largely the concern over this radionuclide, and especially its effects on the developing bones of children, that led to the banning of atmospheric nuclear weapons tests in 1963. ^{90}Sr has been measured in autopsy samples of human vertebrae across Canada. The levels across Canada have decreased to 50 Bq·kg calcium^{-1} by 1978 from a maximum of 150 Bq·kg calcium^{-1} in 1964 (Meyerhof *et al.* 1979).

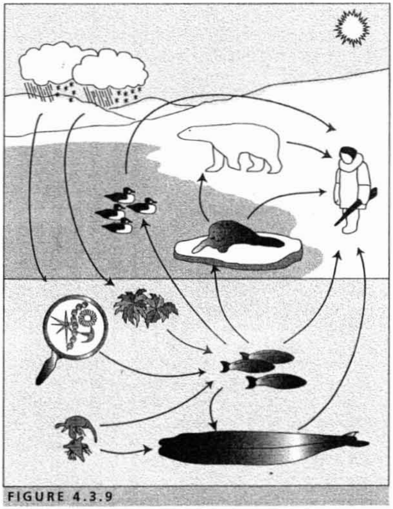

FIGURE 4.3.9

Movement of contaminants through the Arctic marine food chain

FIGURE 4.3.10

Movement of contaminants through the terrestrial food chain

Radioiodine

The chief isotope of radioiodine is iodine-131 (^{131}I)with a half-life of eight days. Iodine-132(^{132}I) also occurs in fresh fallout because its precursor, tellurium-132 (^{132}Te), has a half-life of 78 hours. These radionuclides have entered the Canadian Arctic as a results of weapons fallout (UNSCEAR 1982) and the Chernobyl accident (EHD 1987). Because of their short half-lives, the isotopes of iodine are considered to be a problem only in freshly contaminated foods such as milk and leafy vegetables. They are not concentrated in the meat and fish, which are more typical of arctic diets. An additional isotope, iodine-129 (^{129}I), is released in extremely minute quantities from nuclear fuel reprocessing facilities. It has a very long half-life (16 million years) and contributes a very low but long-term radiation exposure to the entire world population (UNSCEAR 1993).

Radioiodine is chemically indistinguishable from stable iodine and, if ingested, is strongly absorbed by the thyroid gland. Radioiodine has been associated with thyroid cancer (UNSCEAR 1977). This association was seen most recently in children from the former Soviet Union who were affected by fallout from the Chernobyl accident (Kasakov *et al.* 1992). These incidences have all occurred at extremely high levels of thyroid exposure. No effects have ever been observed at the extremely low levels of radioiodine normally found in the Arctic environment.

4.3.3.2 Natural Radionuclides

Lead-210 (^{210}Pb) and Polonium-210 (^{210}Po)

Of all radionuclides today, natural or anthropogenic, ^{210}Pb and ^{210}Po make the greatest contribution to human radiation doses in the Arctic. Both radionuclides are members of the decay series of naturally-occurring uranium-238 (^{238}U). They become airborne as a result of the decay of radon gas seeping from soil. Uranium mining and processing operations may lead to enhanced concentrations of these two radionuclides in the environment. ^{210}Pb has a half-life of 22.3 years. It decays to ^{210}Po (half-life = 138 days).

^{210}Pb and ^{210}Po occur in nature as airborne particulates. They tend to settle out on vegetation such as lichens and are eaten by caribou. ^{210}Po levels as high as 200 to 500 Bq·kg^{-1} were measured in caribou livers and kidneys. Levels in muscle are much lower — about 10 to 30 Bq·kg^{-1}. ^{210}Pb concentrates in bone; values in soft tissue are about an order of magnitude lower than for polonium (Thomas *et al.* 1994, Tracy 1993c). ^{210}Po may also be taken up by marine shell fish (Hunt and Allington 1993).

Blanchard and Moore (1970) have reported ^{210}Po concentrations of up to 10 Bq·kg^{-1} in the livers of Alaska residents who eat caribou. Up to 5 Bq·kg^{-1} of ^{210}Pb were found in the rib bones of these people. An evaluation carried out by Health Canada under the AES/NCP showed that residents in some northern communities may be receiving up to 10 mSv of ^{210}Po per year through dietary sources. In comparison, the normal background radiation does is about 2 mSv per year (Tracy and Walsh 1995). Because these radionuclides are naturally present, it is likely that this has been the situation in the Arctic for several thousand years.

Most of this increased radiation dose from ^{210}Po is due to a changes in the estimated gastrointestinal absorption of polonium by ICRP (1994). This re-evaluation of the absorption factor by ICRP is based on one study of shellfish consumers in the United Kingdom (Hunt and Allington 1993) and should be replicated.

Radium-226 (226Ra)

Radium-226 is also a member of the decay series of naturally occurring uranium-238 (^{238}U). It is present in trace amounts in all rocks and soils. Uranium mining and processing operations may lead to enhanced local concentrations of ^{226}Ra in the environment. This has occurred in the Arctic at abandoned mine tailings at Port Radium, NWT and Rayrock, NWT (Veska and Eaton 1991); enhanced levels of ^{226}Ra have also been observed at trans-shipment points along the Great Bear and Mackenzie Rivers.

Radium-226 has a half-life of 1600 years. Fortunately, it is not very mobile in the environment, and

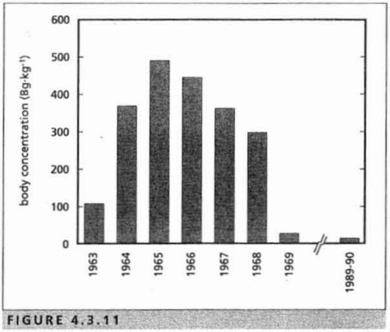

FIGURE 4.3.11

Body burdens of radiocesium — yearly averages across the Canadian Arctic.

rarely migrates far beyond the local source of contamination. It has not been observed to bioconcentrate in food chains. Like strontium-90, radium is deposited in bone. In the early part of the 20th century, bone cancer was found to be an occupational hazard among radium dial painters, although cancers did not occur until the body burdens of radium reached several tens of thousands of Becquerels (UNSCEAR 1977). By comparison, the background level of radium in the human body, including in Arctic residents, is about 0.1 $\text{Bq}\cdot\text{kg bone}^{-1}$ (ICRP 1975).

Radon-222

Radon-222 is a naturally occurring radioactive gas, ubiquitous in indoor air. This gas is not a problem specific to the Arctic. Generally, radon-222 is considered to be the largest single contributor to background radiation dose (NCRP 1987, Ahier and Tracy 1995). Depending on the properties of underlying rocks and soils, and on the type of construction, radon levels in some buildings may reach levels high enough to require remediation. There are not a large number of radon measurements in arctic dwellings, although radon is likely to be less of a problem in the Arctic than in southern Canada. Permafrost and snow cover tend to inhibit the emanation of radon from the soil. The northern practice of building homes on piles, rather than on a full concrete basement, also retards the build-up of radon in indoor air.

Abnormally high occurrences of lung cancer have been observed in uranium miners exposed to high levels of radon in the past (UNSCEAR 1977). Recent epidemiological studies on the association between lung cancer and elevated household levels of radon have proved inconclusive (Létourneau *et al.* 1994). Tobacco smoking, the chief cause of lung cancer, has always been a confounding factor in these studies.

- *The amounts of radionuclides in the arctic environment are generally about the same as, or lower than, levels found in the temperate zone. Of all radionuclides, lead-210 and polonium-210, which are natural in origin, make the greatest contribution to human radiation doses in the Arctic. Both occur in nature as airborne particles and tend to settle out on vegetation (i.e. lichens) thereby entering the terrestrial food-chain (lichens-caribou-humans). Residents in arctic communities may be receiving up to 10 mSv of* ^{210}*Po per year through dietary sources compared with normal background doses of about 2 mSv. This has likely been occurring in the Arctic for several thousand years. Of the anthropogenic radionuclides, the two main isotopes of radiocesium (cesium-137 and cesium-134) are considered to be of greatest concern in the arctic environment. Levels of radiocesium in Arctic residents have declined from about 450* $\text{Bq}\cdot\text{kg}^{-1}$ *in 1965 to roughly 10* $\text{Bq}\cdot\text{kg}^{-1}$ *in 1990.*

4.4 Assessing and Managing Risk

Assessing the human health implications of country food consumption requires a thorough evaluation of what is known about both the risks and the benefits. Weighing risks against benefits is extremely difficult. It is not possible to precisely quantify the degree of risk when there is insufficient scientific information about health outcomes at given contaminant exposure levels, nor is it possible to place a defensible quantitative value on the nutritional, social, and economic benefits of traditional food consumption. The process of risk and benefit assessment is referred to as "risk determination." It is carried out on a case-by-case basis and involves as much good judgement as it does good science. This results in what is often referred to as a "best estimate" of the degree of risk involved in a given activity.

This section explains the process of risk determination, with a focus on application of the process in arctic communities and the special considerations and difficulties faced in the risk management component. An important consideration in arctic communities is the role that risk perception plays in communication. Finally, an evaluation of current risks, and several case studies that illustrate risk assessments and management decisions made recently in arctic communities through the AES are provided.

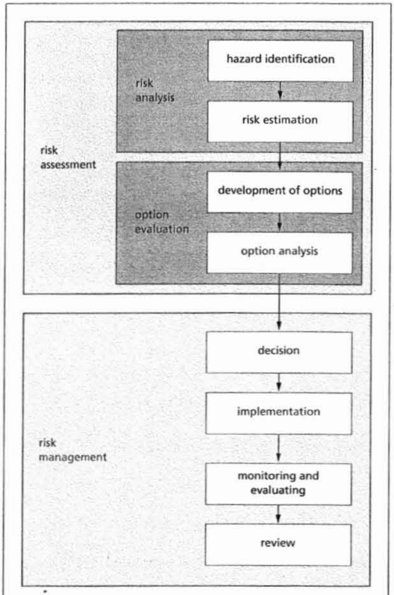

FIGURE 4.4.1

Risk determination (Health Canada 1993)

4.4.1 Risk Determination

Risk assessment and risk management are complementary but distinct processes that together are often referred to as "risk determination" (Figure 4.4.1.). Risk assessment involves the identification of the hazard (adverse health outcome) and the risk (chance) that will occur, followed by the development and evaluation of management options. Risk management involves selecting the appropriate options, followed by monitoring and evaluation. While the risk assessment process is based largely on scientific analysis and evaluation, risk management is a decision-making process that incorporates cultural, economic and other socially important values (Usher *et al.* 1995).

Risk determination is a constantly evolving process. New health outcomes and sociological and economic data constantly emerge and require that risk and benefits be reassessed. Risk determinations are only ever best estimates based on what is known and predicted, making the "review" stage (Figure 4.4.1) a critical feature of the process.

4.4.1.1 Risk Assessment

Risk Analysis

The first step in risk analysis is the identification of a hazard by establishing whether a cause-effect relationship exists. The hazard identification process provides information about the strength of the evidence that supports a relationship between an exposure and a change in health status. Once a hazard is identified, the relationship between exposure and the probability of an adverse effect occurring is assessed. This includes the evaluation of all pertinent toxicological data to establish dose-response relationships for contaminants — that is, identifying those exposure levels known to cause, and more importantly, not to cause toxic effects. The "no observed adverse effect level" (NOAEL) is defined as the dose at which no biologically significant adverse effects are observed in the study population compared to the controls. Well-designed and well-conducted epidemiologic studies with documented exposure assessments are preferred for identification of NOAELs. When these are absent, as they often are, controlled experimental animal bioassays (tests) are used.

While animal bioassays have advantages (defined exposure duration and routes, thorough quantification of toxic responses, collection of chemical toxicity data prior to human exposure), the results obtained have to be applied to humans who differ from experimental animals in terms of body surface area and size, lifespan, pharmacokinetics, etc. Typically, regulatory agencies have employed "uncertainty" or "safety factors" to compensate or adjust for the known physiological and biological differences between experimental animal species and humans, and the fact that there are a range of responses in any given human or animal population. The WHO and the Food and Drug Administration in the US have adopted a default 100-fold uncertainty factor to be applied to NOAELs obtained from animal bioassays (Waltner–Toews and McEwen 1994). The rationale is that humans could be up to 10-fold more sensitive to the toxic effects of a chemical compared with experimental animals and there is at least a 10-fold difference in susceptibility within human populations (inter- and intra-individual variability) (McColl 1989). Based in part on human response to drugs and to a wide range of toxic chemicals, it has been estimated that adopting an uncertainty factor that accounts for this 10-fold difference in individual susceptibility would provide protection for up to 95% of a population (Calabrese 1985). Additional uncertainty factors may be applied if the effects of the chemical are severe (e.g. cancer or birth defects vs. skin irritation or weight loss), or if the animal study used for determining the NOAEL was shorter than a lifetime exposure.

After identification of the appropriate NOAELs and uncertainty factors, exposure guidelines (acceptable or tolerable daily intakes, provisional tolerable daily or weekly intakes: ADI/TDI/PTDIs/PTWIs) can be calculated by dividing the NOAEL by the safety factor. These intakes, when averaged over the course of the entire human lifespan, are thought to represent the exposure for humans that is without appreciable risk of adverse effects, based on the best available information. The terms acceptable daily intake (ADI) and tolerable daily intake (TDI) have essentially identical meanings. Table 4.4.1 lists the TDIs/ADIs for a number of contaminants discussed in this report. These values are subject to change as new data becomes available.

The second component of risk analysis is risk estimation. Contaminant levels in the environment (air,

TABLE 4.4.1

Tolerable daily intakes (TDI) and acceptable daily intakes (ADI) for environmental contaminants[1]

Environmental Contaminants	Canadian ADI/TDI (reported in µg·kg bw^{-1}·d^{-1})
Metals	
Lead	3.57 (adult + child)[2,3]
Mercury	0.471 (methyl Hg) 0. 714 (total Hg)
Cadmium	1[2]
Persistent Organic Pollutants	
DDT/DDE/DDD	20 (S DDT + metabolites)
PCDDs/PCDFs	0.00001 (TCDD TEQ)
Mirex	0.07
HCH	0.3 S(α–HCH, ß–HCH, γ–HCH, δ–HCH)
PCBs (including planar PCBs)	1.0
Chlordane	0.05 S(*cis*–chlordane, *oxy*–chlordane, *trans*–chlordane, *cis*–nonachlor, *trans*–nonachlor)
Heptachlor	0.1 S(heptachlor + heptachlor epoxide)
Toxaphene	0.2
Aldrin/Dieldrin	0.1[4]
HCB	0.27
B(a)P	not available
Radionuclides	International Commission on Radiological Protection (ICRP)
^{137}Cs	Annual Limit of Intake (Bq·a^{-1})[5]–50,000
^{134}Cs	Annual Limit of Intake (Bq·a^{-1})[5]–50,000
^{90}Sr	Annual Limit of Intake (Bq·a^{-1})[5]–30,000
^{131}I	Annual Limit of Intake (Bq·a^{-1})[5]–40,000
^{210}Pb	Annual Limit of Intake (Bq·a^{-1})[5]–1,000
^{210}Po	Annual Limit of Intake (Bq·a^{-1})[5]–800
^{226}Ra	Annual Limit of Intake (Bq·a^{-1})[5]–4,500

[1] *Health Canada 1996. Values are based on current scientific information and may change as new data become available. These values are presently used by Contaminants Toxicology Section, Food Directorate, Health Canada, Ottawa*

[2] *Joint FAO/WHO Expert Committee on Food Additives, 1993*

[3] *This intake for infants will result in a blood-lead value of approximately 5.7 µg·dL^{-1}*

[4] *based on WHO values*

[5] *Annual Limit of Intake is the amount that, if ingested by an adult, would lead to a committed dose of one milliSievert, the limit for a member of the general public recommended by the International Commission on Radiological Protection (1994). [This limit is due to be adopted in Canada by the Atomic Energy Control Board.]*

soil, water) and in consumed foods are used to derive probable daily intakes (PDI) — that is, estimates of total exposure. The PDI is calculated by taking the total exposure from all sources divided by human body weight and is expressed as intake per kilogram body weight per day (intake·kg $bw^{-1} \cdot d^{-1}$). These PDIs are then compared with the exposure guideline values (ADI/TDI) to determine the level of risk in the population (i.e. how much the estimated exposure exceeds the tolerable exposure — TDI or ADI as the case may be). In cases where the PDI exceeds the TDI or ADI, risk management options need to be considered.

Occasionally there is a direct relationship between a contaminant concentration in the body (in blood, hair or an organ), a dietary intake, and an adverse effect. Where contaminant levels in human tissue are known, they can sometimes be used to estimate past exposure and potential risks.

For substances determined to be genotoxic carcinogens (DNA damaging agents which cause cancer), different risk assessment procedures are generally used. For the previously described process, it is assumed that there exists some level of exposure (a "threshold") where no measurable toxic response will be seen. For genotoxic carcinogens, due to the nature and irreversibility of the proposed toxic effect (cancer), a more conservative approach is taken, i.e., no exposure level is considered to be free of risk (non-threshold). The concept that no dose of a genotoxic carcinogen can be tolerated safely has led to the development of mathematical risk assessment procedures. Animal/human study data are used to mathematically predict the exposure doses for various cancer risks, usually the risk range used is a 10^{-5} risk (one additional cancer per 100,000 of population) to 10^{-7} risk (one additional cancer per 10 million of population).

The last step in the risk estimation process is the risk characterization. All of the information about the nature of the risk is reviewed and the major assumptions and major uncertainties involved in the risk analysis process are evaluated in order to assess the strength of the evidence provided by the risk estimation. For example, if high-dose occupational or animal experiments had to be used for the risk analysis then there would be significant uncertainty about the applicability of that data to low-dose and long-term human environmental exposure. In addition, certain assumptions made in risk estimations may not always be valid, for instance, the assumptions that a threshold exists for all toxic effects other than cancer, or that humans are more sensitive to toxins than other animal test species. It is possible that sub-threshold effects occur, or that humans are, in fact, less sensitive to certain contaminants than the test species.

The strength of evidence of a risk assessment can also be influenced by the presence of "covariables" in the studies upon which it was based. Covariables are other factors that may have influenced the outcome of a study and altered interpretation of the results. Covariables include age, genetic/family history, socio-economic factors, education, other disease conditions, lifestyle, and timing of exposure. Covariables can also occur in the form of voluntary exposures to substances (e.g. alcohol consumption, legal and illegal drug use, and smoking). Estimates of how much exposure through food is still safe must account for how much exposure is already occurring through other activities. Another type of "covariable" is the simple reality that human populations are exposed to mixtures of pollutants in the environment, not individual contaminants. Risk assessments are done on individual contaminants and little is known about the interactive effects of these contaminants. Covariables are also important to consider in animal studies although these studies are done in a more controlled environment and are not subject to as many potential covariables.

All of these factors must be considered when establishing the degree of uncertainty involved with the actual risk estimation. This characterization of risk becomes especially significant in the selection of risk management options.

Option Evaluation

The next step in risk assessment involves option development and option analysis. This phase of risk determination takes account of the specific nature of regional issues. Several options — including regulatory measures, advisory measures, and technological measures — are developed and evaluated based on the objectives of the risk determination procedure (Health Canada 1993). Common objectives include minimizing the risk of death, minimizing other adverse health effects, avoidance or elimination of the source of risk, mitigation either through reducing the likelihood of the event, decreasing frequency of exposure or lessening consequences of exposure, or acceptance of the risk through strategies to affect willingness to accept risk (Usher *et al.* 1995).

After all possible options are considered, the options are analysed taking into account public perception of risk, social, cultural and political implications of the option, and the uncertainties in the risk estimation (Usher *et al.* 1995). This provides risk managers with the information they need to propose a risk management strategy.

4.4.1.2 Risk Management

Decision

The results of the option analysis are used to select the most appropriate risk management option. In several risk assessments undertaken in the Arctic in the past, recommendations to continue consuming country foods have been made despite the potential for adverse effects from exposure to contaminants. In some cases, the safety factors surrounding the intake guidelines (TDI/ADI) are large because the science base is poor (e.g. for toxaphene) making it difficult to conclude definitively whether or not a small, or even a significant, exceedance of the intake guideline would constitute a real health risk. By contrast, the benefits of continuing to consume traditional food are often very great. Regardless of the decision taken, the health risks associated with exposure to contaminants remain even though they may be outweighed by the benefits of continued consumption. This poses a large and confusing public, moral and political dilemma.

Wheatley and Paradis (1996a) remind us that the risk management actions taken can do more harm that good, regardless of how well intentioned they are.

> … proceeding to the next step of acting on the theoretical risk can produce real health damage. Conversely, if the risk is real, not just theoretical, then it cannot be ignored and action should be taken. Hopefully ongoing research and discussions will help to resolve this risk management dilemma. In the meantime, there is a need to continue to take a broad, careful approach to risk management decisions which may have negative health effects, especially for populations with different lifestyles and exposure patterns …

Implementation

Once the preferred management option is selected, a strategy for its implementation is developed. All affected parties need to be take part in selecting the management strategy. A protocol has been established by the AES for the release of information related to contaminants in a prompt and understandable way (Figure 4.4.2). The communication of information must take into consideration cultural differences, language differences, and other potential barriers to comprehension such as individual perception of risk. Personal perception of risk can have significant impact on personal and community reaction to the information presented (see section 4.4.3).

Monitoring, Evaluating and Reviewing the Decision

Once a risk management decision has been made, its implementation is carefully monitored and evaluated to see if it is appropriate and effective. If implementation problems occur, or if knowledge of the hazard or the risks changes, the decision should be reviewed. This step is extremely important and illustrates how the process of risk management is an evolving process and is subject to change as new information about the situation is learned and assessed. The approach must be continually modified to suit each situation and each community.

- *Risk determination for contaminants in country food involves a consideration of the type and amount of food consumed and the sociocultural, nutritional, economic, and spiritual benefits associated with country foods. Risk management decisions must involve the community and must take all aspects into account to arrive at a option that will be the most protective and least detrimental to the communities. Regardless of the decision taken, some health risks associated with exposure to contaminants may remain. In the Arctic, these risks are outweighed by the benefits of continued consumption of country food. The uncertainty over risks and benefits often poses a large and confusing public, moral and political dilemma.*
- *Risk management is an evolving process subject to change as new information about the situation is learned and assessed. The approach must be continually modified to suit each situation and each community and the advice monitored to ensure it is providing the best possible health outcome.*

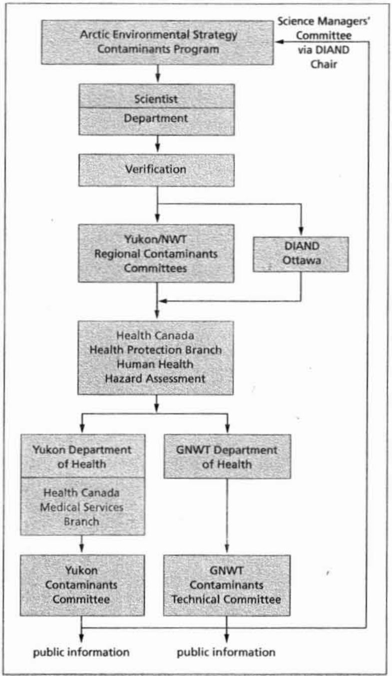

FIGURE 4.4.2

AES Protocol for contaminants health and harvest information release in the NWT and Yukon.

4.4.2 Special Considerations for Risk Management in Arctic Communities

Recent studies of the socio-economic effects of country food contamination in remote Aboriginal communities (Usher *et al.* 1979, Wheatley and Wheatley 1981, Weinstein and Penn 1987, Usher 1992, Scott 1993, Usher *et al.* 1995) have identified several distinct challenges and difficulties in successfully dealing with the contamination of country food.

Risk management decisions concerning the contamination of country food must take into consideration that in Aboriginal communities avoidance of one or more food items can have extensive implications. For most Canadians, the production and distribution of food is divorced from its consumption. Because of this separation of production and consumption, food safety is normally achieved by regulation and inspection of producers and processors, rather than issuing health advisories to consumers. To advise against the consumption of country food in northern Aboriginal communities, however, is also to advise against hunting and fishing. To the extent that Aboriginal identity and the collective sense of well-being is based on subsistence as a social system and as an activity, as well as a dietary staple, then loss of confidence in the quality of country food undermines confidence in identity and society.

For example, the identification of mercury contamination of Aboriginal fisheries in the subarctic was in every known case accompanied by a general decline in subsistence fishery participation and fish consumption. Disruption in these cases occurred gradually, over months or years, because the extent and consequences of contamination was only slowly revealed and communicated. The cumulative effects included fear, suspicion, and hypochondria. People were especially concerned about the health of their children and about the future in general. Explanations were sought for whatever went wrong (most particularly for unusual symptoms or suspicious deaths), or people believe that things had changed for the worse, and contamination was a ready and plausible reason. In the absence of clear and understandable information, "mercury gossip" became the chief determinant of behaviour (Usher 1992). Often, sensational media stories added to the cumulative effects and resulted in virtual cessation (which may be temporary) of eating of traditional foods, especially sea mammals and fish (Wheatley and Wheatley 1981, Wheatley 1994).

Under such circumstances, individual estimation of risk is consequently based on incorrect or untested assumptions, and is frequently wrong, leading to social and economic results that are harmful and undesirable, if not also medically injurious. The collective result may be the abandonment of subsistence to such a degree that the social integrity of the community is seriously compromised. The most extreme example is probably that of Grassy Narrows and Whitedog, Ontario, where elevated rates of mortality, morbidity, and social pathology occurred directly following the destruction of the local fishery by mercury contamination in the early 1970s (Usher *et al.* 1979, Wheatley 1979, Erikson 1994). Although this connection has rarely been examined systematically, the general phenomenon of adverse socio-economic effects of harvest disruption, however caused, has been widely noted (Fienup–Riordan 1986, Wenzel 1991).

For Aboriginal Northerners, virtually any disruption in the use of country foods affects the one part of the local economy over which they exercise the greatest control. This control is critical because few other economic opportunities are present in the North and substitute foods at reasonable cost are not readily available. It is also critical because the loss of country food resources (or the threat of such loss) undermines the basic cultural identity and independence of Inuit and Dene/Métis. In comparison, non-Aboriginal arctic residents are much less vulnerable to exposure to environmental contaminants. They eat much less country food, and it does not have the same social and cultural significance for them.

- *In Aboriginal communities, advising against country food consumption is also to advise against hunting and fishing. To the extent that Aboriginal identity and the collective sense of well-being is based on subsistence as a social system and as an activity, as well as a dietary staple, then loss of confidence in country food undermines confidence in identity and society. If not released with proper communication and consultation, advisories related to country can also result in individual estimations of risk that are often based on untested assumptions and are frequently wrong, leading to harmful and undesirable social and economic results. This may result in the abandonment of subsistence to such a degree that the social integrity of the community is seriously compromised. As such, risk management decisions must be carefully considered and must be implemented in ways that minimize the risk of adverse effects. Options that minimize the extent to which nutritional and sociocultural aspects of Aboriginal societies are compromised should always be considered.*
- *Regardless of the difficulties in the processes of risk assessment and risk management and the different views on their adequacy, we must be guided by one objective. Risk assessment and management decisions are undertaken to serve public health. They must be our "best estimate" and seen in the context in which they were created — an imperfect and ever-changing data base.*

4.4.3 Risk Perception and its Implications in the Arctic

For risk management decisions to be met with the support of the affected individuals, these decisions must respond to the preferences and beliefs of the communities. Assessment of public perception is necessary for decision makers to evaluate the significance to the public of health risks, and the potential success of proposals to reduce these risks. Adequately addressing public perception helps minimize anxiety and avoid any negative repercussions in situations where risk management decisions are implemented.

It has been widely noted that the public does not see risk in the same way as "the experts" (Kasperson 1986, Erikson 1990, Douglas 1992). Risks that are imposed (especially by perceived intent or carelessness), involuntary, perceived to be unfairly shared, not easily controlled by individuals, or associated with anthropogenic hazards or exotic technologies appear greater than more familiar risks (Slovic *et al.* 1982, Douglas 1986). Perception is also influenced by the degree to which it is believed that there will be a benefit as a result of incurring the risk. This disparity in risk perception between public and experts is also evident in the North. It is compounded, however, by the exceptional susceptibility of Aboriginal peoples to contaminants in the Arctic region by virtue of the cultural significance of traditional food and the largely involuntary nature of consumption of contaminated food (as noted in section 4.1). Factors such as language, education, and knowledge transfer systems (discussed in section 4.4.4), economic need, and the apparent invisibility of the risk (Wheatley 1995, 1996) also add to the disparity in risk perception between the public and "experts."

A Santé Québec survey of Nunavik Inuit assessed individual perceptions of environmental contaminants, namely PCBs (Dewailly *et al.* 1994). Awareness of PCBs in Nunavik has developed as a result of at least three factors: national media coverage of the issue of PCB management; a local study of PCB concentration in mothers' milk (Dewailly *et al.* 1989); and the publication of results of a nutrition survey conducted in the NWT that caused alarm among the Inuit population (Allen 1991, Usher *et al.* 1995). The Santé Québec survey revealed that nearly two out of three Nunavik Inuit (62%) had heard of PCB contamination of the food chain, and that the vast majority of respondents (87%) wished to have more information on the subject. Overall, 55% of those surveyed thought that commercial foods were less healthy and less nourishing, while 21% believed that commercial foods were of higher quality either because they were better or more modern or because traditional foods were contaminated. The perception of traditional foods varied significantly according to age with the most favourable attitudes in the 25 to 44 age range. The least favourable attitudes were noted among the young. No variation based on other sociodemographic factors was observed. The survey also indicated that roughly 14% of respondents reported a change in their habits upon becoming aware of PCB contamination of country food, 10.9% reduced their consumption of country food, and 3.5% discontinued consumption altogether (Dewailly *et al.* 1994). This survey did not assess the duration of these responses.

A review of 13 past incidents of country food contamination (Usher *et al.* 1995) noted some distinctive features of the problems encountered with public perception in risk management strategies undertaken in the North prior to the early 1990s. In most of these cases, the community was alerted to the possibility of contamination only by notification of the results of laboratory analysis of animal tissue samples, sometimes obtained for purposes not directly related to local food safety. Otherwise, there was no tangible evidence of hazard. Except for instances of gross visible pollution (for example following the Exxon Valdez oil spill in Alaska), it was only after the presence of the contaminant and its possible toxic effects had been made known to people, that they began to suspect that previous unexplained events, and particularly abnormalities in animals, fish, or humans, might be related.

Almost all of the cases reviewed gave rise to local uncertainty and anxiety. This seems to be related to the absence of straightforward and credible information on toxicity (especially in cases of chronic but low-level exposure) or safe levels. Health authorities tend to set standards with a substantial safety factor. However, this may compound local concern because it suggests that quite small amounts of everyday food are a significant health risk. Where there is fairly strong evidence of low risk (e.g. the cadmium case studies), or of benefits outweighing the risks (e.g. the recent health assessment regarding marine mammal fats), and this is clearly communicated, the problem of uncertainty and anxiety appears to be minimized.

The connection between increased anxiety about the status and health effects of country food, and the actual consumption, is complex. Significant reductions in country food consumption can occur when people are informed of "invisible contaminants" in the local food supply. The term "invisible contaminant" refers to the fact that the contaminant itself, and often its effects on the ecosystem, are not easily detected. Their identification is solely or primarily from scientific monitoring and analysis. Further, the effects on humans, if any, may be delayed for many years and be easily confused with similar effects from other causes. Invisibility alone is not necessarily problematic, because Aboriginal Northerners have long guarded against such hazards as botulism, trichinosis, and vitaminosis, using their experience and traditional knowledge. However, the combination of invisibility, uncertainty, and the long-delayed effects that typifies many chemical or radioactive contaminants, means that community residents have only the word of outside experts that their daily food may be hazardous. They are often unsure as to how they should respond to that hazard. They have had to rely on communications from people whose language and culture are difficult to understand and whose judgements and advice are not common community knowledge. Furthermore, there are frequently competing claims by different experts about the nature and extent of the risks (Elias and O'Neil 1995).

At Lake Laberge, Yukon, the identification of toxaphene in burbot liver and lake trout resulted in concern among the Ta'an Kwach'an First Nation who fish the lake, leading to a reluctance to eat any fish from the lake including migratory salmon (Wheatley 1994a). Similarly, at Salluit, Québec, fear of mercury contamination of traditional food, encouraged by misleading media reports, resulted in Inuit residents ceasing to eat those foods (Wheatley and Wheatley 1981).

However, not all cases of reported contamination have led to decreased country food consumption. A review of contaminant episodes (Usher *et al.* 1995) suggested that identifiable local sources of contamination caused greater social disruption than contamination by long-range transport, perhaps due to more clearly identifiable human involvement and liability, often coupled with apparent inaction, or inappropriate action by those seen responsible for correcting the situation (Usher *et al.* 1995). Residents of the affected communities expected that government and industry would acknowledge the problem, act to clean it up, and restore the environment and their own well-being. Legal action was initiated against the polluters at some point in each, leading eventually to protracted negotiations resulting in remedial or compensation agreements.

There is little evidence that concern over the safety of country food has lead to long-term or sustained reductions in country food consumption (as indicated, for example, by repetitive annual harvest surveys). The exceptions have been where health authorities clearly advised against continued consumption and especially where this was supplemented by evidence of the effects of contaminants (such as the mercury poisoning that occurred in Minimata, Japan in the 1970s — Usher *et al.* 1979, Wheatley 1979, JBNQ 1982), or where there has been visible evidence of contamination with obvious direct consequences (such as oil spills and local industrial contamination — Usher *et al.* 1995).

- *Perception of risk in the Arctic, as in many communities, differs between the public and experts. The predominant effect of inaccurate perception of risk is uncertainty and anxiety often leading to virtual cessation of country food consumption. In many cases, this seems to be related to a lack of straight-forward and credible information about toxicity or safe levels. Adequately addressing public perception can help to minimize anxiety and lessen any negative repercussions from the implementation of risk management decisions.*

4.4.4 Risk Communication in the Arctic

When a problem of contaminants in country food occurs, remedies are most likely to be effective if those concerned have a sound and shared appreciation of the problem and its solutions. This cannot occur without effective communication. To be effective, communication strategies must take into account how people perceive risk and respond to potential hazards. They must also acknowledge cultural differences in perceptions, beliefs and behaviours and differences in communication patterns. Good communication is fundamental to the aims of AES contaminants research.

In their review of several past contaminant incidents in Aboriginal communities, Usher *et al.* (1995) pointed to a deficiency in communications, planning, and evaluation. Communication was typically ad hoc — uncoordinated, and often not budgeted for at the outset. The same study also examined knowledge and information needs in three Inuit communities in the NWT and reported that community members are generally aware of the existence of contaminants, but are unsure where they come from, how they are taken up in the food chain, and what effects they have on humans. People learned of contaminants through the media, especially TV and radio, from pamphlets in local health centres, and from other community members. Such information is usually fragmentary and some people lack the context for the information. There is a tendency to mistrust the word of outside experts who are not personally known and accepted, and to rely primarily on the advice of elders and hunters on matters concerning country food.

Overall, community residents wanted two types of information about environmental contaminants. Where contaminants are regarded as a local problem, people wanted a quick and definitive diagnosis of food safety even though in most cases no absolute statements of this type can be made. They were not satisfied with technical, quantitative information intended to enable them to calculate for themselves the probabilities of food safety. The more threatening the situation, the greater the demand for clear, unambiguous information, and for full disclosure of all available information. Attempts to reassure community residents without providing convincing evidence, or by withholding potentially distressing information, heighten suspicion and mistrust, and make communication difficult.

Many community residents also sought a more general understanding of contaminants: where do they come from, where do they go, and what harm will they do? They wanted both types of information explained in direct and simple ways, by credible sources. Pamphlets and videos are better than dense reports, but interactive communication through media such as radio phone-in shows or through face-to-face discussions, is even more assuring. Public presentations that allow for limited interaction are seldom regarded as useful.

The study concluded that effective communication emphasizes a two-way or multi-directional flow of information, where there are maximum opportunities for feedback (Usher *et al.* 1995). Effective communication requires an exchange of information, ideas or feelings. Communication is not simply media products, such as reports, pamphlets, posters and audio-video materials. These have a role to play, but they should be seen as means to an end (effective communication), not as ends in themselves. They should complement or help facilitate authentic dialogue and exchange. They are not a substitute for discussion. When considering various communication materials, the question is not which product, medium or channel is best, but rather how each can be used to stimulate dialogue, learning and action. Communication is a process, not simply a product. As a social process, communication is often most effective when it is interpersonal and face-to-face, the preferred mode of communication of most Aboriginal people.

In the context of the AES, communication should provide empowerment. Communication should serve to raise awareness, facilitate understanding and enable people to take action or make informed choices. Ideally, communication is an exchange between equal partners. This involves challenges. It also requires patience and respect for the knowledge, perceptions, concerns, and priorities of the public.

Ideally, a contaminants communication program should be based on accurate scientific knowledge and offer a clear workable prescription for how to deal with the perceived problem. However, research and monitoring may not provide clear answers, much less reassuring ones to the lay public. Communicating risk should be a continuous and ongoing process.

Communication becomes more difficult in communities where English is not widely used — this is particularly relevant in Inuit communities (except for the Inuvialuit region). There is no commonly understood framework of concepts and language with which to understand the nature of chemical contaminants. Terms or phrases selected by interpreters of words relating to contaminants are not always widely understood, and do not often make sense (Wheatley 1995). Such concepts and terminology cannot be developed in Aboriginal languages until translators and interpreters understand them well in English. Furthermore, the level of scientific education and knowledge in isolated communities tends to be low, and there is often little experience of chronic, low-level pollution and contamination and their effects. Early attempts to provide translated information about food-borne contaminants were often misleading and confusing (Usher *et al.* 1995). Some recent information tools (videos, posters, etc.) developed in the NWT under the AES program have been translated with apparent success.

Whatever language is used, the information must be presented in a non-technical form, using concrete imagery (instead of abstract statistics) that is understandable to local residents. Understandable language helps to reassure communities if there is no problem, to prevent panic if there is a possible risk, or to justify advisory notices if they are deemed necessary (Wheatley 1996). When information is properly communicated it can serve to promote the cultural and nutritional benefits of traditional foods while educating the public of priority contaminants.

A further difficulty can occur in the differences of the knowledge systems used in the scientific communities versus the traditional knowledge system. Scientific risk assessment is based on hypothesis testing by measurement and statistical analysis. Traditional knowledge, on the other hand, is based on cumulative experience, trial and error, and close observation, which is shared and interpreted locally, and supplemented by knowledge handed down over generations (Wheatley 1996). For contaminants research to become relevant to northern communities, attention must be focused on how these two knowledge systems can be integrated in pragmatic and tangible ways. Otherwise, the risk is the continued rift between the lay community and scientists.

The current research focus is not so much on community vs scientific data interpretation but rather on using the tools for collaborative research, including community involvement — at the initial research — design stage (Wheatley 1994a). Without community involvement, research will often be viewed as "...research on Aboriginal people by representatives of the external society that is invasive and destructive" (O'Neil 1994). The AES Research Guidelines recognize this important dynamic and have sought to encourage the development of community based research initiatives.

- *Transfer of accurate and complete information via good communication plans may help limit the social and cultural effects resulting from the presence of contaminants in traditional food. If this does not occur, people will be forced to draw their own conclusions and will act accordingly based on their perceptions of the situation.*
- *In arctic communities, communication is most effective when it is interpersonal and face-to-face. It should be a*

two-way flow of information where the opportunity for feedback is maximized.

- *Communication should occur from the onset of a study and should be an ongoing process through to the reporting of findings and the development of remedial options. The best studies and the best solutions to local contaminant problems are developed with and by the community.*

4.4.5 Risk Determination Conclusions in the Arctic

All of the contaminants discussed in this document have the potential to affect health adversely. Some levels may already be causing effects to humans in the Arctic. However, the damage is difficult to measure, and it is difficult to state with certainty which chemical or confounding factors caused the effect, or the definitive study has not been (or cannot be) undertaken in Arctic communities.

What we do not yet know about contaminants, or may never be able to measure in the Arctic, cannot be used as a rationale for avoiding our responsibility to protect the health of arctic residents. Nor should what we know — or think we know — be used recklessly in a way that may frighten people who can exert very little control over their exposure levels. It is known that exposure to contaminants in the Arctic must decline and that the benefits of traditional ways must be sustained.

4.4.5.1 OCs

The highest accumulations of OCs are found in the fat tissues of marine mammals that are predators at the highest trophic levels such as seal, walrus, whale and polar bear (Muir *et al.* 1992, see Chapter 3). The level of exposure to contaminants depends not only on the concentration of the contaminant(s) in a particular food, but also upon the extent of use of that food. Consuming a great deal of a food with low levels of a particular contaminant may present similar risk to consuming small amounts of a food with a high level of that contaminant. For example, from Table 4.4.2 it can be seen that whitefish flesh has a relatively low concentration of toxaphene, yet it is the most important source of toxaphene in diets of Sahtú Dene/Métis women because it is widely consumed. Conversely, narwhal blubber has relatively high concentrations of PCBs, yet it accounts for only 11% of PCBs in the diet of Inuit women because it is less frequently eaten (Table 4.4.2). Thus, the entire diet, and not simply those food items known to have elevated contaminant levels, must be considered when assessing risks and benefits of traditional food and diets.

Food preparation is another important factor for the estimation of total oral exposure of organic contaminants from traditional food (Chan *et al.* 1996). Data from Qikiqtarjuaq (Baffin Island community), and two Dene/Métis communities in the 1980s showed considerable differences in levels of organochlorines in raw vs. prepared samples. An ongoing AES project is studying the effects of traditional food preparation (frying, boiling, aging, liquifying, drying, etc.) on organic contaminants in Inuit traditional food including walrus blubber, seal meat and blubber, narwhal meat, mattaq, and blubber. Preliminary results show that levels of some organochlorine compounds may be decreased in prepared food samples (Kuhnlein, personal communication).

Food consumption survey data can be combined with food contaminant concentrations to develop

TABLE 4.4.2

Arctic foods with highest organochlorine contributions to women's diets and corresponding concentrations of Chlordane, DDT, Toxaphene and PCBs (μg·100g^{-1})[1](Kuhnlein *et al.* 1995c)

	CHL[3]		DDT[4]		Toxaphene		PCBs[5]		
Food	% total intake	Conc.	% total intake	Conc.	% total intake	Conc.	% total intake	Conc.	Contribution rating[2]
BAFFIN INUIT									
Walrus blubber	37.0	204.5	21.6	141.9	23.2	212.4	28.8	337.1	High (27.6)
Narwhal blubber	15.5	213.8	18.5	272.6	44.5	843.4	11.0	320.5	Medium (22.3)
Ringed seal blubber	16.2	33.0	20.1	32.7	1.6	16.0	12.9	36.4	Medium (12.7)
Narwhal muktuk	9.5	11.2	18.5	52.2	15.2	83.1	15.5	64.9	Medium (14.7)
Ringed seal meat	9.0	1.8	10.0	1.8	1.3	0.6	15.1	4.7	Medium (8.8)
Caribou meat	1.0	0.2	0.7	0.2	0.7	0.3	4.6	2.5	Low (1.7)
SAHTÚ DENE-MÉTIS									
Whitefish flesh	28.8	0.7	12.0	0.5	39.3	3.9	22.3	5.9	High (25.6)
Caribou meat	4.7	0.03	13.6	0.06	0.0	0.0	18.3	0.4	Medium (9.1)
Inconnu flesh	18.4	2.1	11.4	0.7	23.6	9.1	15.9	3.2	Medium (17.3)
Black scoter meat	7.1	0.1	5.6	0.08	2.8	0.1	3.7	0.3	Low (4.8)
Moose meat	3.8	0.04	5.2	0.03	0.0	0.0	7.6	0.3	Low (4.1)
Loche flesh and liver	2.5	4.4	2.7	1.85	2.6	7.8	4.3	4.3	Low (3.0)

[1] *Values are approximate means of food as consumed following various preparations*

[2] *High: >=20% total intake, Medium: 11–20%, Low: <=10%. Rating is based on average (in parentheses) of all four contaminant contribution values*

[3] *Chlordane = sum of oxychlordane, cis+trans chlordane, cis+trans nonachlor, heptachlor epoxide*

[4] *DDT = sum of p,p'-DDE, p,p'-DDD, p,p'-DDT, o,p'-DDT*

[5] *PCB = sum of congeners*

estimates of exposure in the Arctic. When these estimated probable daily intakes (PDIs) are compared with currently held acceptable daily intakes (ADIs or TDIs) it is possible to gauge the relative, but not the absolute, risk to which people are exposed. Exceeding the TDIs are a problem for an individual if they are exceeded by that individual every day of their life. Much of the data in the Arctic is only for one day's exposures to contaminants. These estimates may over estimate lifetime exposures but do give a good indication which contaminants may be of concern.

General calculations that cover the entire Arctic region are unrealistic. As previously indicated, different Aboriginal groups and different communities in regions of the Arctic eat different species, parts of species and amounts of traditional food annually and seasonally. However, more specific calculations can be made for some groups in some subregions where the data are adequate and these are illustrative of some of the exposure patterns in the Arctic.

Kuhnlein *et al.* (1995c) plotted the range of individual one-day contaminant intakes of seven OCs for Baffin Island Inuit and Mackenzie Valley Sahtu Dene/Métis (Figure 4.4.3). Among Inuit, the mean daily contaminant intake values are higher by an order of magnitude or more for every contaminant shown compared with the Sahtu Dene/Métis. This is expected from the different amount and type of traditional food consumed by these two peoples — that is, marine mammals and fish for the former and caribou and fish for the latter.

These mean contaminant intakes can be compared to current TDI values (Figure 4.4.3). For the Inuit group, several of the mean intake values (chlorobenzenes, HCH, dieldrin and PCB) are within a factor of 10 of the TDI. For chlordane and toxaphene, the mean intakes exceed the TDI by nearly ten-fold. Currently, toxaphene and chlordane both have uncertainty factors of 1,000. For the Sahtu Dene/Métis the estimated mean daily contaminant intakes are well below the TDI values except for toxaphene and chlordane which are only approximately ten-fold below the TDI (Figure 4.4.3).

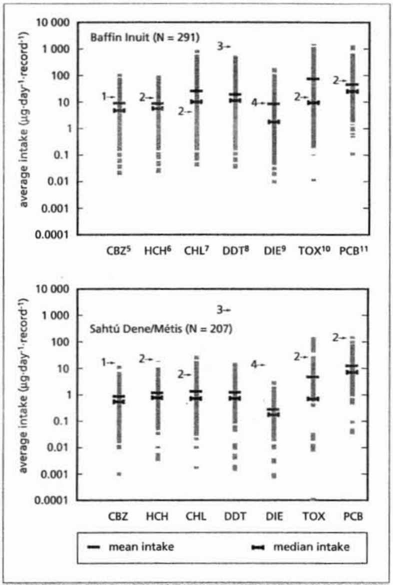

FIGURE 4.4.3

Distribution of OC exposure from traditional food consumed by Arctic Indigenous women. The group mean and median intakes are presented in comparison to reference values: (1) provisional tolerable daily intake (PTDI), Health Canada; (2) tolerable daily intake (TDI), Health Canada; (3) acceptable daily intake (ADI), World Health Organization; (4) tolerable daily intake (TDI), World Health Organization.

[5] *sum of tetra, penta and hexachlorobenzene;*

[6] *sum of alpha- beta- and gamma-hexachlorocyclohexane;*

[7] *sum of oxychlordane, cis and transchlordane and heptachlor epoxide;*

[8] *sum of p,p'DDE; p,p'DDD, p,p'DDT, o,p'DDT;*

[9] *dieldrin;*

[10] *toxaphene;*

[11] *PCB sum of congeners.*

(Adapted from Kuhnlein et al. *1995c)*

TABLE 4.4.3

Proportion (%) of women whose daily intake of contaminants exceeds the tolerable daily intake (TDI)

	CBZ[3]	HCH	CHL	DDT	DIE	TOX	PCB	Hg[4]	Cd[5]
INUIT –									
Baffin Island[1]	9	7	48	0	16	40	16	29	21
Nunavik[2]	6[7]	N/A[6]	75	0[8]	N/A	N/A	4	37	N/A
DENE/MÉTIS Mackenzie Valley[1]	0	0	6	0	0	2	0	N/A	N/A

[1] *Receveur 1996b (dietary interview 1987–88)*

[2] *Ayotte, 1996 (dietary interviews 1992)*

[3] *See figure 4.4.2 for definition of OC abbreviations*

[4] *Mercury*

[5] *Cadmium*

[6] *not available*

[7] *Hexachlorobenzene only*

[8] *DDE only*

Using different methodologies (several 24-hour recalls vs. single 24-hour recall and food frequency questionnaire), Dewailly *et al.* (1996c) also estimated contaminant intakes among Nunavik Inuit women. Ayotte (1996) and Receveur (1996b) have taken the data presented in Dewailly *et al.* (1996c) and Kuhnlein *et al.* (1995c), respectively and calculated the proportion of women in the studies whose dietary contaminant intakes exceeded the TDI (Table 4.4.3) The Inuit in these two studies exceed the ADI/TDI much more frequently and to a greater extent than the Dene/Métis. Although the data sets are incomplete, chlordane, toxaphene mercury and, to a lesser extent, cadmium and PCBs show the most frequent exceedances of the ADI/TDI. These exceedances of some contaminant ADIs/TDIs by Aboriginal groups in Arctic Canada are in contrast to more southern North American populations where the market diet is considered to have negligible amounts of OCs (Connacher and Mes 1993) and in the US where the daily intakes of OCs are usually less than 10% of the ADIs (Gunderson 1988).

The surveys of the two Inuit populations show quite similar results despite the methodological difficulties in comparing food contaminant intake studies. The possible differences in the proportion of ADI/TDI exceedances for chlordane and mercury between the Nunavik and the Baffin Inuit may be due to dietary differences with one group consuming more beluga whale and ringed seal and the other Inuit group consuming more walrus, narwhal and ringed seal (Dewailly *et al.* 1996c, Kinloch *et al.* 1992).

TABLE 4.4.4

Ratio of the concentration of contaminants in Inuit mothers' milk from Nunavik compared with non-Aboriginal mothers' milk from southern Québec

Contaminant	Ratio (North/South)
DDE	5.5
Toxaphene	N/A
Dioxins/Furans (dioxin TEQs)	2.0
Dioxins/Furans/PCB (dioxin TEQs)	2.4
Mirex	10.0
Chlordane[1]	10.0
Hexachlorocyclohexane	N/A
PCB (as Aroclor 1260)	5.6
HCB	4.9

see Section 4.3.1, data collected in 1989/1990

[1] *Comparison between Nunavik (1989/1990) and southern Canada (1992)*

N/A = not available

TABLE 4.4.5

Infant intake of contaminants (infant PDI) via mother's milk relative to the current adult TDI

Contaminant	Ratio of infant PDI/Adult TDI
DDE	0.25
Toxaphene[1]	5.0
Dioxin/Furans (2,3,7,8-TCDD TEQs)	8.6
Dioxin/Furans/PCBs (2,3,7,8-TCDD TEQs)	47.6
Mirex	1.0
Chlordane	0.33
Hexachlorocyclohexane	N/A
PCB (as Aroclor 1260)	13.1
Hexachlorobenzene	2.3

Human milk data from Nunavik Inuit women, 1989/1990 (Dewailly et al. *1992)*

[1] *Data from Stern* et al. *1992*

Support for these data can be found in the cord blood data cited in 4.3.1.11. Since all of the contaminants listed in these studies are persistent, bioaccumulative and cross the placenta, they can be expected to be present in the blood lipid of newborn babies. All of these contaminants occur at higher concentrations in cord blood of Inuit babies from both Nunavik and the western NWT than from Métis/Dene babies. The data also suggest that PCB and DDE levels in Inuit from the eastern Arctic are higher than those in Inuit from the western NWT. In addition, back-extrapolation from maternal blood collected in Nunavik confirm that for those contaminants with well known half-lives and absorption factors, estimated maternal exposures were in the same order of magnitude as those calculated from the dietary intake data shown here (Dewailly, personal communication).

Mothers' milk data comparisons provided in Section 4.3.1 clearly demonstrate that increasing concentrations of all OCs reviewed are associated with increasing consumption of traditional food (in particular fish and marine mammals). The concentrations of OCs in Inuit mothers' milk are 2 to 10 times greater than the levels in milk of southern Canadian women (Table 4.4.4). It is worth noting that even though breast milk levels are considerably higher in the north than the south, the levels of at least one contaminant-DDT, for which there are historical data, indicate that levels in southern Canadian women in 1967 were triple the levels now being reported in Inuit mothers (Mes 1994).

Cord blood data also raise a warning flag in the Arctic. Knowledge of effects of contaminants in animals and humans during fetal development is building and makes a compelling case for reducing population exposures as quickly and as sensitively as possible. Data in 4.3.1.11 indicate that a proportion of Inuit newborns and a smaller proportion of non-Inuit newborns have levels of PCBs in their cord blood that exceed a level of concern. The fetus is vulnerable to the effects of several contaminants, and neither it nor the mother can do anything to reduce exposure during this critical period of cell differentiation and organ development.

Very little is known about the effects of exposure of infants to moderately high levels of OCs during the breast-feeding period. The highest DDE levels reported in the Arctic, are more than 50% lower than the level associated with direct effects on neonatal reflexes and breast-feeding duration (Rogan *et al.* 1986, 1987). Jacobson *et al.* (1992) did not find an association between breast-feeding and developmental deficits in his Michigan fish consumers study. One benchmark that could be applied is a direct comparison between the estimated intake of a 5 kg infant consuming 750 ml of human milk a day containing 3% (22.5g) of fat with current adult TDI values. Clearly TDI values are developed, unless otherwise stated, for adults and are based on a 70-year lifetime exposure. Infants are highly exposed for a short period of life (0.3–2 years) and the toxic effects of exposure during this period of development may be quite different to the toxic effects of the contaminant on a fully developed adult. Thus the comparison of infant exposures to adult TDI is only illustrative not instructive. Table 4.4.5 demonstrates the relative ranking of exposures, and illustrates that total dioxin toxic equivalents (with and without the dioxin-like PCBs), PCB (as Aroclor 1260), toxaphene and hexachlorobenzene are of most concern for infants in the north.

There are many recognized advantages to breast-feeding to infants and to mothers, including improved nutrition, increased resistance to infection, protection against allergies, better parent-child relationships, and possibly a degree of protection of the mother against breast cancer. In the Arctic, alternatives to breast-feeding, such as infant formula, can be difficult to obtain (due to availability and affordability) and can pose difficulties with respect to the maintenance of hygiene in cases where the water supply is compromised. With full regard for the uncertainty over the toxic effects of OCs in human milk, the known benefits of breast-feeding are extensive and serves as a strong rationale for advising mothers to continue to breast-feed their newborns unless cautioned by their local health care worker to reduce or stop.

Evaluating the risks to health posed by the mixture of OCs in traditional foods, especially those from the marine environment, is complex. Clearly, some individuals from some Inuit communities (and some community mean values) exceed "acceptable" exposure levels. Yet the estimates available are based on dietary recall data from individuals for only one to six days a year at best. The different safety margins built into the ADI/TDI values and the uncertainty over the toxic effects of long-term exposure to most contaminants, especially toxaphene and mirex, also have an impact on how to evaluate these findings. An additional concern is the effect that mixtures of contaminants, some of which have similar toxic effects (on ability to cause cancer in animal studies, or to act as weak, endocrine disrupters) may exert on human populations. And there is the issue of how best to protect the developing infant when the scientific data are uncertain. There are no simple scientific answers to these questions in the Arctic.

Whenever an ADI/TDI value is exceeded on a regular basis, in the north or the south, it is prudent for health officials to take measures to reduce exposures. In the Arctic, any dramatic reductions of consumption of traditional food would have far reaching effects on nutrition, economic independence, social and cultural values and spiritual integrity. In short, dramatic reductions in traditional food harvesting and consumption would dramatically affect health.

Management decisions for OCs developed in partnership with all affected groups in the Arctic will need to focus on three strategies.

First, all management options must be based on a case-by-case, community-by-community approach. Co-operative risk management, with all parties at the table, must continue to be the standard for decision making. Elders and health care workers must be part of management decisions for communities, if the management approach is to be understood and applied.

Second, short-term solutions may require that some members of some communities reduce their intake of the one or two food items that contribute the most to their personal exposure. This approach may involve reducing portion sizes or frequency of consumption of some food types such as seal or narwhal blubber in the eastern Arctic and employing food preparation methods that reduce contaminant levels (see section 4.4.5.1). This approach will ensure that the contaminant related risks are reduced and the nutritional, social, cultural, and spiritual benefits retained.

Third, a long-term strategy must be put in place to reduce global use and long-range transport of these contaminants into the Arctic. Many of these initiatives are under way through the United Nations Environment Program (UNEP), the United Nations Economic Commission for Europe (UN-ECE) Protocol on POPs, and the North American Free Trade Agreement's Commission on Environmental Cooperation (NAFTA–CEC) activity on PCB, chlordane, DDT, and mercury.

- *There are many recognized advantages of nursing for both infants and for mothers, including improved nutrition, increased resistance to infection, protection against allergy, better parent-child relationships, and possibly a degree of protection of the mother against breast cancer. In the Arctic, alternatives to breast-feeding, such as infant formula, can be difficult to obtain (due to availability and affordability) and can pose difficulties with respect to the maintenance of hygiene in cases where the water supply is compromised.*
- *The OCs of primary health concern at this time for Inuit adults are chlordane, toxaphene, and PCBs. Exposures in the eastern region are higher than in the western region.*
- *For Dene/Métis, exposure to OCs is in general below a level of concern. However, levels of chlordane and toxaphene exposure are elevated in some individuals and are a cause for concern if individual exposures are elevated on a regular basis.*

- *The developing fetus and breast fed infant are likely to be more sensitive to the effects of OCs than adults and are the age group at greatest risk in the Arctic. Fetus/infant intakes of dioxins and furans, PCBs, toxaphene and HCB through human cord blood/milk are of primary concern even though the toxic effects that might occur are uncertain. In consideration of this uncertainty, the extensive knowledge of the benefits of breast-feeding are a strong rationale for Inuit women to continue to breast feed unless told otherwise by their health care provider.*
- *Human milk data comparisons clearly demonstrate that concentrations of OCs in humans are associated with consumption of traditional food (in particular fish and marine mammals). The concentrations of OCs in Inuit mothers' milk are 2–10 times higher than the levels in mothers' milk of southern Canadians.*
- *Risk management decisions must continue to be developed in cooperation with communities to reduce exposures and to sustain traditional ways, especially the consumption of traditional food.*

4.4.5.2 Metals

Bioaccumulation can occur for metals such as lead, mercury and cadmium (WHO 1989a, 1990, 1992a). Metal levels in organisms show great variations, depending on contaminant source, food chain structure and trophic level. Metals tend to accumulate in organs, particularly liver and kidney (Chan *et al.* 1995a). As was noted for OCs, it is the combination of contaminant concentration and the extent of use of a particular food that determines the amount of human exposure. This is illustrated for mercury, lead and cadmium in Table 4.4.6. Ringed seal liver has the highest concentration of mercury (429.3 µg·100 g^{-1}) but only contributes 11.9 % of total intake while ringed seal meat has a much lower concentration (24.8 µg·100 g^{-1}) yet contributes greater than 40% of total mercury intake.

Lead

In general, current levels of lead in the Arctic do not pose a significant threat to health and based on the declining emissions of lead globally are not likely to pose a threat to the health of Northerners in the future. Local emissions of lead from current or future smelting operations may increase lead levels regionally because lead can be transported by air relatively long distances (Skerving 1988).

It is clear that average adult blood lead levels are slightly higher in Arctic peoples than in the south and that they are associated with increased consumption of marine mammals and fish and with smoking (section 4.3.2.2). Nevertheless, these levels are in the 10 µg·dL^{-1} range (Nunavik) or lower (NWT) and below a level of concern based on Canadian and WHO blood guidelines.

For children, who are most at risk from lead exposure during fetal development and early infant development (quantifiable IQ point reductions at 20 µg lead·dL^{-1} or more and reduced gestation periods at 5 µg·dL^{-1} or more) there is a necessity to monitor cord blood and infant blood in the eastern Arctic, including Nunavik, to ensure lead levels do not increase. Currently average cord blood levels have been reported as 5.2 µg·dL^{-1} .

Although in general, marine mammals and fish appear to be the major pathways for lead intake in the Arctic, it is not known which foods contribute the majority of lead on a community-by-community basis. However, lead shot contamination in waterfowl may contribute to human exposure and is under investigation (Dewailly *et al.* 1996c). Lead levels are not often determined in country food samples because they are often so low.

- *Current levels of lead in the Arctic do not pose a significant threat to health and, based on declining emissions of lead globally, are not likely to pose a threat to health.*
- *Many adult and infant blood lead levels are below Canadian and WHO guideline values, however, because of the toxic effects of lead on children, cord blood and*

TABLE 4.4.6

Baffin Inuit foods with highest heavy metal contributions in women's diets (% total intake) and corresponding contaminant concentrations (µg·100g wet wt.$^{-1}$)[1] (Chan *et al.* 1995)

	MERCURY		CADMIUM		LEAD		
Food	% total intake	Conc.	% total intake	Conc.	% total intake	Conc.	Contribution rating[2]
Ringed seal meat	40.7	24.8	71.9	51.1	23.5	11.4	High (45.4)
Narwhal muktuk	21.5	108.1	1.7	3.7	5.4	8.7	Low (9.5)
Arctic char flesh	2.7	15.0	1.4	10.6	18.4	29.6	Low (7.5)
Caribou meat	7.2	6.6	3.4	3.6	10.0	50.0	Low (6.9)
Ringed seal liver	11.9	429.3	5.0	213.6	0.2	3.3	Low (5.7)
Walrus meat	2.3	18.1	3.1	21.1	5.6	23.1	Low (3.7)
Kelp	0.4	10.6	5.8	173.0	3.8	50.7	Low (3.3)

[1] *Values are approximate means of food as consumed following various preparations*

[2] *High: >= 20% total intake; Medium: 11–20%; Low: <=10%.*
Rating is based on average (in parentheses) of all 3 contaminant contribution values

infant blood monitoring should continue to ensure local or regional lead levels are not increasing.

Cadmium

Cadmium exposure in the Arctic comes primarily from smoking and secondarily from consumption of organ meats of caribou and moose and marine mammals. For non-smokers, Inuit, Dene/Métis, and southern Canadian populations have similar blood cadmium levels of between 0.2 and 0.9 $\mu g \cdot L^{-1}$ (Table 4.3.12). Based on a conversion factor proposed by Louekari *et al.* (1992), non-smoker intakes in Nunavik Inuit would be in the range of 0.4–0.5 $\mu g \cdot kg\ bw^{-1} \cdot d^{-1}$ which is half of the WHO PTDI. Recent results for Inuit, Dene/Métis and non-Aboriginal mothers from the NWT (Table 4.3.12) have not yet been broken down by smoking history but suggest that there are few real differences between the three groups in terms of mean cadmium levels and ranges in levels.

A recent dietary intake study of eastern Arctic Inuit women indicated that 21% consumed amounts of cadmium in diet greater than the current WHO PTDI (Table 4.4.3). These exceedances are not likely to be far greater than the PTDI but indicate further monitoring is needed in the Baffin region.

For smokers, cadmium levels are 20 to 30 times higher than for non-smokers in the Nunavik region and southern Canada (Ontario) respectively (Table 4.3.12). The exposures are again similar north and south, however the blood levels suggest that exposure to cadmium through smoking leads to large exceedances of the TDI.

Cadmium levels in caribou kidney tend to be highest in herds in the Yukon and those between Great Slave Lake and Hudson Bay. It is also in these areas that caribou consumption is greatest. Risk assessments have been completed for various age ranges of caribou and endorsed by community leaders. More importantly, for the protection of health from the point of view of exposure, is a reduction in tobacco smoking which leads to exceedances of the TDI without any contribution of cadmium from traditional foods whatsoever.

- *Several studies have indicated that cadmium intakes by non-smokers in the Arctic, whether Inuit or Dene/Métis, are low and similar to intakes reported in southern Canada. One study has shown that some Inuit may exceed the TDI for cadmium.*
- *Smokers have 20 to 30 times higher mean blood levels of cadmium than non-smokers. These intake levels exceed the current WHO TDI value several-fold and are not related to consumption of traditional food.*

Mercury

Methylmercury is a highly toxic substance and poses a significant risk to health in the Arctic, especially in the northern and eastern regions. Larger percentages (45–75%) of Inuit (NWT and Québec) and Cree had levels above 20 $\mu g \cdot L^{-1}$ than did Dene, Inuvialuit (NWT)and Yukon First Nations in the central and western Arctic (3–25%) (Section 4.3.2.3). Extensive research by Wheatley and his coworkers indicate the extent and range of the problem (Wheatley and Paradis 1995, 1996a and b). Blood mercury levels range from 1–660 $\mu g \cdot L^{-1}$ in the 38,571 samples examined between 1970 and 1995. Many of these measurements among Eastern Arctic Inuit and Cree were above the 200 $\mu g \cdot L^{-1}$ level suggested by the WHO (1990) as a guideline (i.e. 5% of the population above 200 $\mu g \cdot L^{-1}$ blood or 50 $\mu g \cdot g^{-1}$ hair may be at risk of developing neurological paraesthesia). While neurological signs have been observed clinically in some highly exposed native peoples it has as yet been impossible to irrefutably diagnose methylmercury poisoning in this range of exposure levels. Recently, two studies have become available that indicate that some Arctic Aboriginal communities may have lower levels of mercury (Walker *et al.* in prep., Dewailly *et al.* 1994b).

The fetus may be five times more sensitive to the effects of mercury than the adult (Rice 1995). The WHO (1990) has proposed that there is a 5% risk of neonatal neurological damage associated with peak methylmercury exposures measured at 10–20 $\mu g \cdot g^{-1}$ in maternal hair. Using the conversion factors proposed by Clarkson (1992) and Phelps *et al.* (1980), these hair levels can roughly be converted to 30–70 $\mu g \cdot L^{-1}$ blood. While all mean values for Dene, Yukon First Nations and non-Aboriginals reported in Table 4.3.14 are below this range, 12 out of 15 mean values reported in the same table for Cree and for Inuit from Canada and Greenland are within this range.

A 1987–1988 food intake study of Inuit in the eastern Arctic (Table 4.4.3, section 4.4.5) confirms these findings. Receveur (1996) reports that 29% of the women in the study had daily intakes of mercury that exceeded the WHO TDI for adults. This assessment does not comment on the implications for the fetus. Stern (1993), Gilbert and Grant–Webster (1995) and Rice (1995) have derived intake levels for mothers for protection of the fetus ranging from 0.025 to 0.07 $\mu g \cdot kgbw^{-1} \cdot d^{-1}$. Based on these derived values a pregnant 60 kg woman should not regularly exceed a daily intake of 1.5 to 4.2 µg of mercury.

The findings for mercury are complex. It is clear that mercury intakes in some communities exceed WHO TDIs and it is also clear that some adults and infants are in the 5% risk range for adverse effects. It is not known if the abundance of selenium in the traditional diet counteracts the effects of mercury in northern peoples as it does in experimental animals. It is also not known precisely how much mercury is of natural origin and how much enters the Arctic from long-range transport. In light of the uncertainties there remains a compelling argument for international action to reduce global mercury emissions and to consider selective reductions of intake of the traditional foods contributing the most to mercury exposure. This selective reduction approach will be essential for reducing mercury exposure for the fetus, newborn, and adults while ensuring a highly nutritious, culturally necessary diet of country foods.

Dietary surveys are underway (or nearing completion) in many communities. When the survey results are coupled with contaminant analyses, they will guide decisions in the communities on what dietary items, if any, should be considered for reduced consumption. Continual hair monitoring is also highly desirable to determine whether or not mercury levels are declining.

- *A number of the adult blood mercury measurements reported from 1970 to 1992 are above the 5% risk range (200 mg·L*$^{-1}$*) proposed by the WHO for onset of neurological paraesthesia. Recently data has become available under the AES program which indicates that some Arctic Aboriginal communities may have lower levels of mercury.*
- *12 of 15 mean blood values reported for Cree and Inuit from Canada and Greenland are within the 5% risk range for neonatal neurological damage (30–70 mg·L*$^{-1}$*) proposed by the WHO.*
- *One food intake survey confirms that a large percent (29%) of Inuit women are exposed to mercury levels above the current WHO TDI.*
- *Continued hair monitoring and community-by-community dietary survey results will guide communities in their decisions on what, if any, selective food intake reductions are warranted.*

4.4.5.3 Radionuclides

As indicated in section 4.3.3 the concentration of various radionuclides in Arctic air, water or soil are generally about the same or lower than, levels found in the temperate zone. Due to concentration in the food chain and consumption of large amounts of traditional food, Aboriginal peoples are exposed to higher levels of some natural and anthropogenic radionuclides.

The carcinogenic effects of radiation have been well documented in studies of human populations exposed to high levels of radiation. The question remains as to whether these effects can be extrapolated to the low exposures encountered in the environment. The number of induced cancers at these levels would be very small and statistically indistinguishable from cancers produced by other causes. Radiation protection authorities prudently assume that the risk of cancer is linearly related to dose with no threshold. This assumption should be regarded as a worst case scenario in a situation where the true risk from environmental radiation could be as low as zero.

The dose assessment methodology for radiation exposures has evolved to the point where it is possible to carry out a summation over all the doses from various radionuclides and pathways and to combine the resulting total dose into one single risk factor. The dose received by a person who consumes a radioactive contaminant in an exposure medium (e.g., air, food, water) is a product of three factors:

TABLE 4.4.7

Total radiation dose calculated for residents of Old Crow, Yukon

Radionuclide	Main Pathway[1]	Radionuclide Concentration[2] (Bq·kg^{-1})	Intake[3] (kg·y^{-1})	DCF[4] (mSv·Bq^{-1})	Radiation Dose[5] (mSv·y^{-1})
NATURAL					
external gamma	direct				0.8680
lead-210	mammals	0.97	497	7.0E–04	0.3375
polonium-210	mammals	14.8	497	1.2E–03	8.8267
radium-226	mammals	0.63	497	2.8E–04	0.0877
radium-226	vegetables	0.601	73	2.8E–04	0.0123
thorium-228	fish	1.26	25	2.7E–04	0.0085
thorium-230	fish	2.0	25	3.5E–04	0.0175
thorium-232	mammals	0.91	497	1.8E–03	0.8141
uranium	fish	0.59	25	3.9E–05	0.0006
uranium	vegetables	6.17	73	3.8E–05	0.0171
radon-222	air	11.2[6]		2.0E–02	0.2240
Total Natural					**11.21**
ANTHROPOGENIC					
cesium-134	mammals	0.5	497	1.9E–05	0.0047
cesium-137	mammals	9.5	497	1.3E–05	0.0614
strontium-90	water	0.0146	700	2.2E–05	0.0002
Total Anthropogenic					**0.07**
TOTAL: Natural and Anthropogenic					**11.28**

[1] *The pathways listed contribute over 98% of the total radiation dose (Beak 1995)*

[2] *The concentrations of radionuclides in the food items/exposure media are averages based on the best available measurements (Beak 1995)*

[3] *Intakes are for a person who derives their entire food supply from country foods (Beak 1995)*

[4] *DCF = Dose Conversion Factor, or the dose in millisieverts from each becquerel of radioactivity, ICRP (1994)*

[5] *Radiation Dose derived by multiplying radionuclide concentration by intake by DCF*

[6] *Concentration is in Bq·m*$^{-3}$

i the concentration of the radionuclide in the exposure medium [Bq/kg]

ii the amount of that exposure medium taken in/consumed per year [kg]

iii the dose conversion factor [Sieverts/Becquerel] for the specific radionuclide

This process is illustrated for one particular community, Old Crow, Yukon, in Table 4.4.7. The total individual radiation dose is 11.28 $\mathrm{mSv \cdot y^{-1}}$ and 99% of this dose is from natural radionuclides (11.21 $\mathrm{mSv \cdot y^{-1}}$ for natural radionuclides vs. 0.07 $\mathrm{mSv \cdot y^{-1}}$ for anthropogenic radionuclides). Polonium-210 and lead-210 supply 82% of the total natural radionuclide dose through the consumption of red meat. The additional contribution from organ meats is not available at this time.

Table 4.4.8 summarizes the doses from the main groups of radionuclides for six Arctic communities for which at least some dietary information is available. In the first five communities the doses are derived for persons who depend entirely on traditional or country foods. In the sixth community, Yellowknife, NWT, the doses are for persons who depend on food primarily from the southern commercial market. The derived doses for Yellowknife would be typical for most urban communities in Canada. The doses in Table 4.4.8 vary considerably from one community to the next. The results from the five communities depending on traditional foods give an average dose of 6.85 $\mathrm{mSv \cdot y^{-1}}$ from natural radionuclides and 0.29 $\mathrm{mSv \cdot y^{-1}}$ from anthropogenic radionuclides. $^{210}\mathrm{Pb}$ and $^{210}\mathrm{Po}$ contribute most of the natural radionuclide dose; Cs-137 contributes most of the man-made radionuclide dose. The corresponding doses for Yellowknife are 0.95 $\mathrm{mSv \cdot y^{-1}}$ for natural radionuclides and 0.05 $\mathrm{mSv \cdot y^{-1}}$ for anthropogenic radionuclides.

Although one would need to include more communities to characterize the entire region, the above doses can be used to derive an upper limit on the health impact of radionuclides in the arctic environment. Assuming an Aboriginal population of 70 000 dependent on country food and the International Commission on Radiological Protection (ICRP) fatal cancer risk of 5×10^{-5} per mSv, then

> Natural radionuclides:
> $6.85\ \mathrm{mSv \cdot y^{-1}} \times 5 \times 10^{-5}\ \mathrm{mSv^{-1}} \times 70\ 000 = 24$ fatal cancers per year
>
> Anthropogenic radionuclides:
> $0.29\ \mathrm{mSv \cdot y^{-1}} \times 5 \times 10^{-5}\ \mathrm{mSv^{-1}} \times 70\ 000 = 1$ fatal cancer per year

The corresponding radiation-induced cancer risk for an urban population of 70 000 receiving the same doses (natural plus man-made) as those calculated for Yellowknife residents would be:

> $1.00\ \mathrm{mSv \cdot y^{-1}} \times 5 \times 10^{-5}\ \mathrm{mSv^{-1}} \times 70\ 000 = 3.5$ fatal cancers per year.

The total number of fatal cancers from all causes in a population of 70 000 would be about 14 000. Assuming a 70-year lifespan, this would amount to about 200 cases per year. This would indicate that Aboriginal Peoples of the arctic who exclusively consume traditional foods would have a 12.5% higher cancer rate (25/200) due to environmental radiation while those people who consume a southern market diet would have a 1.8% higher cancer rate (3.5/200) due to environmental radiation. Available information indicates that Inuit cancer rates in the NWT, for all cancers except lung cancer, are significantly lower than the Canadian average (Gaudette 1996). The higher lung cancer rates in NWT Inuit are believed to be due to the much higher smoking rates among Inuit.

It must be emphasized that the above health impacts should be regarded as upper limits only. The calculations assume that the cancer risks derived for high levels of radiation apply equally well to low level radiation. They also assume that the entire northern Aboriginal population derive their entire food supply from traditional sources. It is also important to recognize that this evaluation only

TABLE 4.4.8

Radiation doses for six arctic communities[1] ($\mathrm{mSv \cdot y^{-1}}$)

Radionuclide	Broughton Island	Baker Lake	Makkovik	Rae-Edzo	Old Crow	Yellowknife
NATURAL						
external gamma	0.36	0.55	0.58	0.63	0.87	0.71
Pb/Po-210	1.99	10.10	3.20	1.99	8.85	0.00
Ra-226	0.02	0.11	0.04	0.08	0.11	0.00
other U/Th	0.25	1.03	0.34	1.06	0.95	0.01
Radon	0.22	0.22	0.22	0.22	0.22	0.22
Total Natural	**2.84**	**12.01**	**4.39**	**3.99**	**11.00**	**0.95**
Average for five communites with traditional diets = 6.85						
ANTHROPOGENIC						
Cs-137/134	0.08	0.55	0.72	0.25	0.07	0.05
Sr-90	0.0002	0.0002	0.0002	0.0002	0.0002	0.0002
Total Anthropogenic	**0.08**	**0.55**	**0.72**	**0.25**	**0.07**	**0.05**
Average for five communites with traditional diets = 0.29						

[1] *See footnotes to Table 4.4.7 for dose derivation. Pathway, local radioculide concentration and intake, are based on Beak (1995).*

considers the effects of radiation on cancer rates. Other possible effects of low level radiation have not been considered here because they have not been clearly documented in the literature.

- *Consumers of traditional foods are exposed to an approximately seven-fold higher radiation dose than non-consumers of traditional foods. More than 95% of this increased radiation dose is due to the bioaccumulation of natural radionuclides in the food chain.*
- *This increased radiation dose gives consumers of traditional foods a cancer risk that is approximately 10% higher than that compared with consumers of a southern diet. This increased risk is in fact not seen in NWT Inuit cancer statistics where Inuit have a significantly lower rate of all cancers, with the exception of lung cancer, than the Canadian population.*

4.4.6 Risk Determination/ Communication Case Studies

Several recent risk determination activities in the Arctic illustrate the application of the major steps of this process and underscore the complexities involved. The first four case studies exemplify the evolution under the AES from 1990 to 1996 of the evaluation/communication process pertaining to contaminants in traditional foods. There has been an increasing involvement of the Aboriginal partners in both the design of the research projects and final dietary advice to Northerners. The fifth case study, "PCB: Inuit of Nunavik," examines a research project on contaminants in human tissues and discusses the risk management decisions and risk communication process used.

These case studies also demonstrate the value placed on a high quality data base, cooperative strategies to understand the risks, and communication/ consultation initiatives to ensure clear understanding of risk reduction and health promotion. Further information on these and other case studies can be found in Usher *et al.* (1995) and Grondin *et al.* (1996).

4.4.6.1 Toxaphene in Lake Laberge Fish

Fish from Lake Laberge were tested for the presence of OC pesticides and PCBs in 1990 and 1991. The results were unexpected: high levels of toxaphene, DDT and PCBs were present. The concentration of PCBs, DDT, and toxaphene were 4 to 10 times greater in lake trout muscle and in burbot liver from Lake Laberge than in the same species from other lakes in the Yukon and the NWT. Health Canada reviewed the results and recommended maximum consumption limits for burbot (ling cod) livers and lake trout flesh from this lake. After consultation among all concerned groups and agencies, including First Nations, the medical Officer of Health for the Yukon issued a health advisory recommending that people limit consumption of lake trout to two meals per month and refrain from eating burbot liver. Contaminant concentrations and risk assessment conclusions are summarized in table 4.4.9. In this case study the risk management decision was simply the announcement of the risk assessment results.

As a result of the findings, several risk management actions were undertaken. The territorial government responded by buying out the commercial fishery on Lake Laberge and moving the First Nations Fisheries from Lake Laberge to a nearby lake to allow the overfished lake trout population an opportunity to recover. Signs stating the consumption limitations were posted in English, French, and Southern Tutchone at strategic locations around Lake Laberge. The Council of Yukon Indians, fearing widespread contamination, called for a territory-wide investigation. A spokesperson for the Council suggested that people refrain from eating fish caught anywhere in the Yukon until the extent of the contamination was determined.

In response to growing concern about contaminants in the aquatic environment, several sampling programs were initiated through the AES. In addition, the Yukon Contaminants Committee (YCC) was created in 1992 as a successor to the Lake Laberge Working Group.

This was the first hazard assessment from Health Canada recommending that Yukoners limit their consumption of fish from freshwater lakes as a result of elevated contaminants. The YCC, with representatives from federal and territorial governments, First Nations, public interest groups and the Yukon College, coordinated Yukon-based contaminant studies. They also released all contaminants data generated under the AES.

Communications of contaminant data soon became one of the most important and difficult responsibili-

TABLE 4.4.9

Toxaphene in Lake Laberge fish (1990–1991)

Species/Tissue	Toxaphene Concentration (range $ng \cdot g^{-1}$)	Risk Assessment Recommendations	Risk Management Recommendations
Trout Flesh	280–1400	Limit consumption to less than 130 $g \cdot week^{-1}$ or approximately two meals per month	Limit consumption to two meals per month
Burbot Liver	NA[1]–8500	Refrain from eating burbot liver	Refrain from eating burbot liver

[1] *NA = not available*

ties of the YCC. Based on the experiences gained through the first advisories on Lake Laberge a protocol for the release of future data (including health advisories) was established. Some of the basic concepts included involvement of the community prior to and during sampling, full accessibility of all data to the public, education and communications programs, timely turn around of results and the submission of data to Health Canada for assessment.

It was also the consensus that individual First Nations should be directly involved in the development of any future health advisories issued in the Yukon. Prior to any recommendation, all factors including the cultural and nutritional benefits should be considered. It is up to each individual community to make an informed choice on the consumption of traditional foods.

- *Based on high levels of PCBs, DDT, and toxaphene in lake trout muscle and in burbot liver from Lake Laberge, a health advisory was released recommending that people limit their consumption of lake trout to two meals per month and refrain from eating burbot liver. The Council for Yukon Indians then suggested that people refrain from eating fish caught anywhere in the Yukon until the extent of the contamination was determined. Based on the experience with this advisory, a protocol for the release of future data (including health advisories) was established.*

4.4.6.2 Toxaphene in Slave River Burbot

In the fall of 1992, a Health Canada hazard assessment concluded that people eating burbot liver from the Slave River, near Fort Smith, NWT, should limit their consumption to less than 13.4 g of liver per day or 93.8 g per week because of elevated toxaphene levels. The NWT Policy Advisory and Technical Committee reviewed the results and developed a plan to inform people living in Fort Smith about this finding as quickly as possible. The ensuing community meeting included presentations by the regional medical Health Officer, a representative from GNWT Renewable Resources, DIAND, the Dene Nation and the Métis Nation. An advisory was issued in which residents were warned not to eat more than one burbot liver per person per week (Usher *et al.* 1995). Key contaminant concentrations and risk assessment/management conclusions are summarized in table 4.4.10. In this case study the risk management decision involved the conversion of the consumption restriction advice from "grams per week" to the more easily understood quantity "livers per week."

Since this information was communicated, anecdotal reports from this suggest that some people have stopped eating any burbot liver. From this advisory, the importance of considering consumption advice in the context of the seasonal availability of the species was clear. In some areas, burbot are only available for a few months of each year, yet the advisory was communicated as though they were available all year. Conveying the information as a recommendation not to eat more than fifty-two livers per year would perhaps have been more useful to people from this community.

- *An advisory was issued in the Fort Smith/Slave River area recommending that residents not eat more that one burbot liver per person per week from Slave River. Reports from the community suggest that some people stopped eating any burbot liver. Based on the seasonal availability of burbot, it may have been more useful to the community to have the advisory expressed as an annual consumption limit (maximum of 52 livers per year).*

4.4.6.3 Cadmium in Caribou (NWT and Yukon)

A study undertaken by the Canadian Wildlife Service examined caribou in the Beverly, Southamptom and Porcupine herds, and muskoxen herds on Banks and Victoria Islands for cadmium levels. Based on the findings and assumptions about the average weekly intake of cadmium from smoking and commercial food, the researchers concluded that the consumption of a single meal of caribou kidney would lead to an exceedance of the tolerable weekly intake for cadmium. Conclusions were also drawn for caribou liver and muskox kidney. This information was made public prior to a review by Health Canada, the Yukon Territorial Government Health Department, or the GNWT Health Department.

In the Yukon, the response to the information release was a consultation with potentially affected communities about the cadmium results. Communities were presented with the benefits of eating caribou, and the information about the amount of cadmium present in caribou meat and organs. For example, in the evaluation of cadmium in Finalyson caribou, a 60 kg person could eat up to 28 kidneys a year of a caribou less than 2 years old, 8 kidneys per year from

TABLE 4.4.10

Toxaphene in Slave River burbot (1989–1991)

Tissue	Toxaphene Concentration (range $ng \cdot g^{-1}$)	Risk Assessment Recommendations	Risk Management Recommendations
Burbot Liver	137–1887	Limit consumption to less than 13.4 $g \cdot day^{-1}$ or 93.8 $g \cdot week^{-1}$	Do not eat more than one burbot $liver \cdot person^{-1} \cdot week^{-1}$

caribou aged between 3 and 6 years, and 4 kidneys from animals between 7 and 15 years of age. Although an advisory was not issued, the full information set was posted for anybody interested in more information. It must be remembered that caribou levels vary among caribou herds and specific consumption advice will therefore also vary.

In the Northwest Territories, data collection of contaminant levels in caribou herds was already underway by GNWT Department of Renewable Resources as part of a territory-wide survey of caribou herds. The information from the Canadian Wildlife Service was combined with the data collected to that date and was published in the monthly newsletter, *Epi-North*.

When the final results of the GNWT data and of the Health Canada risk assessment of the data were available, results were communicated in a three-page summary (GNWT 1995). Among other information, the summary stated: "Although caribou kidneys and livers contain moderate levels of cadmium...people can still eat up to 50 caribou kidneys each year throughout their lifetime without any health risk." This recommendation was based on Health Canada's hazard assessment which made recommendations for caribou kidney consumption based on the age of the animal and the weight of the consumer. Based on these age and weight categories, the range in maximum allowable kidneys consumable per year was between 50 and 182. The more conservative number of kidneys (50) was communicated in the summary, both for consistency of information between herds, and also to avoid a possible misunderstanding that the recommendation was for people to increase their annual intake of caribou kidneys. The summary was distributed by the NWT Technical Committee to regional health agencies, health centres, hunters and trappers associations and committees, regional Aboriginal organizations, Renewable Resource officers, and federal agencies. Key contaminant concentrations and risk assessment/management conclusions are summarized in table 4.4.11. In this case study the risk management decision involved the conversion of the consumption restriction advice from grams per week to the more easily understood livers or kidneys per year.

- *Contaminant levels were measured in caribou and muskoxen herds in the Yukon and the NWT. An advisory was not issued in the Yukon although communities were consulted about the findings and the information was posted for people wanting further information. In the NWT, the information from the initial study was combined with other findings and released in a short summary document that people could consume up to 50 caribou kidneys a year.*

4.4.6.4 Health Risk Assessment of Organochlorines in Marine Mammals

In March 1993, at the request of DIAND (Ottawa), Health Canada commenced a health risk assessment for fish and marine mammal data collected and analysed by the federal Department of Fisheries and Oceans (DFO) under the AES–NCP, as well as for DFO data collected prior to the AES–NCP. These data were originally collected for studies of spatial and temporal trends of contaminant levels in wildlife (the results are summarized in Chapter 3 of this report). However, the results from assessments in Yukon fish (see Lake Laberge case study), indicating that toxaphene was a potential human health concern in the arctic food chain, prompted a retrospective analysis of other NCP data. Following the *Protocol for Contaminants Health and Harvest Information Release* (Figure 4.4.2) developed under the AES, copies of the raw data and all related correspondence (e.g. letters requesting that the assessment be conducted, letters requesting status reports) were also forwarded to Northern Aboriginal Peoples' organizations, the GNWT and the two territorial contaminants committees.

In May 1995, as the Health Canada assessment was originally scheduled to be nearing completion, DIAND met with HC representatives to discuss the anticipated assessment results and to ensure that the most up-to-date data were being included in the

TABLE 4.4.11

Cadmium in liver and kidney of caribou in the Yukon and the NWT (1992–1994)

	Caribou Tissue	Cadmium Concentration (range $\mu g \cdot g^{-1}$)	Risk Assessment Recommendations ($g \cdot week^{-1}$)	Risk Management Recommendations[2]
Yukon[1a]	Kidney	5.4–79.6	7–42[3]	Although no advisory was issued the full information was posted. It was indicated that 4 to 28 kidneys or 2.5 to 6 livers could be safely consumed each year.
	Liver	0.78–19.69	70–175[3]	
NWT[1b]	Kidney	0.7–18.98	75–265[3]	Can safely consume up to 50 kidneys per year
	Liver	0.09–1.38	400–970[3]	No limitation given

[1] Based on selected assessments ([1a] – Mar 19/93, [1b] – Apr 26/95)

[2] Muscle was also analysed for cadmium. As only low levels of cadmium were found no limitation on consumption was required

[3] Older animals have higher concentrations of cadmium and the more restrictive recommendations apply to these animals. Cadmium levels vary among different caribou herds and will result in different consumption advice.

assessment. The outcome of these discussions, indicating that recommended maximum consumption levels for these tissues were likely to be quite low, were then communicated to the NWT Technical Committee on Arctic Contaminants (TCAC) during a teleconference held on June 29, 1995. Although results at that time were incomplete, and no recommendations had been made by HC, DIAND wished to allow the NWT Committee lead time to prepare in the event that Health Canada's assessment resulted in recommended maximum consumption levels of marine mammal tissues which could be of concern to Inuit consumers.

In November 1995, the NWT TCAC met in Yellowknife to develop the principles and a process for communicating health advisories. Although HC conducted the assessment, the GNWT health authorities are responsible for delivering the information and the health advice which follows from that information. DIAND Ottawa staff participated as guests in this meeting, due to their specific interest and involvement in the Health Canada assessment of organochlorines in fish and marine mammals, the preliminary results of which were presented (by HC) at the meeting.

Due to the concentrations of toxaphene and chlordane in beluga and ringed seal blubber, the HC assessment recommended maximum weekly intake (RMWI) of these tissues which were likely to be lower than quantities actually consumed by many Inuit. This was based on thousand-fold safety factors employed to address factors such as uncertainties in the available toxicological information and extrapolation of animal data to humans. For the species of most concern due to their contaminant levels (ringed seal and beluga), Table 4.4.12 provides the ranges of concentrations for chlordane and toxaphene, the risk assessment recommendations and the final risk management recommendations. Other contaminants such as DDT had much less restrictive RMWIs (2,415–31,381 $g \cdot week^{-1}$).

Notwithstanding the uncertainties, it was clear that at face value these low RMWIs warranted concern and a closer examination. Therefore, based on these results, the Inuit Tapirisat of Canada (ITC) chose to take immediate action to inform their regional representatives of the assessment's preliminary results.

Together with DIAND Ottawa, ITC held, on December 8, 1995, a meeting of 'Experts' with representatives from all Inuit regions across Canada (including Québec and Labrador who would have a direct interest in the issue), Health Canada, CINE, Fisheries and Oceans Canada, Santé Québec Community Health Department and GNWT Health and Social Services. The final assessment results were presented by HC at this time. The HC results provided, based on toxicological information (i.e. with no consideration of benefits) and the limited available dietary information , recommended maximum weekly intake (RMWI) figures for fish and marine mammal tissues. The assessment results for fish indicated no need for concern. However, based on the organochlorine pesticides chlordane and toxaphene, it was felt that there would be people eating more than the HC recommended maximum for beluga and ringed seal blubber (i.e. the RMWI figures were thought to represent quantities that would be lower than the amounts actually consumed by many Inuit. The following three key factors were discussed:

i) the large safety factors and related uncertainties of the toxicological information, including the long-term subtle nature of potential effects, and unknown effects of these chemicals in combination
ii) the certain and considerable benefits of continued consumption of the tissues in question, and
iii) the serious shorter-term, consequences which would result from restriction of consumption of these tissues.

The meeting participants agreed that, based on information available at that time, a recommendation to change diets which include beluga and ringed seal blubber would be premature and unwise.

At the December 8 meeting, it was agreed that the Inuit leadership would make a decision on initial release of information relating to the assessment, while GNWT would expedite the process which had been designed at the November 1995 meeting in Yellowknife. As a result, Rosemarie Kuptana, President of the ITC, made a public announcement through the northern media stating that they had received assessment results that indicated elevated

TABLE 4.4.12

OCs in marine mammal blubber from the NWT (1992–1994)

Species/ Tissue	Organochlorine Contaminants Concentrations Chlordane	Toxaphene	Risk Assessment Recommendations	Risk Management Recommendations
	(range, $\mu g \cdot g^{-1}$)		(RMWI $g \cdot week^{-1}$)[1]	
Ringed Seal Blubber	322–1291	175–601	28–480	Based on the uncertainty of the toxicological information for these chemicals and the dietary benefits of these foods, it was decided not to recommend dietary changes at this time.
Beluga Blubber	119–3311	319–11 283	8–375	

[1] *The most restrictive consumption advice, recommended maximum weekly intake (RMWI) for the above samples was due to chlordane contamination. Other contaminants had much less restrictive RMWIs, such as PCBs (70–994 $g \cdot week^{-1}$) or DDT (2 415–31 381 $g \cdot week^{-1}$).*

levels of contaminants in beluga and ringed seal blubber, but the information on risks, weighed against the known benefits of this diet, were such that people were advised not to alter their diets. DIAND notified provincial health officials in Québec and Newfoundland/Labrador of these events, also forwarding the assessment results. However, the only direct contact with Inuit in these two provinces was through ITC. At the same time, GNWT Health and Social Services developed an assessment information sheet for distribution, through the NWT TCAC, to representatives at the local level (federal, aboriginal and territorial). This was to be complemented by a series of contaminants and nutrition fact sheets and videos previously prepared and distributed to communities by GNWT. In addition, it was suggested that GNWT host a workshop specific to the issue and focused on key community communicators (e.g. health representatives, hunters' associations). Such a meeting has, to-date, not yet been arranged. However, a workshop to exchange information on contaminants issues and communication was hosted in March 1996 by the NWT-TCAC. The workshop was held in Cambridge Bay with members of Pauktuutit, the national Inuit women's organization, in recognition of the important roles and concerns of women regarding food, breastfeeding and family health. Attending this workshop were women who had been elected as Pauktuutit representatives by their communities in Nunavut, Nunvik, Labrador and the Innuvialuit settlement region.

Throughout the process, all participating agencies were informed of one another's activities, and these communications ensured that common and consistent messages were conveyed by all parties concerned. The process followed for this major assessment, which encompassed hundreds of tissue samples and dozens of communities, was important in opening the dialogue with those at greatest risk of effects from contaminant exposure associated with consumption of traditionally harvested foods. It resulted in serious discussion of the consequences of altering diets which rely on traditional foods, and the significance of the benefits of those foods. It initiated discussion of potential effects, and clearly demonstrated our need for more information to allow a better evaluation of risks. In doing so, opportunities for better information transfer and streamlining of action were identified, and lessons were learned about the types and formats of information which are most useful to different audiences. The assessment demonstrated that the NCP's partnership structure and evolving communication networks allow for an open and direct discussion of issues as they arise, and facilitate active and immediate collaboration of aboriginal organizations, governments and the scientific community in assembling, evaluating, responding to and communicating the available information. However, it also clearly points out the compelling need to augment existing knowledge about the potential consequences and human health implications of current levels of organochlorine contaminants in marine mammal tissues. This issue continues to be important to the NCP, and concern about improving the situation and our understanding of it remains high.

- *Based on insufficient information about the risks and the significant known benefits of traditional food, people were not advised to significantly alter their current diets.*

4.4.6.5 PCBs: Inuit of Nunavik

In 1986, the Province of Québec conducted a survey of PCBs in milk of 600 women from various parts of Québec and found very high levels of organochlorines in the milk of Nunavik women. Based on the fact that there are not enough data to formulate definitive advice on potential risks, and the knowledge that there are clearly established benefits of breast-feeding (in terms of health promotion and economic costs), the Nunavik and other health authorities decided that further information was required to determine if a health advisory was needed.

A surveillance program was set up in 1989 to monitor PCB levels in human milk. The objectives of the program were to measure the biological exposure of Inuit women to organochlorines, to identify epidemiological data relevant to the human milk levels, to assess the health risks involved, and to make recommendations regarding breast-feeding.

In order to keep the northern population informed about the progress of the study and to ensure that people in Nunavik did not react to information about PCB contamination without adequate supporting information, a PCB Resource Committee was put together (renamed the "Food, Contaminants and Health Committee" in 1994). When initiated, the Committee consisted of the project field coordinator, two midwives from Povungnituk, an auxiliary nurse from Kuujjuaq, an information officer from Kuujjuaq Regional Community Health and Social Services (KRCHSS), a community representative, and a member of the Makivik Corporation. Communication activities continued throughout the monitoring program and included the preparation and publication of information, participation in a number of radio interviews or discussion forums, the preparation of a video, and discussion of the issue of food chain contamination and benefits associated with country food and breast-feeding with the various people concerned with health in Nunavik.

To date, the main recommendation communicated to the public has been to continue eating traditional diets of fish and sea mammals. Furthermore, nursing mothers were strongly urged to continue breast-feeding their babies (Grondin *et al.* 1996).

The research to complete a comprehensive risk assessment of the situation is ongoing. A risk management process has also been initiated with the support of regional partners in order to bring together the major stakeholders working on issues related to food chain contamination in Nunavik and Labrador.

- *Research required to complete a comprehensive risk assessment of PCBs in human milk is ongoing. To date, the public has been advised to continue eating traditional diets of fish and sea mammals and nursing mothers have been strongly urged to continue breastfeeding their babies.*

4.5 Conclusions

The two key objectives of this chapter were to assess the impact of exposure to current levels of environmental contaminants in the Canadian Arctic on human health and to identify the data gaps that need to be filled by future human health research.

4.5.1 Aboriginal Perspectives on Food and Health

1. Assessing the impact of contaminants on the health of people living in Aboriginal communities must be undertaken within the context of the social, cultural and spiritual dimensions of health recognized by Aboriginal peoples. "Traditional" or "country" food is an integral component of the concept of health in Aboriginal communities because it contributes significantly to spiritual and cultural identity. The harvesting, sharing, processing, and consumption of traditional food is an important part of cultural identity. Cultural identity is expressed in traditional values of respect and responsibility for the environment. A degraded environment is considered a threat to health.

4.5.2 Traditional Food Consumption in the Arctic

2. Country foods are an economic necessity for many Aboriginal Northerners. Estimates of the cost to purchase equivalent amounts of imported meats in local stores indicate that the annual replacement value of traditional food in the NWT is well over $10,000 per Aboriginal household per year.
3. Traditional diets are an important source of nutrition with many of the essential lipids, vitamins, protein, and minerals coming from the traditional portion of the diet. Imported foods contribute more to daily caloric intake due to their higher saturated fat and higher simple carbohydrate (e.g. sugar) content.
   - As well as being a concentrated source of energy, lipids consumed in traditional foods act as carriers of fat-soluble vitamins, and are a source of n-3 fatty acids not easily found in imported foods. A diet rich in omega-3 fatty acids, which are found at high levels in fish and marine mammal tissues, have been associated with a decreased incidence of thrombotic and ischaemic disease.
   - Thiamin, riboflavin and niacin intakes in the North are reasonably adequate due to the fact that traditional food (meat) is a major contributor of these vitamins. Fish and game contribute substantial amounts of vitamin B^{12} and pantothenic acid. Total intakes of these vitamins are likely higher than in the general Canadian population. However, some reports indicate that vitamin A, calcium, and vitamin C intakes may be below recommended levels.
   - In arctic communities, a significant portion of the mineral (especially selenium, iron, zinc) and protein requirements are fulfilled by traditional foods.
4. Limiting the supply of traditionally harvested fish and game would drastically reduce protein. Similarly, if consumption of traditional food resources, particularly fish and wildlife, were discontinued, the mineral nutrition of most arctic populations would be compromised to such an extent that nutritional deficiencies could occur.
5. Dramatic reductions in country food consumption in northern Native populations has been associated with decreased physical activity, obesity, dental carries, anaemia, lowered resistance to infection, diabetes, and would likely result in increased heart disease.
6. Estimates of country food consumption in northern communities show that traditional foods are widely consumed. The key food resources, depending on the region (coastal or inland), are marine mammals and large ungulates (caribou, and moose). Country food consumption varies seasonally and annually as well as regionally, according to income and access to urban centres, and as a factor of age and gender. Country food consumption in the communities studied ranged from 292 $kg \cdot yr^{-1}$ at Sanikiluaq, to 27 $kg \cdot yr^{-1}$ in the Wood Buffalo Park area.
7. The diet of Aboriginal Northerners consists of a mix of country food and imported food. A trend of increased imported food use has been noted in the North with increased availability and to some degree affordability of these foods. This is due to many factors associated primarily with industrialization, improved transport, and "modernization." This trend has been most marked in the younger segments of the population.

4.5.3 Environmental Contaminants

8. The levels of OCs such as DDE, PCBs, Mirex, dioxins and furans, and chlordane are two- to ten-fold higher in milk of Inuit women from Arctic Canada than non-Aboriginal women from southern Canada. Comparative values for toxaphene and HCH are not available.
9. The findings of higher concentrations of various OCs in Inuit mothers' milk is supported by studies of OCs in newborn cord blood. Concentrations of several OCs in Inuit newborn cord blood were two- to four-fold higher than those seen in Dene/Métis or non-Aboriginals from the NWT and the general population from southern Québec. There are also differences in the levels of some contaminants between Inuit groups in Arctic Canada and these may be due to amount and type of country food consumption, age of mothers, number of children, length of breast-feeding and number of children breast fed.
10. Few studies of the health effects of OCs on arctic populations have been undertaken. A preliminary study of Canadian Inuit associated PCB levels with impaired immune responses in infants/children. This Canadian Inuit study is supported by a Dutch study which found subtle clinical, immunological and neurobehavioural alterations in infants/children associated with dioxins and furans and dioxin-like PCB levels in human milk.
11. Lead (Pb) does not appear to be a contaminant of significant concern in Arctic Canada. Levels in children's blood are as low as those found currently in southern populations.
12. Among Inuit the influence of dietary and environmental sources of cadmium exposure appear to be minor compared to the contribution from smoking. Smokers have 20 to 30 times higher mean blood cadmium levels than non-smokers regardless of where they live in southern or Arctic Canada.
13. Methylmercury is a potent neurotoxin and human exposure in the arctic is almost exclusively through food consumption, especially fish and marine mammals. Blood mercury levels are highest in northern and eastern Inuit communities and there was no convincing downward trend in blood mercury levels between 1972 and 1987. Two recent surveys conducted under the AES program suggest that some Dene and Inuit groups may have lower blood mercury levels.
14. The amount of radionuclides from atmospheric fallout in the Arctic environment are about the same as or lower than levels found in the temperate environment. However, due to bioconcentration in the food chain, residents of the Arctic who consume large amounts of traditional foods may be exposed to higher amounts of anthropogenic and natural radionuclides. Today, the natural radionuclides, ^{210}Pb and ^{210}Po contribute most of this additional radiation dose in the Arctic. These levels of exposure are likely to have existed in the Arctic for thousands of years. Levels of radiocesium in humans — the anthropogenic radionuclide of greatest concern — have decreased from about 450 $Bq \cdot kg^{-1}$ in 1965 to roughly 10 $Bq \cdot kg^{-1}$ in 1990 with the cessation of above ground nuclear testing in the early 1960s.

4.5.4 Assessing and Managing Risk

15. In the eastern Arctic, a large number of Inuit women (48% and 75%) in two communities studied exceeded the Tolerable Daily Intake (TDI) for chlordane based on dietary survey estimates. A number of these women also exceeded the TDI for toxaphene, hexachlorobenzene, hexachlorocyclohexane, dieldrin, and PCBs. In any given day, a much smaller percentage of Dene/Métis (approximately 6%) exceeded the TDI for one or both of chlordane and toxaphene. Comparisons of human milk data clearly demonstrate that increasing concentrations of many OCs are associated with increasing consumption of traditional foods (mainly fish and marine mammals). Some individuals need to reduce their intake of the most contaminated food items to lower their exposure. These individuals should consume other traditional foods to ensure adequate nutritional, social, cultural, economic and spiritual benefits.
16. The developing fetus and the breast fed infant are likely to be more sensitive to the effects of OCs than adults and are the age groups at risk in the Arctic. Fetal/infant exposures to dioxins and furans, PCBs, toxaphene and HCB through human milk/cord blood are of primary concern though the toxic effects that might occur are uncertain. A proportion of Inuit newborns have levels of PCBs that exceed a level of concern. Balancing the uncertainty associated with the effects of the contaminants with the extensive knowledge of the benefits of breast-feeding there remains a strong rationale for Inuit women to continue to breast feed unless told otherwise by their health care provider.
17. Two recent surveys of eastern Arctic Inuit indicate that in any given day 21% to 37% of women consuming traditional foods exceed the WHO TDI for mercury and cadmium respectively. Other surveys have indicated that the only smokers exceed the TDI for cadmium. Extensive sampling of human tissues in the Arctic indicate that a significant proportion of Cree from Canada and Inuit from Canada and Greenland had mean blood mercury levels within the 5% risk range (30–70 $\mu g \cdot L^{-1}$) proposed by the WHO for neonatal

neurological damage. Continued hair monitoring for mercury and community by community dietary survey results will guide communities in their decisions on what, if any, selective food intake reductions are warranted.

18. Though levels of environmental radiation in Arctic Canada are similar to more temperate regions, consumers of traditional foods in the Arctic are exposed to a three- to twelve-fold higher radiation dose than non-consumers of traditional foods. More than 95% of this increased radiation dose is due to concentration of natural radionuclides in the food chain. This increased radiation dose gives an approximately 10% increased risk of cancer in the population consuming large amounts of traditional foods. It is uncertain, however, whether the risk of cancer from high dose exposure of nuclear bomb survivors can be extrapolated to low dose environmental exposures (primarily natural sources). In fact, the rate for all cancers, with the exception of lung cancer, is lower in NWT Inuit than in the general Canadian population.
19. Risk determination for country foods considers the potential risks from exposure to contaminants and the sociocultural, nutritional, economic, and spiritual benefits associated with country foods. Regardless of the decision taken, the health risks associated with exposure to contaminants remain though they may be outweighed by the benefits of continued consumption. This poses a confusing public, moral and political dilemma.
20. The transfer of accurate and complete information via good communication may help limit the social and cultural effects resulting from the presence of contaminants in traditional food. If it does not occur, people will be forced to draw their own conclusions and will act based on their perceptions of the situation. Communication should be a two-way flow of information with an opportunity for feedback. It should also occur at the onset of the research/monitoring studies and should be an ongoing process.
21. Risk management decisions for communities must continue to be developed with communities to reduce exposure and to sustain traditional ways, especially the consumption of traditional food.
22. All of the contaminants discussed in this document can affect health adversely and will exert their effects as exposures increase. Exposures to these contaminants must decline and the significant benefits of consuming traditional food must be sustained. Eliminating traditional food from the diet is unrealistic and unhealthy. In the short term, reduced intake of some organs/tissues of some species may be needed to reduce contaminant intakes, provided they are replaced by increased consumption of other highly nutritious traditional food available locally. In the long term, there is the need for committed international effort to curb the global manufacture and release and to increase the rate of destruction of these contaminants so they no longer enter the Arctic bioregion at their current rate. This global action will bring down contaminant levels in the Arctic and reduce known and as yet unknown, risks associated with exposure. Action to prevent pollution of the Arctic cannot await definitive and extensive proof of harm to human populations or to the Arctic ecosystem.

4.5.5 Knowledge Gaps

This assessment has revealed significant exposure to and possible effects of contamination in arctic communities. It has also revealed a number of significant data gaps. Some data gaps, such as those related to monitoring contaminant levels in human blood/milk, are being addressed by ongoing studies and will take several years to complete. Others, such as those dealing with neurobehavioral, immunological and reproductive impacts, have undergone investigation at the pilot study stage and need to be funded as full studies. Finally, studies have not yet been initiated on the toxic effects of long term exposure to components/metabolites of toxaphene and chlordane in the environment. These studies are essential to understand the human health impacts of contaminants in the Arctic and will enable health agencies and Aboriginal leaders to provide better advice to communities on the risks posed by contaminants.

The majority of the research needs identified below pertain directly to the Arctic. However, some of the animal studies (see points 3 and 4) are not only of direct benefit in the Arctic but also serve a more general need to understand the effects of contaminants on the fetus (placental transfer) and the newborn (human milk).

4.5.5.1 Toxicology

1. Little information is available on the human health effects of long-term exposure to toxaphene components found in fish and marine mammals (not technical/commercial toxaphene). More toxicological information is required to identify the levels of exposure at which adverse health effects will be seen. Animal feeding studies containing toxaphene components such as T_2, T_{12} and others are needed to fill this data gap.
2. The effects on human health of long term exposure to chlordane components/metabolites found in fish and marine mammals (not technical/commercial chlordane) are not well known. Animal feeding studies using chlordane and its components/metabolites are required.

3. Environmental contaminants in traditional diets occur as complex mixtures. The additive/antagonistic/synergistic effects are not known. Animal studies must explore the interactive effects of contaminants in order to better understand the potential human health effects of these contaminant *mixtures* in the Arctic.
4. Due to the rapid development of all body systems, the fetus is sensitive to many contaminants that cross the placenta. Animal feeding studies are needed to further investigate the effects of maternal contaminant mixtures on the fetus with emphasis on neuropsychological development, and immune system or reproductive impairment.
5. The newborn, whose body systems are continuing to rapidly develop, is exposed at higher levels to a mixture of contaminants present in breast milk. Animal feeding studies are required to further investigate the effects of contaminants on the neuropsychological development and immune system of newborns.

4.5.5.2 Epidemiology

6. Studies conducted to date indicate that the fetus and neonate are likely at greatest risk from exposure to contaminants. Further study is required to evaluate the neurobehavioral, immunological and reproductive organ status of newborns in the Arctic. Pilot studies are complete.
7. High intakes of cadmium may lead to kidney disease. Smoking appears to be the major contributor of cadmium for most people living in the Arctic and elsewhere, though consumers of large amounts of traditional foods such as liver and kidney from caribou and moose are also exposed to higher levels of cadmium. Evaluations of kidney function and disease are required in arctic communities, notably among consumers of high levels of cadmium-containing foods as well as smokers and non-smokers.
8. In certain areas in the Arctic, the level of human exposure to mercury is at a level which exceeds the TDI and some adults and infants are in the 5% risk range for adverse effects. Because the fetus may be five times more sensitive to the effects of mercury than the adult, the evaluation of fetal effects of methylmercury is a priority. Research is currently underway around the world and must also be done in the Arctic, an area of high exposure.

4.5.5.3 Exposure

9. Accurate evaluations of exposure are essential for risk assessment. Exposure to environmental contaminants varies over a lifetime and the few arctic dietary studies focus on one or a few age groups. Studies must be done looking at community contaminant exposure profiles for various age groups: 0–1, 1–12, 13–18, 19–29, 30–49, 50–70.
10. Maternal/cord blood monitoring data is needed across the Canadian Arctic to allow an accurate assessment of human fetal exposures. This monitoring is underway in the NWT and Northern Québec and must be initiated in the Yukon and Labrador.
11. Human milk monitoring must be completed for all high-exposure populations in the Canadian Arctic to identify potential at-risk groups. Preliminary sampling in Northern Québec is complete.
12. Consumers of traditional foods are exposed to higher levels of radiation mostly due to the ingestion of the natural radionuclide ^{210}Po (polonium-210). The recently increased ^{210}Po absorption factor used by ICRP (1994) is based on one study in the United Kingdom. This needs validation for northern diets.

4.5.5.4 Food Surveys

13. Accurate food consumption information assists the estimation of contaminant and nutrient intakes. This information is essential in a risk-benefit evaluation of country foods for specific communities. Although Western Arctic communities have participated in food consumption surveys, there remains a need for these surveys in some eastern Arctic and Labrador Inuit communities.
14. Traditional foods in the diet contribute to both significant nutritional benefits and contaminant exposure. Recent information on contaminants may have caused significant changes in the diets of Northerners. These dietary changes need to be quantified in communities that have already participated in food consumption surveys.
15. There is little information about the contaminant and nutrient composition of "as consumed" foods in Arctic communities. Studies are required to assess the effects of cooking and processing of country foods on both nutrient content and contaminant levels. This information is essential to doing an accurate risk-benefit evaluation of country foods.

4.5.5.5 Risk Determination

16. A major difficulty in the risk determination process is the comparison of the risks from contaminant exposure with the benefits (nutritive, social, cultural, physical, economic) of traditional food consumption. A good model/approach for comparing risks with benefits must be developed to remove the subjective aspects of risk determination and allow a more rigorous approach to

weighing the risks and benefits of traditional food consumption.

17. While some information exists on the effect of contaminants in isolation, little is known about the effects of contaminants in the presence of common confounders such as poor nutrient intake and exposure of the fetus to alcohol or the products of maternal smoking. An important priority for research in this area is an evaluation of growth, disease, and resistance to infection in children related to the combined effects of these confounders and relatively high contaminant exposure.

4.5.5.6 Communication

18. Communication and management play a critical role in the success or failure of the risk determination process as they are the mechanisms through which the key information about diet and contaminants in Aboriginal communities is conveyed. It is important to determine whether or not the advice is reaching the community and whether this information was clear and usable so that subsequent communication/management strategies can draw upon past experiences.

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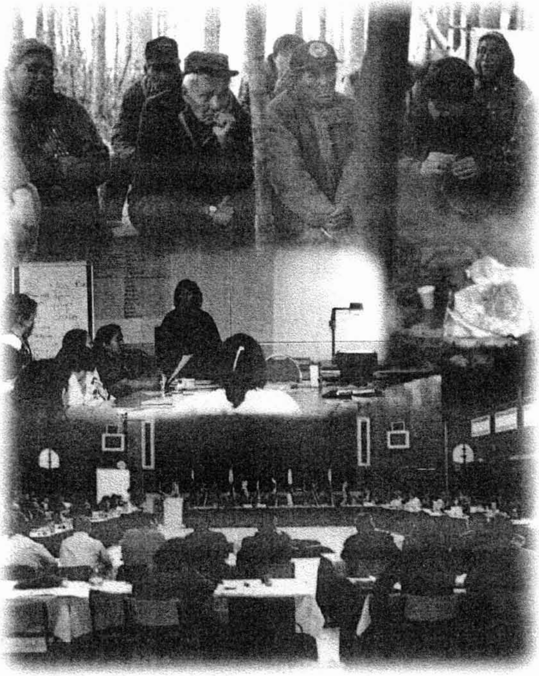
Chapter 5: From Research to Action – Using the Program Results



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CHAPTER 5 From Research to Action – Using the Program Results

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5.1 Understanding the Issue

5.1.1 Introduction

Findings from studies conducted under the Northern Contaminants Program (NCP) are presented through publication in the open literature (e.g. refereed scientific journals), and "gray" literature (e.g. government reports) and through presentations at scientific conferences. However, the research program of the NCP is also intended for practical application.

Northerners must have access to research results that are in a relevant and meaningful form, to make informed decisions regarding contaminants, traditional foods and lifestyle choices. Respecting, yet superseding the purely academic aspects of scientific research, projects approved under the NCP are intended to provide northerners with the information they need concerning contaminants in the Arctic ecosystem, in particular in traditional food species. To achieve this, the scientific research of the NCP must also respond to concerns and priorities at the community level, from the research questions and process through to presentation of results in an understandable form. This may occur on a project-specific basis, as well as on a program-wide basis.

A central challenge to communication about the NCP is the wide range of languages and cultures among the participants in the NCP. The scientific managers and researchers of the NCP generally speak only English or French, whereas the Aboriginal communities — who are the primary clients of the NCP — often communicate primarily in their own languages, with English being a second language. Many scientific terms related to contaminants are difficult to translate in a meaningful way into the Aboriginal languages. Translation from one language to another is more than an issue of terminology; there are fundamental differences in the world views that underlie these languages. All cultures develop a system for categorizing the things that make up the world and for understanding how those things fit together. The contemporary scientific world view has evolved over centuries as one defining aspect of western industrial cultures. Similarly, Aboriginal peoples have developed their own knowledge systems for understanding the world. In both cases, the languages of the people involved possess the vocabulary required to express their respective world views. Yet understandably, they do not necessarily have the vocabulary to express each others' world views.

Although the cross-cultural issues of communication pose many challenges to the NCP, the commitment to partnership between the scientific and Aboriginal communities has led to some innovative initiatives to overcome these challenges. Within the management structures of the NCP committees, the scientists and the Aboriginal partners who comprise the committees, have strived to make their respective world views understandable. This has required much effort in trying to match the information needs of the Aboriginal communities with the research commitments of the scientists and the NCP technical requirements for data.

The use of NCP results extends beyond the generation and presentation of research results. It involves the use of those results at the political level, both domestically and internationally, to effect change through regulation of the chemicals of concern; the goal being an ultimate decline in contaminant levels in arctic biota. Findings in the Arctic present clear evidence of the global nature of contamination from a number of pollutants, and demonstrate that transboundary effects can be exerted over great distances, thus requiring international cooperation on a wide scale. The characteristics of arctic food chains, particularly in the marine environment, exemplify the importance of bioaccumulation and biomagnification and the resultant threat posed by even minute levels of certain contaminants in the abiotic environment. Furthermore, the potential threat to human health, resulting from the biomagnification of environmental contaminants in the food chain, is given substance in the Arctic where Northern Peoples are exposed through their traditional diet. In combination, these facts give Canada a strong scientific case for arguing against the continued global release of the chemicals of concern.

In the short term, NCP results should provide people with an understanding of the issue of contaminants in the Arctic. This encompasses the information required to make decisions on consuming traditionally harvested foods, given the risks posed by contaminants in these foods versus the considerable benefits obtained through continued consumption of the traditional diet. In the long term, it is hoped that the NCP results will contribute to political actions that will ultimately realize significant reductions in the quantities of contaminants reaching the Arctic.

5.1.2 Accomplishments

The NCP has broken new ground in a number of different scientific, policy and program implementation areas. Research generated through the NCP represents leading edge science in the field of environmental contamination with persistent pollutants. NCP results constitute a large proportion of the global knowledge base on arctic contamination in all major ecosystem components. NCP research on global transport and pathways of contaminants demonstrates the occurrence of many chemicals far from any sources, and provides critical evidence for long-range movement of many key pollutants of concern, thus providing indisputable substantiation for the need to control chemicals based on their long-range transport potential. NCP data on contaminant levels in arctic biota, and on human exposures through the arctic food chain, illustrate the critical role of biomagnification, even for those substances that are barely detectable in the abiotic environment, thus providing compelling arguments for control of even minute releases of such substances. Finally, NCP data quantifies the nutritional and dietary importance of traditionally harvested foods to northerners, providing context for the concerns that contamination of the food chain might engender, but also heightening awareness of northerners' dependence on these foods, and therefore the critical and immediate need for action to reduce contaminant levels in those foods.

During the course of the NCP, significant action to address persistent and bioaccumulative contaminants has taken place on both the international and the domestic fronts. It can be argued that much of the international action that has occurred since 1989 has been driven by NCP results and NCP participants. Not only have the data generated by the NCP been the keystone of calls for international action on long-range transported, persistent, bioaccumulative and toxic substances, but Canada's leadership role at the United Nations Economic Commission for Europe (UN ECE), as well as the growing momentum for global action, have largely been precipitated and facilitated by this program. Nationally, NCP data have strengthened, accelerated and substantiated domestic policies, such as the federal government's Toxic Substances Management Policy, and other legislation on toxic substances, particularly those that are persistent and bioaccumulative.

From a policy and implementation perspective, the NCP has also been highly innovative. Through a continuously evolving process, the NCP is setting new standards for community participation in scientific programs. The partnerships forming the management structure of the NCP are unique and have proven highly productive and mutually beneficial. Through this structure, the NCP has been given the capacity to address Arctic ecosystem contamination using approaches that ensure scientific integrity and relevance, while responding to community concerns and priorities. For example, all individual proponents are required to report their results annually and must re-submit a proposal each year for approval to ensure that the project remains relevant to the overall program objectives and that the program itself remains flexible (to respond to emerging issues), scientifically defensible and robust. Community concerns and priorities can be related not only to the specific types of studies done, but also to how the studies are conducted and communicated, and to how the communities can initiate or take part in NCP research and communication activities, thus increasing their capacity to address contaminants issues.

The NCP as a whole is continuing to adapt its priorities, policies and overall approach as a result of the ongoing dialogue among northern communities, northern Aboriginal organizations, scientists and governments. This is demonstrated by the range of interests represented on the program management committees, the NCP commitment to community participation in research (expressed through policies such as the Researcher Guidelines for Planning Communication and Community Participation), and NCP support of capacity-building at all levels, from the Aboriginal partners and institutions (such as the Centre for Indigenous Nutrition and Environment (CINE)) to actual community project proposals.

This chapter describes how NCP results are used in order to further the short-and long-term objectives described above. Section 5.2 describes the structure of the AES-NCP, elaborates on the role of the Aboriginal organizations, and discusses community participation in the program. It also details activities and issues relevant to communication with northern communities, including discussions of risk and benefit. Section 5.3 covers the linkages of the NCP with other national initiatives and reviews some of the key concerns and issues that have been expressed by community members. Section 5.4 illustrates how research from the NCP is relevant to Canada's activities at the domestic and international levels.

5.2 Innovative Partnerships

The NCP was designed to address a specific Arctic human health issue: contamination of traditionally harvested foods with persistent, bioaccumulative, toxic substances. This situation has arisen largely due to contaminant movement on a global scale and is inextricably linked to contaminant dynamics that can only be understood when approached on a "whole ecosystem" basis. Technically, the issue encompasses a broad range of scientific areas ranging from (among others) the global movement of air and water currents to the behaviour at the molecular level of specific substances, and from feeding and reproductive strategies of a wide range of marine, freshwater and terrestrial species to the nutritional and cultural importance of traditional foods to northern Aboriginal peoples. On a political level, the issue of arctic food chain contamination is one that is multijurisdictional, being of concern to federal, territorial and Aboriginal governments; yet it is also one that can only be addressed through cooperation from the international community on a global scale. Finally, the needs and the concerns that must be addressed in relation to contamination of arctic food chains must yield information that meets internationally acceptable scientific standards, but they must also respond to the needs (for information, as well as capacity-building, participation etc.) expressed at the community level, and at the level of individual consumers of traditional foods.

A program that addresses such a scientifically and politically complex issue requires well-developed management, planning and implementation structures and strategies. The multidisciplinary nature of the NCP has allowed it to develop such structures and strategies, as described in the following section.

5.2.1 Program Infrastructure

Much of the strength of the NCP is derived from the partnership approach that forms the basis of its management process. This encompasses representatives who speak to the key areas of arctic contaminants research (based on an ecosystem approach), northern community concerns, needs and priorities, and the international and domestic agendas for control of toxic substances. The NCP is managed by DIAND on a partnership basis by a series of interrelated committees:

- The **AES Partners Committee** made up of representatives of five Aboriginal organizations (Inuit Tapirisat of Canada (ITC), Inuit Circumpolar Conference (ICC), Métis Nation-NWT, Council for Yukon First Nations (CYFN), Dene Nation and DIAND officials from the Yukon, the NWT and headquarters. This committee deals with overarching issues within all four programs of the AES, but it plays a special role in the NCP in relation to issues such as ethics and community involvement in research.
- The **Science Manager's Committee on Contaminants in Northern Ecosystems and Native Diets** is chaired by DIAND and is responsible for establishing NCP policy and research priorities and for allocation of funds. It is a multidisciplinary committee comprising managers from the federal Departments of Environment, Fisheries and Oceans, Health Canada and DIAND, representatives of the Yukon and Northwest Territories' governments, and delegates from the five Aboriginal organizations of the AES Partners Committee.
- The **Technical Committee on Contaminants in Northern Ecosystems and Native Diets,** chaired by DIAND, is responsible for annually reviewing and prioritizing project proposals and making recommendations to the Science Manager's Committee. Its membership is drawn, at the technical level, from the agencies described above, as well as from academic institutions.

There are also two regional committees:

- The **Yukon Contaminants Committee** develops and coordinates a comprehensive regional contaminants research program and establishes priorities for the Yukon. The committee, chaired by DIAND, develops a Yukon position for consideration by the Technical Committee. It evolved out of the AES and provides residents of the Yukon with a direct channel to the AES programs. All contaminants research projects conducted in the Yukon are either developed and managed, or reviewed and approved by the YCC. Communications on contaminants in the Yukon are mediated through the Committee, and one-on-one consultations, which occur on an "as required" or "as requested" basis, are an important part of this process. In addition, a number of fact sheets have been developed for distribution to local educators (including Yukon College), references have been collected and made available through Yukon College, and annual workshops have been conducted with community members and scientists to review studies that have taken place in the Yukon.
- The **NWT Policy Advisory and Technical Committees on Arctic Contaminants** were formed by the Executive of the NWT Legislative Assembly.

This occurred after detection of elevated levels of PCBs in the blood and milk of people from Broughton Island. These committees were created to position the territorial government to respond quickly and effectively to similar situations, as well as to the contaminants issue in general. The NWT Technical Committee on Arctic Contaminants (TCAC) acts as one of the territorial links to the AES-NCP. It develops strategies and priorities for collaborative study, and provides information to the public about the presence and possible effects of contaminants in the NWT. Regional Health Boards and committees are linked to these committees through the membership of the NWT Aboriginal organizations as well as through a reporting relationship with NWT government Health representatives. The TCAC is chaired by the GNWT, and comprises representatives from Health Canada, the Science Institute of the NWT, the regional offices of a number of federal departments (Fisheries and Oceans, Environment Canada, Indian and Northern Affairs), and several Aboriginal organizations (Dene Nation, Métis Nation-NWT, Inuvialuit Regional Corporation, Nunavut Tunngavik Incorporated, Inuit Tapirisat of Canada).

5.2.2 Capacity-building

Capacity-building within communities provides committees or individual community members with first-hand experience in project development and implementation. General public education initiatives are an important element in building general public awareness of issues, and to date, this has been achieved most often through community workshops (described in Section 5.3.1). However, capacity-building is also being created through the involvement of the Aboriginal Partners in the overall management of the AES-NCP, from both the technical and the policy perspective, as described above.

Through the partnership between the northern Aboriginal organizations (Métis Nation-NWT, CYFN, ITC, ICC, and Dene Nation), these organizations have been able to develop their own internal capacity to work on contaminants issues and other important environmental issues with their constituents. The Aboriginal Partners play several roles within the NCP. By participating in the NCP management structures, they provide advice and represent northern Aboriginal interests to DIAND and the other NCP government partners, contributing input on communications as well as research priorities. For instance, the Aboriginal partners have been able to work closely with many of the scientific partners involved in Arctic research, advising them on issues such as methods for improving community involvement, appropriate contacts in regions and communities, and specific communications problems related to their work. As a result, the NCP is better able to respond to the needs and wishes of northerners when designing and delivering programs and projects.

Another crucial element of the Aboriginal Partners' work with respect to the NCP is in developing communications links. By working to improve and systematize communications with their own constituents at the regional level, they have become better able to represent those priorities within the NCP. Equally important, the Aboriginal partner organizations and communities have been able to make better use of NCP information and opportunities for funding. As a result, capacity to address contaminants issues at the community level has in recent years increased significantly. This is demonstrated by the community-driven project proposals that have been submitted to the NCP and approved for funding. Such projects (some of which are discussed in Section 5.2.4) contribute to technical as well as administrative capacity at the community level, and range from specific scientific contaminants studies to communication projects by local environment committees.

Finally, the capacity developed within the Aboriginal organizations has permitted them to participate at the national and international levels to ensure their positions are considered in policy development, including arguments for control of contaminants. This is well illustrated by their active participation in the Arctic Environmental Protection Strategy and the Arctic Council, as well as the important role played by ICC in pushing the UN ECE Executive Body to move forward with a protocol for Persistent Organic Pollutants. Within Canada, the Aboriginal organizations' increased facility for addressing the contaminants issue has enabled them to have a stronger voice at national consultations (e.g. the Dene Nation has a seat on Environment Canada's National Air Issues Coordinating Committee Consultation Group on POPs).

5.2.3 Community-Researcher Relationships

The NCP, as a partnership of scientific and Aboriginal organizations, is committed to optimizing the working relationships between NCP scientists and northerners. In the fall of 1993, at the request of the NCP, the Inuit Tapirisat of Canada prepared an issues paper, *Negotiating Research Relationships in the North*, to stimulate discussion on these issues. Subsequently a special AES workshop was dedicated to formalizing the NCP's response to this issue.

As a result of that workshop, the NCP prepared a document entitled, *Researcher Guidelines for Planning Communications and Community Participation*. This document outlines a series of requirements set out for researchers to integrate community participation and communication in all stages of their research, from proposal development to the reporting of project results. These Guidelines were endorsed in 1994 by the Science Managers and issued as part of the application package for all research proposals to the NCP starting in 1994/95. Funding from the NCP is contingent upon demonstration (to the satisfaction of the Technical and Science Managers Committees) of the project plan for adherence to these guidelines.

It should be noted that because the Researcher Guidelines only recently (1994/95) became a condition of funding, there has been no opportunity to evaluate their effectiveness or conduct an exhaustive review of the specific activities undertaken on a project-by-project basis.

5.2.4 Cooperation in Research

While all NCP projects must adhere to the Guidelines, the level of community interest and involvement can vary according to the project and the community. However, in the case of research on human health (i.e. collection of data on humans, thereby directly involving individual community members) the level of community participation in the planning and management of a project and subsequent dissemination of results is necessarily very high.

Three institutions in particular, have been heavily involved in the human health research projects of the AES. GNWT Health and Social Services has been working to establish a program for baseline monitoring of contaminants in human tissues, specifically maternal and umbilical cord blood and mothers' milk. The Centre for Indigenous Peoples Nutrition and Environment (CINE) examines many aspects of the diet of Northerners, including: consumption of store-bought and traditional foods, nutritional value and contaminant levels in various foods, relative contribution of various foods to intake of protein, fat, energy and essential nutrients; and effects of food preparation methods on contaminant levels in food. Centre Hospitalier de l'Université Laval (CHUL) conducts a wide range of human health research projects in Nunavik and Labrador, including: human tissue monitoring for contaminants, investigation of biomarkers for biological effects, dietary surveys and exposure assessments, and studies on developmental and immunological effects in infants.

Although not all of the above-noted work involves the sampling of human tissues, all has the potential to be considered personally intrusive, and all requires the full support and participation of individuals and communities as a whole. As such, all of the researchers doing studies in this field have devoted a significant effort toward developing good working relationships in the communities where projects are conducted, prior to the commencement of any investigative work.

Laying the groundwork for a research project may involve any or all of the following:

- formal negotiated community-researcher agreements
- one-on-one consultations with expectant mothers
- training and education of community members who will assist in the research
- extensive consultation and information exchange with key community members in issue identification and project planning
- information sessions for the community at large
- training and education for all study participants
- education of health professionals in the communities, and
- follow-up and results reporting

Each of GNWT Health and Social Services, CINE and CHUL have adhered to the principles that respect the necessity of taking a cooperative approach to research of this nature. They have, among other things, put in place infrastructure, established contacts, developed communications tools and materials and/or negotiated research agreements with the communities in which work is being conducted. The following section describes, from a communications perspective, the work that each of these organizations has been doing as part of their human health research.

5.2.4.1 GNWT Health and Social Services — Contaminants Baseline Monitoring

GNWT Health is undertaking a baseline monitoring program for contaminants in the umbilical cord blood of mothers in three administrative regions: Mackenzie-Kitikmeot, Keewatin (Kivalliq) and Baffin. Consultation, sampling and analysis have been completed in the Mackenzie-Kitikmeot region. Sampling has been undertaken in the Baffin region and consultation is being initiated in Kivalliq.

Once funding for the blood monitoring program was approved, consultative working groups were established in the regions. Among the activities that were then undertaken were consultation with health boards, identification of issues related to contaminants and development of materials such as protocols for participant recruitment, consent forms, communications strategies and tools, and information packages for potential participants. The programs are administered at the level of the Regional Health Boards, with a designated Contaminants Project Coordinator in each region leading activities. This includes in-service training for community health nurses, hospital physicians, ward staff and research assistants, as well as workshops and regular project updates with regional health board trustees (who represent each community).

5.2.4.2 Centre for Indigenous Peoples' Nutrition and Environment (CINE)

Through the involvement of the Aboriginal Partners and funding from the AES, CINE was established at McGill University as a permanent research and education resource for indigenous peoples. CINE undertakes community-based research and education related to traditional food systems and nutritional well-being. The knowledge of the environment inherent in indigenous societies is incorporated in all of its efforts. CINE provides research training to community members through each of its research projects as well as in community workshops. CINE is directed by a governing board consisting of representatives from the Dene Nation, the Métis Nation–NWT, the Inuit Tapirisat of Canada, the Inuit Circumpolar Conference, the Council of Yukon First Nations, and the Assembly of First Nations, with the Mohawk community of Kahnawake acting as host.

CINE has established a response system to address questions or concerns arising from communities. If information already exists to answer the questions, it is made available to the community in an understandable form. If the questions pose a new research need, CINE will work with the community in developing a research project to address the questions.

All of CINE's AES work is done in response to community requests which are submitted directly, or through the CINE Governing Board. The latter decides which projects should be pursued and seeks funding through either external (e.g. as project proposals to the AES) or internal (from core funding) sources. Community meetings and discussions with band or hamlet councils lead to the development of written research agreements that outline study purpose and scope, extent of community training and participation, information collection, sharing, distribution and archiving, confidentiality or anonymity requirements, communication (e.g. of project progress to the community, media and outside parties, funding agencies), expected benefits and potential problems to community and expected benefits for the researchers. The intent of such agreements is to ensure a balanced relationship between the researcher and the community.

Return of information to the communities is an ongoing process involving progress reports addressed to local leaders and collaborators, radio interviews, local newspaper articles, posters, and invited presentations at relevant meetings. For example, as results from the project "Variance in Food Use in Dene/Métis communities" were becoming available, presentations were made at the Annual General Assemblies of the Dene Nation and Métis Nation–NWT, and at various environmental workshops sponsored by NWT Aboriginal organizations such as the Dene Nation, Inuvialuit Game Council, Gwich'in Tribal Council, and the Sahtu Wellness Initiative. Communities were also kept informed through CINE's Newsletter. CINE staff also undertakes short training courses at the request of communities and organizations, such as two Sahtu nutrition courses offered in Fort Good Hope in December 1995.

For CINE, the returning results may signify the end of a particular project as well as the beginning of another, as community members become interested in developing other nutrition-related initiatives and additional capacity is developed. In K'asho Got'ine (Fort Good Hope) for example, results of previous assessments showed inadequate calcium and vitamin A intake. A project was then designed in partnership with community members involved in previous dietary assessments to analyse factors affecting food selection and identify underutilized sources of these nutrients. The final intervention to promote appropriate traditional and market food sources of calcium and vitamin A is expected to be developed as a community initiative.

5.2.4.3 Centre Hospitalier de l'Université Laval (CHUL)

CHUL conducts its research in Nunavik (northern Québec), and communication activities in this region have been carried out since the first contaminants study was set up by the Kativik Environmental Health Service in 1989 (prior to the AES-NCP), at which time a community-based contaminants committee was created. The "PCBs Committee," comprising Health Centre representatives such as midwives and nurses, as well as members of the local Health Board and the Puvirnituq Women's group was, until 1994, a forum for on-going training on the issue of contaminants in the food chain and for dissemination of related information to the communities.

Discussions between researchers and regional members of the committee helped develop skills required to fulfill the mandates of the committee regarding community information. This enabled committee members to provide community members with information about the project purpose and methodology, and background on the issue, on an ongoing basis. The role of nurses and midwives on the committee included coordination of the collection of milk samples and gathering of data (via questionnaire) in their Health Centre. Immediately after the first meeting, a television phone-in show was broadcast across the North and a pamphlet on PCB contamination was produced in English and Inuktitut. In the three years following the beginning of this study, a few local phone-in shows were organized but one-to-one exchanges of information frequently occurred because most committee members provided interactive information when attending to mothers participating in the study.

In 1994, the Committee was renamed "The Food, Contaminants and Health Committee." Its expanded mandate included northern nutrition, discussion of risks and benefits, educational and health promotion programs, economic and cultural values. Its main priority was to define and make available, for the people of Nunavik, data from different knowledge sources on the issue of food and contaminants in a manner relevant to a broader definition of health (i.e. encompassing physical, mental, socio-cultural and economic perspectives).

The Committee collaborated with Taqramiut Nipingat (a Nunavik radio and television producer) to produce a video and a series of four public service announcements (PSAs) targeted at pregnant and nursing women, families, and teenagers. The PSAs were still in preparation as this report was in production, and were to be pre-tested before presentation on television in autumn 1996. The video, available in Inuktitut and in English, was aired in the spring of 1996. It provided background information on how contaminants are transported to the Arctic and how they enter the human body and presented the conclusions of the studies (a survey of organochlorines in human milk and Santé Québec's health survey among the Inuit) and recommendations linked with the proven benefits of eating country food.

In 1996, the Public Health Department of the Nunavik Regional Board of Health and Social Services decided to establish a permanent regional advisory committee to define and promote policies and programs on nutrition and health, including perspectives related to physical, emotional, spiritual and mental well-being. The two existing committees dealing with nutrition topics (Food, Contaminants and Health and the Steering Committee of the Avativut/Ilusivut Program), merged and were joined by members of organizations such as the Kativik Regional Health Department and Makivik Corporation. The expertise of the two committees is now consolidated into a unique forum with a broader scope to examine all nutrition issues related to health in Nunavik. The mandate of the committee is to advise and support the Public Health Department, and other organizations on:

i) the development of a food policy in Nunavik,
ii) the development of nutritional components within Regional Health Programs,
iii) research projects regarding nutrition and to support them when relevant and needed,
iv) communication with the population on all issues linked to nutrition and health, and
v) to fulfill the role of the Steering Committee for the Nunavik component of the Avativut/Ilusivut Program.

Complementing the AES-NCP activities, many of the health research communications activities undertaken in Nunavik through CHUL result from the Avativut/Ilusivut ("our environment/our health") Research Program. This is a three-year project funded by Environment Canada through a grant from the Tri-council Secretariat to study contaminants in the food chain of Inuit of the eastern Canadian Arctic. A key objective of the program is to collaborate with Inuit groups to formulate appropriate risk management strategies and to meet the needs of the residents of Nunavik and Labrador. The first regional risk management workshop was organized in January 1995 in Nunavik and Labrador, bringing together researchers, Steering Committee members, field coordinators and key individuals from local communities including elders and hunters. The Avativut/Ilusivut Program also publishes a quarterly newsletter in English and Inuktitut that is distributed in Nunavik, across Canada and in other circumpolar countries.

CHUL researchers generally follow a specific consultative process for individual projects. Dr. Gina Muckle and collaborators are conducting research on transplacental exposure to PCBs and infant development. The first stage of this project involved a pilot study that determined the feasibility of such a study in Nunavik. The intent was to determine if the small population size and cultural and linguistic differences between Inuit and a "typical" southern test population might influence the measures that would be used in the study. During this pilot study, Dr. Muckle worked with the midwives of the Inuulitsivik Health Centre in Puvirnituq. The final stages of research began in November 1995 after Dr. Muckle organized meetings and activities with various northern agencies and community groups such as the Board of the Nunalirtuquait Ikaluqatigiitut (for prevention of alcohol and drug abuse), the Mayor and the Municipal Council, the Health Committee, and women's group representatives from the five participating municipalities (Kuujjuaq, Salluit, Purvirnituq, Inukjuak and Kuujjuarapik). Furthermore, four phone-in shows were organized on the local FM radio stations and an article was published in the Avativut/Ilusivut newsletter.

The process with individual study participants began with presentation of information about the objectives and procedures for the study. Written consent was obtained from all individuals wishing to participate in the study. A child development specialist worked with each mother to evaluate her infant, provide feedback, and answer questions about the infant's development. As agreed with the Board of the Nunalirtuquait Ikaluqatigiitut and the mayors and councils of participating municipalities, Dr. Muckle will personally present the study results, with the support of the Nunavik Nutrition and Health Committee.

5.2.4.4 Other Projects

While the human health projects provide the best examples of community participation, other studies have also taken a cooperative approach to research. It has long been recognized that Aboriginal peoples have valuable information about natural systems. The challenge, however, is to go beyond a mere cataloguing of this knowledge.

Ed Carmack and Robie Macdonald, from the Institute of Ocean Sciences (Dept. of Fisheries and Oceans), worked with the late Jimmy Jacobson, an elder from Tuktoyaktuk, to incorporate traditional knowledge into the study design for an investigation of the oceanography of Husky Lakes. In this project, the authors found four specific examples of traditional knowledge that led to focused scientific enquiry. The first study used local knowledge of ice characteristics and fish abundance as a basis for interpreting tidal dispersion and its effect on water mixing during winter. The second concerned the maintenance in winter of a small ice-free area, used by caribou as a salt lick, by heat flux associated with channel flow. The third concerned overflooding caused by westerly winds in mid-winter. The final example described the importance of wind direction to ice conditions and brine formation. By using dialogue to guide the study, the authors were able to apply oceanographic techniques to topics relevant to the use of the region by Aboriginal peoples.

The Fisheries Joint Management Committee (FJMC), a co-management body established pursuant to the Inuvialuit Land Claim Final Agreement, conducted a research project on the mercury and organochlorine content of beluga whales. The study was carried out in cooperation with scientists from the federal Department of Fisheries and Oceans (DFO). It is an example of a study in which the need and interest in a specific issue was common to both the NCP and the local people, and which had been identified at the community level. Within the region, community members were able to work with scientists and bring together the relevant expertise in developing a project that addressed the identified needs. Tissue samples were collected by local hunters and DFO regional field representatives, according to agreed-upon protocols, and sent to DFO laboratories for analysis. As per agreement with the FJMC, the study results were not made available to other agencies until the FJMC had an opportunity to review the data and reports generated by DFO.

The Fort Resolution Environment Committee submitted a proposal to the NCP in March 1996, for study of organochlorine and metal levels in fish and sediments in the Fort Resolution Domestic Fishing Zone. This proposal was approved for funding, and a partnership with Dr. Marlene Evans (of the National Hydrology Research Institute) who had previously conducted similar work in the area and who had also submitted a proposal for 1996, was facilitated through the NCP. As a result, the Fort Resolution Committee and Dr. Evans met and developed a collaborative project that began in summer 1996.

Another similar project was initiated by the Kaska First Nation in the Yukon, who submitted a project proposal to the AES via the Yukon Contaminants Committee. Working with a local consultant, samples from a wide range of country foods were collected and analysed for metals and organochlorines. Identification of the species to be sampled, collection of samples, and reporting of results were coordinated in agreement with the Kaska.

5.3 Communication with Communities

Recognizing the importance of communication at the community level, the NCP has in recent years, and as scientific results have become available, devoted an increasing proportion of program resources to the mutual exchange of information and ideas between the program and northern residents. This includes dissemination of program results, but also extends to issues such as education, gathering of community concerns and priorities regarding contaminants, development and provision of resources, research on past communication activities, etc. Although spearheaded by the Aboriginal organizations that participate in management of the NCP, communications are now considered to be a responsibility of all program participants, with certain organizations (e.g. GNWT Health and Social Services) having an active role in many of the NCP communications initiatives. Some of these activities are described below.

5.3.1 Community Workshops

An element common to all of the Partners' activities is the conduct of environmental workshops targeted at community people in the Partners' respective constituencies. ITC has conducted annual environmental workshops throughout the north for the past five years, the Dene Nation organized Contaminant Workshops (for each of five regions) at the 1995 Dene National Assembly, as well as an environmental workshop for youth, elders and scientists in 1996, and the Métis Nation–NWT conducted a Contaminants Communication Workshop at their 1995 NWT Annual General Assembly. Other AES-funded workshops have been joint initiatives involving several organizations. For example, through the NWT-TCAC, the Gwich'in Renewable Resource Board, Inuvialuit Game Council, territorial health department (GNWT), DIAND-NWT, and the Science Institute of the NWT collaborated to organize a 1995 workshop in Inuvik. In 1996 the GNWT territorial health department coordinated a workshop in Cambridge Bay with Pauktuutit, the national Inuit women's association. Since 1992/93, the Yukon Contaminants Committee has held annual environmental workshops in cooperation with CYFN, territorial departments, Yukon College and local environment committees and public interest groups, such as the Yukon Conservation Society.

These workshops allow for the exchange of information on contaminants, AES activities and research results with leaders, youth and other community members. They also allow for the exchange of traditional knowledge and create an opportunity for the NCP to learn first-hand, via the Aboriginal Partners, the grass roots concerns and information gaps with regard to contaminant information. Feedback from these workshops is discussed in Section 5.4.

5.3.2 Aboriginal Partners Projects and Programs

The issue of communication and mutual information exchange between the NCP and northern communities, particularly Aboriginal northerners who are the main consumers of traditionally harvested foods, is multifaceted and can be approached from many different perspectives and to varying degrees of detail. The Aboriginal Partners, while increasing their internal capacity to address contaminants issues, also work to find ways of improving the NCP's communication and working relationship with its primary 'client' group, consumers of country foods. This is an ongoing activity, with one-to-one contacts, workshops, newsletters, and fostering of local contaminants committees, local environment committees, or regional program officers being among the initiatives that the Partners continue to pursue. However, the Partners also engage in specific individual projects that may address single needs or particular issues identified over the course of their ongoing activities. Some of these are described in the following sections.

5.3.2.1 Inuit Tapirisat Canada

ITC research staff, through annual environmental workshops and other activities, have been listening to the concerns of Inuit and have passed on these concerns in presentations to research associations. The ITC paper *Negotiating Research Relationships in the North* formed the basis for planning and discussion at a workshop organized by ITC and DIAND that resulted in the NCP developing and adopting the *Researcher Guidelines for Planning Communications and Community Participation* (discussed earlier). Work is underway for a guidebook for communities on their rights and choices when it comes to negotiating research relationships.

Recognizing that ineffective communication has been a significant barrier to Inuit understanding of contaminants at the local level, ITC examined how communication occurred in a variety of contaminants incidents involving country food in Aboriginal communities, and identified Inuit knowledge and communication needs regarding contaminants in three Inuit communities. The research report, *Communicating about Contaminants in Country Food,* was also widely distributed in plain language summary form in English and Inuktitut, in Inuit communities.

5.3.2.2 Dene Nation

The Denendeh National Office and Mackenzie Regional Health Service developed nutrition fact sheets to illustrate the nutritional and socio-economic benefits of Dene traditional foods and assist communities in finding alternative traditional food sources, should health advisories affect their nutrition intake.

In addition to generating information for distribution to communities, the Dene Nation is working to enable efficient outreach to communities. For example, an electronic communications network was developed with the Dene Cultural Institute to allow for remote access of communities to information on contaminants, the AES and other environmental issues. Environmental committees, now established in a number of communities, can act as a contact point for AES research. These committees are developing the capacity to feed into the NCP, as illustrated by the project proposals approved for funding in 1996, which were developed and submitted to the AES by some of the local committees.

5.3.2.3 Métis Nation – NWT

One of the key priorities for NCP communications, is the provision of general knowledge on the issues and concepts that are fundamental to understanding contaminants in arctic food webs. The Métis Nation Contaminant Education Program Contaminants Curriculum Project integrates contaminant information and materials with existing grade school curricula and the adult education programs across the NWT. Wherever possible, these materials are cross-referenced to Dene Kede and Inuuqatigiit school materials that validate traditional values and perspectives. This education program will be distributed to more than 70 grade schools across the NWT, Aurora and Arctic College campuses and Community Learning Centres. It will also be available to the Yukon and other interested educational jurisdictions.

Further supporting the theme of education on contaminants has been the development of a general overview booklet on the NCP. The Métis Nation–NWT assumed the responsibility for publication of a lay person's guide to the NCP. This illustrated booklet, prepared in collaboration with DIAND, was published in English and uses non-technical language to explain the main issues and concerns of the program. Standard graphic icons were developed to represent the contaminants, and concepts such as biomagnification were illustrated with appropriate northern species. The booklet also guides readers to further sources of information.

5.3.2.4 Council of Yukon First Nations

CYFN communications activities under the NCP have been integrated into a holistic environmental education program that has included workshops and one-on-one consultations with community leaders and environmental representatives. Over a three-week period in March 1995, workshops were conducted in Whitehorse to explore a broad range of environmental issues having both local and international scope. The workshops were targeted at First Nations people who work in the fields of environment or renewable resources, and were also open to community members or groups with a specific interest in environmental issues. While part of the workshop was devoted specifically to contaminants in the food chain, mining, forestry and the environmental assessment process were also covered, including training seminars addressing resource management plans (land use and forestry).

Follow-up to the March workshops took place over the summer as one-on-one discussions with leaders and environmental representatives in the communities. The consultations were to determine what level of understanding members of the community had relating to environmental issues, specifically addressing contamination in traditional food and informed decision-making about diet, and to find out what kind of education on environmental issues was desired. In some areas, the use of traditional foods and medicines has increased.

5.3.2.5 Inuit Circumpolar Conference

The ICC held a contaminants workshop as part of their 1995 General Assembly. The General Assembly, held in Nome, Alaska brought together approximately 500 participants from Chukotka, Canada, Alaska and Greenland, as well as artists, media delegates, scientists, and government representatives from each country. One of the recommendations from workshop participants was to develop an Inuit International Action Plan to assist Inuit in working with contaminants researchers. This plan would also outline strategies for dealing with producers and users of pesticides and for promoting the benefits of country foods. The workshop participants also pointed to the need for further research efforts to focus on defining acceptable levels of environmental contaminants.

5.3.3 Communicating About Health Benefits and Risks

An important responsibility of the NCP is to ensure that people have an accurate understanding of the risk of contaminants in traditionally harvested foods in relation to the benefits of a traditional diet and the consequences of abandoning this diet. The NCP has attempted to develop an integrated view of risk with respect to contaminants in country foods, with the intent of enabling people to make judgments about risk based upon the most complete information possible. This includes recognition of the unique and important social and cultural values of traditional food systems to Aboriginal people, over and above conventional nutritional values.

The AES-NCP works to improve risk communications by:

- filling information gaps about contaminants and thereby contributing to greater certainty
- placing emphasis on communications and partnerships to develop strategies for managing risk
- bringing together scientists and community representatives, thereby helping scientists understand what kinds of questions the communities are asking
- supporting diet surveys and nutrition research to highlight the benefits of country food.

5.3.3.1 A Process for the Conduct and Communication of Health Risk Assessment and Management

In previous years, the process of information flow about Human Health Risk Assessments did not pass from the scientists and responsible government agencies to the communities through the Aboriginal organizations. Like the communities, the latter would only see the end result, not the data, assessment and reports that led to a decision on the content and issuing of a Health Advisory. In response to urging from the AES Partners, a change to this process was initiated under the NCP as it became clear that Aboriginal organizations must have the knowledge and capacity to be involved with health assessment decisions concerning their member communities. Where the NCP and its participants are concerned, the health risk assessment and management process now involves three main stages.

1. **Initiation and conduct of assessment** (refer to Chapter 4, Figure 4.4.2). As agreed to by the Science Managers, this stage involves the transfer of raw data from the initiating party (i.e. government departments, Aboriginal organizations, communities) to Health Canada, and the distribution of this data and all relevant correspondence to the other government departments and the Aboriginal organizations. Completion of the health assessment by Health Canada leads to stage two of the process.
2. **Evaluation of assessment results.** This is mediated through territorial contaminants committees. Each committee will adapt its response based on the results and communities affected. The processes are essentially similar in that consultation with regional and community representatives is undertaken. This allows for the consideration of nutritional, cultural and social benefits of continued harvesting and consumption — factors that are not incorporated into the Health Canada assessments. It also ensures evaluation of the significance of the assessment with respect to factors such as species harvested, tissues analysed and safety factors used in establishing recommended maximum weekly intake levels.
3. **Communications.** Contact with communities and the public at large is mediated through the territorial contaminants committees. All participating parties reach agreement on key messages and release of the assessment and evaluation results. In the Yukon, ultimate legal responsibility for issuing health advice rests with the federal government (Health Canada) while in the NWT this responsibility has been devolved to the GNWT Department of Health and Social Services.

5.3.4 Community Feedback

Communication of general information about contaminants and NCP activities is a continuously evolving process, as program results become available and as community needs, priorities and expectations change.

Throughout the course of the NCP, program participants (scientists, territorial and federal governments, and Aboriginal organizations) have had opportunities to interact with people at the local level and receive feedback on areas of concern and interest. Sometimes, through formal environmental fora such as workshops, and often during informal contact with researchers during their work, questions are asked, information is provided and knowledge is exchanged. While the NCP must continue to work toward improving this dialogue, the workshops, research projects, and research agreements that have been realized over the six years of the program's existence have yielded information that can contribute to improving NCP activities in the coming years. Also feeding into the policy deliberations are the recommendations and perspectives that have come out of workshops held by the Aboriginal partners and territorial contaminants committees, and reports such as the ITC report *Communicating about Contaminants in Country Food.*

5.3.4.1 Community Information Needs and Priorities

The Program has come to recognize the holistic view of health and the environment that Aboriginal Northerners have. This therefore influences how information is presented, since it is not possible to discuss "long-range" transported contaminants in isolation of a wide variety of other environmental considerations. Contaminants are regarded by most northern people as part of food, health and the environment, and not necessarily narrowly defined as long-range airborne chemicals or metals. For example, in Baker Lake, contaminants in Inuktitut is expressed as *ulurianaqtuq* (the product of something dangerous), which can include chemicals, oil spills, garbage, viruses and even alcohol. Two basic types of information are needed: specific information about a contaminants situation and general background information on contaminants.

The following provides an unedited overview of needs and priorities related to contaminants as expressed by communities through the many workshops and meetings held by the NCP partners:

- What information do people want?
  - Safety of traditional foods.
  - Levels of contaminants in plants and animals.
  - How contaminants move through the food chain.
  - Why restrictions are put on food consumption.
  - Benefits and risks of eating traditional food.
  - What can be done to minimize the effects of contaminants.
  - Procedures for submitting samples for testing.
  - Other health risks.
  - About the Arctic Environmental Strategy.
  - Local contaminants.
  - Diseases in wildlife.
  - Radionuclides.
  - Contaminants in store bought food.
  - List of current environment and health committees.
  - Connections between cancer and local/distant contaminants.
  - Effects of oil spills on safety of beluga and bowhead.
  - What migratory birds eat during migration.
  - How do mercury, PCBs and other contaminants get into water, air and land.
  - How to tell if an animal is contaminated before you eat it.
  - Long-term versus short term effects of contaminants (on human health and the environment).
  - What do different contaminants look like in animals and what are their effects?
  - Where do contaminants come from, and how are they produced?
  - How are assessments done?
- Community concerns
  - Contaminants (including radiation) from mines and other local sources such as garbage dumps, Dew Line sites, other military activity.
  - Radionuclides and caribou.
  - Ways to deal with people who violate the environment.
  - Cross border pollution from pulp and paper industry.
  - Need for greater community involvement and use of traditional knowledge.
  - Security and confidentiality of personal information, e.g. blood monitoring.
  - Heavy metal contaminants.
  - Community control over research.
  - Observations of deformities in wildlife — what can people do about this.
  - Mercury from hydroelectric development.
- Recommendations for future action
  - Progress on international conventions.
  - Local action on clean-up.
  - Increased capacity of communities and regions to address environmental concerns and set priorities.
  - More emphasis should be placed on individual responsibility.
  - Increased funding to bring information back to the community.
  - Encourage openness about research.
  - Involvement in baseline monitoring, particularly before greater expansion of mining industry.
  - Focus on community action initiatives.
  - Community petitions for changes in research attitudes and policies.
  - Increased involvement of Community Health Representatives (CHRs), health committees and board members and community agencies.
  - More basic information on environmental issues should be provided through formal channels.
  - Improved access to information for single mothers.
  - Incorporation of traditional knowledge.
- How do people want to receive information?
  - Published list of contacts where more information is available.
  - Information should be in the schools.
  - Posters.
  - Use of local media, especially local radio and TV.
  - Local agencies should be encouraged to present information.
  - Local clean-ups to raise awareness.
  - Visual presentation of information or in a non-technical language.
  - Slides, videos and photos.
  - Environmental specialists in each region.
  - Workshops on specific topics.
  - Translated and interpreted information.
  - Glossary of contaminants terminology — consistent translations.
  - Use northern produced information/education materials — not southern.
  - Presentation by community members to band/hamlet councils, and politicians.
  - Interesting materials in health waiting rooms.

  - From a trusted, locally credible person.
  - From elders.
  - Face-to-face.
  - From members of the Hunters and Trappers Association.
  - Information should be delivered during the hunting/fishing season.

5.3.4.2 Traditional Knowledge and Local Observations

Many of the questions that researchers are working to answer involve information needs that can only be met through long-term observation, detailed familiarity with the environment, capacity to recognize changes and abnormalities in the environment, and continued sampling and monitoring. Typically, for 'western' science, collection of this type of information is very costly, logistically difficult, time-consuming and often only seasonal. However, the traditional knowledge that is held in communities represents the accumulation of generations of year-round observation, sampling and monitoring, combined with an intimate familiarity with the local environment. A number of individual NCP projects have already incorporated traditional knowledge, although often the focus has been on identifying good sampling locations and times. However, there is a growing recognition that traditional knowledge can also contribute to program planning.

Among other things, traditional knowledge may reveal some general areas of contaminants research that would benefit from the information available on changes in the environment. In addition, the potential research assistance that could be offered by people with extensive current and historic knowledge of the local environment has significant potential to act as, for example, a basis for planning future monitoring programs. The possibility of using traditional knowledge to improve 'western' scientific approaches in hypothesis formation, research planning and policy development exists. It represents a significant challenge because there are few, if any, practical models of specific application of traditional knowledge in scientific program planning. However, the NCP represents a unique opportunity to develop working examples of this integration.

For example, Aboriginal elders across the north have reported a number of changes in terrestrial and aquatic wildlife. The causes of these changes are unknown, but they do lead to questions about the plausibility of a relationship to anthropogenically induced environmental changes, including contamination. Similarly, certain wildlife species might be at greater risk of effects from contaminants. But if such effects are being experienced, they are likely to be subtle and, in light of the intergenerational nature of many of the suspected effects, perhaps not easily detectable within the timeframe of a "typical" scientific research project. Given the associations that have been made between organochlorines and effects such as reproductive impairment, developmental and behavioural abnormalities, it is clearly important to be aware of all of the observations that elders and hunters have made, and the long-term record that these may provide.

To date, the NCP has not formally attempted to collect and analyse traditional knowledge. However, through feedback received at workshops and community meetings, information on local observations has been recorded on an *ad hoc* basis, and it is described here as an example of the types of information that can be gained through consideration of traditional knowledge. The observations described below have not been sorted or selected for a possible direct relationship to contaminants program research, nor has any attempt been made to link them with 'western science' knowledge. They are presented to give an overview of the breadth of knowledge that is currently available. The observations are arranged according to the organization that collected and contributed them, and have been collected from workshop reports, the ITC report *Communicating about Contaminants in Country Food* and other research reports.

Inuit Tapirisat of Canada

- Sanikiluaq
  - Association of "invisible" contaminants with trichinosis in walrus.
  - Consumption patterns and chain of animals, e.g. ravens and gulls are unfit for human consumption because they eat feces. Resident animals can be eaten raw because their food sources are known, while migratory birds are cooked.
  - In the Inuit tradition, pregnancy is a time of restraint, when the mother's diet and behaviour need to be closely regulated. This may make it easier to communicate risks to the group that is most vulnerable to contaminants risks.
- Baker Lake (Qamanittuaq)
  - Elders would not eat animals that were wounded or had sore joints, or if they had a lemming in the stomach (lemmings carry a lot of diseases).
  - Look for oddly shaped or out of place organs.
  - Observed change in the texture of caribou meat. Meat is pale, almost whitish, while in past it was a good, deep red.
  - Undernourished caribou with sores on body or shedding badly.
- Tuktoyaktuk
  - Observations that incidence of brucellosis and tape worms in caribou has increased over the past 15 years.
  - Increase in white spots on internal organs and meat as well as swollen joints in caribou.

- Smaller, thinner and softer appearance of ocean-caught fish, fish with spotted, shrivelled or discoloured livers, and herring with more white spots on their flesh (and different taste).
- Increased hair loss in community members from Tuktoyaktuk.
- Concerns that cancer is more widespread.
- Recent marked decline in rabbit and ptarmigan is blamed primarily on local industrial activity and non-local sources.

Avativut report

- Nunavik and Labrador
  - Bluish caribou livers, especially in young, other changes.
  - Fewer ducks, geese, other birds.
  - Changes in caribou fetus weight and size, non-pregnant females.
  - Abnormal caribou behaviour, staggering.
  - Caribou lungs stuck to rib cages, some with black spots.
  - Unexplained caribou deaths (no marks on carcasses).
  - Sores on the insides of seals.

Dene Nation

- Changes in water quality, especially the Mackenzie River.
- Changes in the number of migratory birds (decreasing).
- Changes in animal behaviour.
- Changes in fish quality and quantity.
- Increases in cancer.
- Increases in deformities in harvested animals.

Yukon First Nations

- Decreases in the quantity and size of fish (whitefish and trout) eggs.
- Changes in texture and consistency of fish flesh.
- Altered behaviour in spawning salmon (e.g. reduced distance travelled upstream).
- Decreases in size of male caribou in the northern Yukon.
- Decreases in the size of caribou calves.
- Increased incidence of deformities in moose and caribou horns.
- Changes in colour of caribou and moose liver (darker colour), changes in taste of kidneys (bitterness).

5.4 Impact of the NCP: Domestic and International Action on Contaminants

The use of NCP results spans a wide range — from responding to the community level information needs described above, to substantiation of a need for action under domestic and international policy. NCP results have been critical in demonstrating the importance of long-range transport of contaminants on a global basis. NCP participants, supported by leading edge scientific evidence, have raised domestic and international awareness of the immediate human health and environmental concerns of long-range contamination, thus stimulating an accelerated pace of action toward control of such substances, both nationally and internationally.

5.4.1 Domestic Initiatives

5.4.1.1 Toxic Substances Management Policy

The federal Toxic Substances Management Policy (TSMP), announced by the Minister of the Environment in June 1995, focuses on anthropogenic substances that are characterized by persistence, a tendency to bioaccumulate, and toxicity. The compounds of concern under the NCP share these properties, in addition to being semi-volatile and therefore prone to long-range transport. Under the TSMP, substances that are predominantly anthropogenic and meet (or exceed) the prescribed criteria for persistence, bioaccumulation and toxicity are scheduled for virtual elimination from the environment (considered 'Track 1' chemicals). This means that no measurable release to the environment from any source is acceptable, and Canada will take action at both the domestic and the international level. Other toxic substances that meet some, but not all, of these criteria undergo a life-cycle management process to prevent or minimize their release into the environment ('Track 2').

Research results from the AES-NCP contribute evidence of persistence and bioaccumulation for a number of substances that have been specifically identified in the TSMP as 'Track 1' chemicals — those targeted for virtual elimination. AES-NCP results also form a substantial part of Canada's case for international control of such substances. Through the TSMP, many of the compounds of concern in the Arctic will be controlled domestically. The TSMP also provides the mandate for Canada to act internationally with respect to a number of the substances of concern in the Arctic.

5.4.1.2 The Priority Substances Assessment Program under the Canadian Environmental Protection Act (CEPA)

Canada has taken a conscious policy direction to deal with toxic substances by assessing and prioritizing these substances based on the combined human and environmental factors established in the Canadian Environmental Protection Act (CEPA). A comprehensive review of each substance or group of substances is undertaken.

Since 1990, 69 substances have been examined by expert panels established by Environment Canada and Health Canada. Forty-four substances were contained in the first Priority Substance List and in late 1995, an additional group of 25 substances was recommended to the Ministers of Environment and Health by an Expert Advisory Panel. The goal is to complete examination of 100 priority substances by the year 2000. Of the 69 examined to date, 25 have been classified as "toxic" under the definition included in the CEPA.

The CEPA reviews are undertaken on single compounds, groups or classes of substances, effluents and emissions. No attempt is made to deal with mixed or combined substance effects. Management strategies for key sources of most of the substances declared "toxic" are now being developed under the Strategic Options Process (SOP). Under the SOP, Environment Canada and Health Canada work with stakeholders through Issue Tables (consultative groups representing federal and provincial governments, industry and non-governmental organizations) to develop strategic options to manage the CEPA toxic substances, and to recommend to the Ministers of the Environment and Health how best to address the problems associated with these substances. The SOP is being guided by the Federal TSMP and emphasizes implementation of best technologies and pollution prevention, supported by socio-economic analysis, followed by environmental monitoring programs where feasible.

5.4.1.3 Federal Sustainable Development Plan

The Government of Canada is creating a Commissioner of the Environment and Sustainable Development to hold government accountable for application of environmental considerations in its policies, operations and programs. Furthermore, a step towards sustainable development is being taken through the requirement for all federal departments to develop their own comprehensive sustainable development strategies. Each department is required to ensure appropriate consideration is given to the economic, social and environmental components of sustainable development. Given that health and economic prospects are influenced by the state of the environment, the Northern Contaminants Program becomes a critical part of the whole sustainable development strategy.

Objectives that figure prominently in domestic and international sustainable development programs include protecting the health of people and of ecosystems. With respect to contaminants, the challenge posed by sustainable development is to protect the environment and human health in view of the chemical products that are used or unintentionally emitted or discharged as by-products or wastes. *A Guide to Green Government* suggests this is best accomplished through application of pollution prevention methodology and recycling of products. Suggested actions include virtually eliminating anthropogenic, persistent, bioaccumulative, toxic substances. Long-term health and environmental implications result from the accumulation of these substances in the tissues of plants and animals. Because they persist in the environment, they threaten the use of country food as a sustainable resource, and have therefore been the focus of much research under the AES-NCP. The federal TSMP sets out its approach for assessing and managing the risks associated with toxic substances. Management techniques may be applied to prevent their release into the environment or to phase them out if containment is not possible.

5.4.1.4 Canadian Environmental Quality Guidelines

National Canadian environmental quality guidelines (CEQGs) for the protection of aquatic and terrestrial ecosystems provide useful science-based tools for the assessment and management of arctic contaminants. These resource-based guidelines are required under Part 1, s.8(1) of CEPA that states:

> For the purposes of carrying out the Minister's functions and duties related to the quality of the environment, the Minister shall formulate … environmental quality guidelines specifying recommendations in quantitative or qualitative terms to support and maintain particular uses of the environment.

Canadian environmental quality guidelines are developed in cooperation with the provinces and territories (Canadian Council of Ministers of the Environment) using nationally approved protocols and a formalized peer review and approval mechanism. CEQGs are recommended concentrations of substances in various media (water, sediment, biota tissue, and soil) to protect, enhance, and restore designated resource-uses of the environment such as drinking water, aquatic life and wildlife, recreation and aesthetics, and agriculture. For example, tissue residue guidelines, which address persistent bioaccumulative toxic substances, are levels in tissue of aquatic biota (e.g. fish) recommended to protect wildlife that consume aquatic biota.

Numerical guidelines provide scientific benchmarks for environmental quality that are easily understood, communicated, and implemented as the basis for management decisions. For persistent, bioaccumulative toxic substances (PBTS), the ultimate management goal under the federal Toxic Substances Management Policy is the virtual elimination of these substances from the environment. These types of substances tend to accumulate in Arctic ecosystems as a result of long-range transport. Environmental quality guidelines can be used to determine interim management objectives and as performance indicators to measure progress in national and international virtual elimination strategies (as per the Canada-Philippines International Experts Meeting on Persistent Organic Pollutants, meeting statement, June 4–8, 1995, Vancouver, BC). As well, CEQGs can be used to evaluate the significance of adverse risks to environmental and human health to help determine priorities, targets and schedules for further assessment, monitoring, and management actions so that limited environmental protection resources can be allocated most effectively.

For non-persistent or non-bioaccumulative substances that may be directly deposited into Arctic ecosystems, CEQGs are essential tools used to interpret the significance of measured levels of substances in the Arctic environment and, in turn, to estimate the degree and likelihood of adverse effects. These guidelines also provide scientifically based measures of sustainability for use in risk management for selecting and implementing appropriate management actions in conjunction with legal, and socio-economic factors. Furthermore, these guidelines can be used to determine priorities, select strategies and timelines to help achieve the goal of life-cycle management to prevent or minimize releases of these substances to the environment.

5.4.2 International Initiatives

Canada participates in and leads a number of international initiatives (Table 5.1) that are directly relevant to the assessment and control of contaminants that are of concern in the Arctic ecosystem. The NCP has been a driving force behind Canada's political commitment to pursuit of international action. Data from the NCP have provided a significant proportion of the scientific substantiation of long-range transport of such substances from source regions to remote areas of the world such as the Arctic. The studies have revealed the biological concern that can be generated by even barely detectable levels of these compounds in the abiotic environment. Using this evidence, while demonstrating the very real human health concerns that arise from the global transport, deposition, and subsequent biomagnification of persistent contaminants in the Arctic, the NCP has raised the profile, urgency and level of action on contaminants on the international agenda.

5.4.2.1 Arctic Environmental Protection Strategy (AEPS)/Arctic Council

At Rovaniemi, Finland in June 1991, Ministers from all eight Arctic countries signed a declaration adopting the Arctic Environmental Protection Strategy (AEPS). Ministers met again in Nuuk, Greenland (1993) and in Inuvik, Canada (1996) and expanded their commitments under the AEPS by signing two further declarations: the *"Nuuk Declaration on Environment and Development in the Arctic"* and the *Inuvik Declaration on Environmental Protection and Sustainable Development in the Arctic.*

The AEPS sets out the shared environmental objectives of these eight countries, and identifies the principles that should guide their implementation. It also reviews the nature of Arctic environmental problems and assesses the existing institutional and legislative arrangements for dealing with them. Finally, it identifies specific actions that should be implemented. One of these actions committed the eight governments to establish a comprehensive Arctic Monitoring and Assessment Programme (AMAP) dealing specifically with anthropogenic pollution of the Arctic environment. The AEPS also includes components that address Protection of the Arctic Marine Environment (PAME), Conservation of Arctic Flora and Fauna (CAFF), Emergency Preparedness, Prevention and Response (EPPR) and Sustainable Development and Utilization (SDU).

Specific to contaminants, the AEPS has also acted strategically to influence the wider international agenda by supporting or promoting action to control contaminants of concern in the Arctic. For example, in the Nuuk Declaration, the eight Arctic Ministers urged the UN ECE to pursue control of POPs and heavy metals under the Convention on Long-range Transboundary Air Pollution (LRTAP).

In September 1996, the Arctic Council was inaugurated and the AEPS was incorporated as a cornerstone of the Council's broader agenda. The Council provides a forum for the circumpolar countries, in close collaboration with Indigenous peoples, to seek consensus-based solutions for a wide range of arctic issues. This includes environmental protection as well as economic development, community health and development, cultural development and transportation and communications.

5.4.2.2 Arctic Monitoring and Assessment Programme (AMAP)

The mandate of AMAP is to monitor the levels of, and assess the effects of, anthropogenic pollutants in all components of the Arctic environment. It was

TABLE 5.1

Hazardous air pollutants (HAPs) targeted for action.

Substances	UN ECE LRTAP[1]	UNEP	Proposed Can/US Bin'l Strategy	NAFTA/CEC
Lead (*Alkyl lead)	X		*	
Mercury	X		X	X
Cadmium	X			
Aldrin	X	X	X	
Chlordane	X	X	X	X
Chlordecone	[X]			
Short-chain Chlorinated Paraffins	[X]			
DDT (+DDD+DDE)	X	X	X	X
Dieldrin	X	X	X	
Endosulfan	[X]			
Endrin	X	X		
Heptachlor	[X]	X		
Hexabromobiphenyl	X			
Hexachlorobenzene	X	X	X	
Lindane	[X]			
Mirex	X	X	X	
Octachlorostyrene			X	
PAHs	X		X	
PCBs	X	X	X	X
PCDDs (dioxins)	X	X	X	
PCDFs (furans)	X	X	X	
Pentachlorophenol	X			
Quintozene (pentachloronitrobenzene)	[X]			
Toxaphene	X	X	X	

[1] *Decisions have not yet been taken on whether to include these substances in the initial POPs Protocol.*

envisaged that AMAP should, as far as possible, be built upon existing national and international programs, which were to be developed into a circumpolar framework. Initial priority was given on a circumpolar basis to persistent organic pollutants, selected heavy metals and radionuclides, and on a regional basis (Fennoscandinavia) to acidification.

To implement AMAP, a Working Group was established comprising delegates from each of the participating nations (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden, United States), observing countries (Germany, Netherlands, United Kingdom), and Indigenous Peoples Organizations (Inuit Circumpolar Conference, Nordic Saami Council, Russian Association of Peoples of the North). A number of international organizations (e.g. United Nations Environment Programme, United Nations Economic Commission for Europe, World Wide Fund for Nature, International Union for Circumpolar Health) are also observers. Canada is presently the Chair of the Working Group and Sweden is Vice-chair. Norway has provided a Secretariat, based in Oslo. Work is underway to consider the possible role of AMAP in its second phase under the new Arctic Council. It is not anticipated that detailed proposals will be developed until the spring of 1997.

The Monitoring Programme for AMAP was completed in 1993, providing direction for monitoring activities for the Atmosphere, the Marine, Freshwater and Terrestrial environments, and Human Health. The requirements of the Monitoring Programme are fulfilled by the sum of the member countries' Implementation Plans. Canada's Implementation Plan has largely comprised projects being conducted under the AES-NCP.

Output from AMAP will take the form of comprehensive "State of the Arctic Environment" assessment reports for the eight circumpolar Ministers, the first of which will be presented to Ministers in June 1997. This assessment will have two products: the Arctic Assessment Report (AAR), a fully referenced, comprehensive technical and scientifically presented assessment of all validated data available on the status of the Arctic environment relative to the AMAP mandate; and the State of the Arctic Environment Report (SOAER). The latter, based on the AAR, will be more concise, presenting the results of AMAP and its assessment. It will be prefaced by an executive summary with recommendations specifically addressed to Ministers.

5.4.2.3 Canada/Russia Program on Scientific and Technical Cooperation in the Arctic and the North

In October 1991, an agreement was reached between Canada and Russia for cooperation on contaminants in the Arctic. A work plan, which ultimately proved to be overly ambitious given the present period of economic transition in Russia, was established and two major projects were achieved. First, an air monitoring station was established at Dunay Island in the Russian Arctic that is a replicate of the three existing stations in the Canadian Arctic. This new station has contributed to efforts to set up a circumpolar network for air monitoring. The second achievement was the exchange of environmental samples between Russian and Canadian laboratories for analytical intercalibration purposes. Future efforts will be directed toward establishing a proposed second Russian Arctic air monitoring station on the coast north of the Ural mountains and carrying out further analytical intercalibration exchanges with laboratories in Murmansk (Academy of Sciences), St. Petersburg (Arctic and Antarctica Research Institute), Moscow ("Typhoon" Laboratories, and the Institute of Global Climate and Ecology), and possibly elsewhere. In addition, Canada will continue to work with the other Arctic nations to assist Russia in meeting its AEPS/AMAP responsibilities.

5.4.2.4 NAFTA Commission for Environmental Cooperation

In October 1995, Environment Ministers from Canada, the US and Mexico signed the Resolution on the Sound Management of Chemicals, under the auspices of the NAFTA Commission for Environmental Cooperation. Country-specific action plans for PCBs, DDT, chlordane and mercury (all of which have been shown by NCP research to be concerns in the Arctic) were submitted for approval in late 1996. Criteria are being developed to select additional substances for action in the future.

5.4.2.5 Binational Virtual Elimination Strategy

On the regional level, Canada is working with the United States to develop the Canada-US Strategy for the Elimination of Persistent Toxic Substances in the Great Lakes Basin, which will provide for a coordinated program for the virtual elimination of specific persistent, bioaccumulative and toxic substances between 1996 and 2005. The long-range transport of pollutants into the basin is a major issue that will be addressed under the strategy. The strategy is expected to be completed by fall/winter 1996/97.

5.4.2.6 United Nations Economic Commission for Europe (UN ECE) Convention on Long-Range Transboundary Air Pollution (LRTAP) Preparatory Working Groups on Persistent Organic Pollutants (POPs) and Heavy Metals

A task force on persistent organic pollutants (POPs) led by Canada and Sweden was established in 1991 under the UN ECE Convention on Long-range Transboundary Air Pollution (LRTAP). In April 1994, the task force completed its work and concluded that the documentation that it had compiled provided adequate substantiation to support the preparation of a POP protocol under the Convention. In December 1994, the Executive Body to the Convention took note of these findings and established a new "Preparatory Working Group on POPs" with a mandate to assist the Working Group on Strategies with the preparation of a POP protocol.

On a parallel track with the UN ECE POPs efforts has been a similar initiative for heavy metals. A task force on heavy metals, led by the Czech Republic, was established in 1989 under the UN ECE LRTAP Convention. The task force completed its report in February 1994, concluding that there is the need for a protocol to control the long-range transport and deposition of heavy metals. The UN ECE Working Groups on Technology and on Effects met in July 1994 and noted concerns related in particular to the significance of natural sources and forwarded the Task Force Report to the Executive Body. In December 1994, the Executive Body to the Convention established a new "Preparatory Working Group on Heavy Metals" with a mandate to assist the Working Group on Strategies with the preparation of a heavy metals protocol.

The Preparatory Working Groups are preparing draft text for negotiation. A complete composite negotiating text for a POPs protocol was prepared in late 1996, with negotiations commencing in Geneva in January 1997. All of the POPs of concern in the Arctic are on the UN ECE list of 16–18 priority substances. Negotiations are targeted for completion by late 1997 or early 1998. Table 5.1 indicates substances to be addressed under the protocol.

5.4.2.7 United Nations Commission on Sustainable Development (UN CSD), International Experts Meeting on Persistent Organic Pollutants: Towards Global Action

At the spring 1994 session in New York of the United Nations Commission on Sustainable Development (UN CSD), Canada offered to host a meeting of world experts in the field of persistent organic pollutants to consider how all nations can work together to solve the problems associated with these substances. This meeting was hosted jointly by Canada and the Republic of the Philippines, thus enabling the perspectives of both the developed and the developing worlds to be considered in the experts' deliberations. Held in Vancouver, BC, June 4–8, 1995, this meeting had the following objectives:

- highlight any significant concerns posed to human and environmental health by certain persistent organic pollutants
- identify and analyse the technical, institutional, social, economic, financial and management issues inherent to certain persistent organic pollutants, and
- identify key elements for action to manage concerns posed by certain persistent organic pollutants.

At the conclusion of the meeting, participants agreed that enough evidence exists on the adverse human health and environmental impacts of POPs to substantiate the need for immediate action. This action should be taken at the national, regional and international levels, and it should include bans and phase-outs for certain POPs. It was agreed that existing international laws and agreements do not provide for effective global POPs management, and that domestic regulatory arrangements alone would not be adequate. It was also noted that action would only be effective if it takes into account the significant social, economic, political and cultural differences throughout the world. The need for development of effective, affordable and accessible alternatives was emphasized and many recommendations were made regarding, for example, donor and development agency policies, trade and economic strategies, and bans on the use of POPs in agriculture.

5.4.2.8 United Nations Environment Programme Global Action Plan

In May 1995, the 18th session of the Governing Council to the United Nations Environment Programme (UNEP) agreed to invite "the Inter-Organizational Programme for the Sound Management of Chemicals (IOMC), working with the International Programme on Chemical Safety (IPCS), and the Intergovernmental Forum on Chemical Safety (IFCS) to initiate an expeditious "global assessment process" concerning POPs. The assessment was conducted by an *ad hoc* Working Group that met in Canberra, Australia in March 1996, and in Manila, the Philippines in June 1996. The focus was on a short-list of 12 POPs derived from work conducted under the LRTAP Convention initiative. Based on the results of this process and the Declaration from the (October 1995) Washington

Intergovernmental Meeting on Marine Pollution From Land-Based Activities (at which countries expressed their intent to develop global legally binding actions on POPs), the IFCS *Ad Hoc* Working Group has developed a comprehensive set of recommendations for consideration in 1997 by UNEP Governing Council and the World Health Assembly (WHA). These include recommendations that Governing Council and the WHA decide that immediate action on the 12 specified POPs should be initiated to protect human health, that action should include expeditious development of a global legally binding instrument, and that an International Negotiating Committee be convened to prepare that instrument.

5.5 Communications Materials

5.5.1 Overviews, Technical Reports and Summaries

Contaminants in Northern Canada, an overview booklet of the AES-NCP. 1995. Métis Nation-NWT.

Contaminants and Your Health: NWT Fact Sheets Resource Book. 1995. GNWT Health and Social Services.

Communicating about Contaminants in Country Food: The Experience in Aboriginal Communities. 1995. Available as a full report and as a bilingual (English/Inuktitut) plain language summary. Inuit Tapirisat Canada.

Inuit Perspectives on Environmental Contaminants. Report on Avativut/Ilusivut Risk Management Workshops in Nunavik and Labrador (1995): available at the Institute for Risk Research.

Contaminants in the Yukon: 1997. (in press). Contact Yukon Contaminants Committee.

5.5.1.1 Videos

Environmental Contaminants in the North. 1993. GNWT Health and Social Services.

Environmental Contaminants and Your Health. 1995. GNWT Health and Social Services.

The Arctic Environmental Strategy. 1993/94. Council of Yukon First Nations.

The Northern Food Dilemma. 1996. Nunavik Food Contaminants and Health Committee. Available in English and Inuktitut. Contact Suzanne Bruneau.

Traditional Food: Is it Safe? 1996. CINE/CYFN.

5.5.1.2 Newsletters, Fact Sheets, Curricula, Public Service Announcements

Graphics package of contaminants icons and illustrations. Métis Nation-NWT.

Lesson plans for junior high, high school and Adult Education. 1995/96. Métis Nation-NWT.

Fact sheets on toxaphene, PCB, DDT. 1993-1996. Yukon Contaminants Committee.

CINE's Newsletter. CINE.

As Long as This Land Shall Last. Denendeh Environmental Newsletter.

Nutrition fact sheets for Dene traditional foods. 1996. Dene Nation

Nutrition fact sheets for Inuit traditional foods. 1996. GNWT Health and Social Services

The Avativut/Ilusivut Newsletter published quarterly. Editor: Susie Bernier.

Public Service Announcements. 1996. Nunavik Food, Contaminants and Health Committee.

1) Regional food contributes to a healthy pregnancy and a healthy baby;
2) Breast-feeding is the best thing for the health of the baby;
3) Family sharing in food harvesting activities: passing on essential knowledge;
4) Teenagers and regional food: counteracting the lure of fast food.

5.5.2 Electronic Information

5.5.2.1 Métis Nation — NWT Northern Contaminants Program Database

This data base provides useful information regarding northern contaminants projects to teachers, students, community environmental committees, researchers, health professionals, Aboriginal organizations, etc. The data base can cross-reference NCP information with community profile data, school boards, schools, colleges, community learning centres and wildlife distribution information.

5.5.2.2 Northern Aquatic Food Chain Contaminants Database (NAFCC)

Contains results of contaminant levels from analyses completed up to and including April 1995 for organisms in the marine aquatic food chain across the Arctic. Based on published data as well as AES-NCP researcher unpublished data.

5.5.2.3 Northern Information Network (NIN)

Consists of a Directory and a Bulletin Board Service (BBS). The **Directory** contains information about Northern Canada. Details are provided about the ownership of data bases, categories of information, keywords, size of the data bases, contacts and certain technical information. One category provides additional descriptive material that is specific to contaminants (e.g. medium, sampling and storage methods, analytical techniques). For on-line access to the Directory dial (819) 997-0840. Upon connection, the password is **USER.**

5.5.2.4 NIN Bulletin Board System

Provides modem access (offering a 1-800 number, enabling access to users at no additional cost) to an electronic bulletin board upon which information on contaminants can be posted and exchanged. Examples of posted information are: GNWT nutrition fact sheets, overview booklet on Contaminants in Northern Canada. The BBS allows you to send and receive messages, download the NIN Directory and periodic updates, as well as the directory of federal government data bases. User manuals, help and information files, AES reports, announcements and documents, including documents about work on contaminants can be accessed.

5.5.3 Contacts

For further contaminants information contact the following organizations:

Avativut/Ilusivut: Susie Bernier, Quebec Public Health Centre (tel: (418) 666-7000 ext. 408, fax: (418) 666 2776, e-mail: sbernier@cspq.qc.ca).

Institute for Risk Research: (tel: (519) 888-4567 ext. 3355, fax: (519) 725-4834).

CHUL: Suzanne Bruneau (tel: (418) 666-7000 ext. 292, fax: (418), 666-2776, e-mail: sbruneau@cspq.qc.ca).

CINE: Macdonald Campus, McGill University, 21-111 Lakeshore Road, Ste-Anne-de-Bellevue, Québec, H9X 3V9. (tel: (514) 398-7544, fax: (514) 398-1020).

Council For Yukon First Nations: Norma Kassi, 22 Nisutlin Drive, Whitehorse, Yukon. (tel: (403) 667-7631, fax: (403) 668-6577).

Dene Nation: Environment Office, Box 2338, Yellowknife, NWT, X1A 2P7. (tel: (403) 873-4081, fax: (403) 920-2254, e-mail: dene-nation@ssimicro.com).

GNWT Health and Social Services: Jody Walker, Department of Health, Box 1320, Yellowknife, NWT, X1A 2L9. (tel: (403) 920-8868, fax (403) 873-7706).

Inuit Tapirisat Canada: Research Department. 170 Laurier Ave. West, Ottawa, Ontario, K1P 5V5. (tel: (613) 238-8181, fax: (613) 234-1991).

Métis Nation-NWT: Bill Carpenter, Box 1375, Yellowknife, NWT, X1A 2P1. (tel: (403) 873-3505, fax: (403) 873-3395).

NIN Administrator: Room 612A, Natural Resources and Environment Branch, Department of Indian Affairs and Northern Development, Ottawa, Ontario, K1A 0H4 (tel: (819) 997-9480, fax: (819) 953-2590, e-mail: ninman@INAC.gc.ca).

NIN-BBS access: modem connections 8-N-1, dial: (819) 994-2557 in the Ottawa/Hull area, (800) 567-6935 in other parts of Canada. **Internet access:** http://www.inac.gc.ca. **Telnet access** is also possible through nin.inac.gc.ca To upload or download files, use **ftp**. The command is open nin.inac.gc.ca.

Yukon Contaminants Committee: c/o Mark Palmer, DIAND, 345–300 Main Street, Whitehorse, Yukon, Y1A 2B5, (tel: (403) 667-3272, fax: (403) 667-3206).

Chapter 6: Conclusions

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Assessment Report, Indian and
Northern Affairs Canada, Ottawa 1997.

Citation:

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CHAPTER 6 Conclusions

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6.1 Introduction

The contaminants of concern in the Canadian Arctic include persistent organic pollutants (POPs) such as the organochlorine (OC) pesticides, polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), and the heavy metals mercury, cadmium, and lead, and radionuclides. These contaminants are present in the air, water, sediment, soil, snow, plants, animals, and humans of the Canadian Arctic. Many of these compounds bioaccumulate and biomagnify. In more temperate ecosystems, toxicological effects have been detected as a result of the presence of many of these compounds, raising concern for the health of the Arctic ecosystem and its peoples. Northern residents, especially Aboriginal Northerners, are susceptible to the exposure and potential adverse effects of contaminants due to a dietary dependence on arctic fish and wildlife (country or traditional foods).

The research efforts of the Arctic Environmental Strategy Northern Contaminants Program (AES-NCP) have focused on the sources, pathways, fate, and distribution of contaminants in the north, with an emphasis on animals and plants consumed as traditional foods. This information provides scientists, government agencies, and northern communities with a knowledge base to address broad issues of concern regarding the presence of contaminants in the Canadian Arctic and the potential human health implications.

This chapter summarizes the major conclusions based on the scientific results presented in Chapters 2, 3, and 4. The discussion attempts to synthesize the conclusions from the various components of the AES-NCP (sources, occurrence, and pathways, ecosystem effects, and human health effects) into a cohesive, general discussion that outlines the current state of knowledge regarding contaminants in the Canadian Arctic. The discussion is intended to be as holistic as possible by presenting the information in an interdisciplinary fashion, when appropriate, to corroborate the conclusions from these three chapters. Throughout the discussion, the implications for Arctic residents, and in particular Aboriginal Northerners, who consume traditionally harvested food, is emphasized.

6.2 Contaminants in the Canadian Arctic

6.2.1 Human Health

A diet based on traditional foods has many nutritional, socio-cultural, and economic benefits that have been discussed in detail in Chapter 4. A decrease in the use of traditional foods has been documented for some Canadian Aboriginal Northerners for a variety of reasons, especially among younger generations (Kuhnlein *et al.* 1995a, Morrison *et al.* 1995, Wein 1995) A shift from the consumption of traditionally harvested foods to market foods would generally result in the loss of many of the nutritional and health benefits associated with a traditional diet. For example, a traditional Aboriginal Northerner's diet is high in protein and zinc, moderately high in fat, and low in carbohydrates. The consumption of meats and local wild greens and berries provides the necessary dietary intake of vitamins and minerals. In particular, fish and game contribute substantial amounts of vitamin B_{12} and panthothenic acid to the diet. Although the total intake of these vitamins by northern Aboriginals is probably higher than that of the general Canadian public, a few studies indicate that vitamin A and C intakes may be below recommended levels. In addition, marine mammals and fish are rich sources of omega-3-fatty acids, which are thought to protect against cardiovascular disease. Compared with a traditional diet, a diet based on market foods provides less protein, iron and zinc, more fat (especially saturated fat), and a much higher carbohydrate intake.

Dietary shifts are also associated with "lifestyle" changes; less time spent hunting results in a more sedentary lifestyle. Poor diet and other lifestyle changes are considered to have contributed to the observed increase in the incidence of certain diseases, such as diabetes and cardiovascular disease, among northern Aboriginals (Lawn and Langner 1994, Kuhnlein 1995). Historically, the low incidence of these diseases among the Inuit has been attributed to a dependence on a traditional lifestyle, including the consumption of traditional foods. A change in diet can result in a loss of traditional knowledge, as hunting and food preparation is an integral part of Aboriginal Northerners' culture. Harvesting, sharing, processing, and the consumption of traditional food all contribute to the strong bond among Aboriginal communities, which forms the basis of cultural identity. The harvesting of traditional food provides an opportunity to teach humility, spirituality, and responsibility for the environment. A degraded environment is considered a threat to health.

For Aboriginal Northerners, the risk associated with a shift in dietary preference balanced with the risk associated with the presence of contaminants in arctic wildlife consumed as traditional food must be assessed. At present, the greatest risk associated with the consumption of traditionally harvested food is exposure to certain OCs, especially chlordane, toxaphene, and PCBs, and exposure to the heavy metals, cadmium and mercury. In certain instances, concern for exposure to these particular contaminants has led to human health risk assessments and decisions based on risk management (Appendix III).

For example, in the Yukon Territory, public health advisories were issued on Lake Laberge in 1991 in response to observed elevated toxaphene levels in lake trout muscle and burbot liver. However, other species of fish (e.g., whitefish), from this lake and all fish from nearby lakes are safe for consumption. Similarly, in the NWT, a health advisory was issued in the Fort Smith/Slave River area for the consumption of burbot liver, due to elevated levels of toxaphene. Furthermore, concern for the level of human exposure to cadmium through the consumption of caribou liver and kidney resulted in an advisory to limit the consumption of caribou kidney from specific NWT herds. Actual consumption levels of this tissue, however, rarely exceed the recommended maximum consumption levels. In fact, smoking is the most important route of exposure to cadmium in the Arctic, and elsewhere in the world. Caribou meat (i.e., the muscle), has low levels of contaminants and is safe for consumption.

Risk assessment and the resulting risk management in the north does not always result in the recommendation of restrictive consumption guidelines for country foods. For example, samples of several species of fish and marine mammals collected over several years from the NWT and Yukon were submitted for a health risk assessment. Recommended maximum weekly intakes for toxaphene, chlordane, PCBs, dieldrin, chlorobenzene, cadmium, and mercury were made available by Health Canada as part of the risk assessment. However, because the benefits of a traditional diet were well known and documented, whereas information about the human health risks posed by some of the contaminants measured (especially toxaphene and chlordane) was, and remains, relatively uncertain, the health risk management decision was such that it was unnecessary to issue restrictive consumption guidelines. This decision was reached after a review of the health risk assessment results by a number of organizations, including representatives of regional Inuit organizations in the NWT, Labrador, and northern Québec, Health Canada, the Centre for Indigenous Peoples Nutrition and Environment (CINE), the GNWT Health Department, and other government agencies.

For organic contaminants, the human health risks associated with exposure to these compounds through country foods is uncertain both for the Arctic and for other regions (e.g., the Great Lakes basin), where they are present (Swain 1991). The few health studies that have been conducted suggest the greatest health concern is for the fetus, through transplacental exposure, and for newborns through breast-feeding (Dewailly *et al.* 1993a,b, Fein *et al.* 1984, Jacobson *et al.* 1990, Rogan and Gladen 1991). In the Canadian Arctic, cord-blood studies have been conducted in the Mackenzie-Kitikmeot region, Nunavik Broughton Island region, and in Greenland to assess transplacental exposure to contaminants. The implications for human health, based on these initiatives, have yet to be determined. Relatively high levels of organochlorines, mainly PCBs, have been measured in the cord-blood of newborns from some communities in the eastern Arctic. These levels are attributed to a greater dietary dependence on marine mammals, which tend to accumulate organochlorines, and are discussed later in this chapter in the context of spatial trends. Recent research associates dioxin, furan, and PCB levels in human milk, and subtle clinical, immunological and neurodevelopmental alterations in a group of infants/children from the Netherlands (Koopman-Esseboom *et al.* 1994a,b). This research is supported by a preliminary study of Canadian Inuit, which found an association between increasing PCB, dioxin and furan levels in human milk and immune system deficits in infants (Dewailly *et al.* 1993c). In addition, very little is known about the intergenerational effects associated with long-term exposure to these contaminants.

In the Canadian Arctic, concern regarding the level of human exposure to lead, cadmium, and mercury depends on the metal. For example, lead does not appear to be a heavy metal of concern in Arctic Canada, as levels in children's blood in the north are as low as those found in southern populations. Northern residents may be exposed to cadmium through the consumption of certain traditional foods, such as organ meats (liver and kidney). However, the major route of exposure to cadmium in the north is cigarette smoking. People at the highest risk of exposure to cadmium are smokers, the elderly, and diabetics. High cadmium levels in smokers may have irreversible effects on kidney function.

Human exposure to mercury is of particular concern because methylmercury, the organic form, is bioaccumulative, biomagnifies, and is a potent neurotoxin. Humans are exposed to methylmercury in the Arctic through food consumption, especially fish and marine mammals. At present, there are no adverse human health effects that are directly attributed to exposure to methylmercury in the Canadian Arctic. This does not, however, mean that there have been no effects. Isolating the specific effects of a particular compound, such as methylmercury, is extremely challenging due to the many confounding factors, such as the presence of other contaminants and socio-economic conditions. The complexity of this issue is exemplified in other Aboriginal communities, for example two Ojibwa communities on the English-Wabigoon River system in northern Ontario, where levels of methylmercury have been elevated in human tissues. Despite measured levels, there was no provable clinical evidence of methylmercury poisoning (Wheatley and Paradis 1995). Methylmercury, in addition to other factors, however, has likely collectively contributed to past and present socio-cultural problems within these communities. Additional factors that may contribute to the lack of observed clinical effects in the north include mercury in human tissues at levels only just into the range of concentrations at which subtle symptoms or signs of poisoning may be detected (WHO 1990, Wheatley and Paradis 1996) and the hypothesized, albeit controversial, protection afforded by selenium against mercury toxicity through co-exposure to selenium and mercury (Endo *et al.* 1977, Marier and Jaworski 1983, Magos 1991).

The amount of radionuclides in atmospheric fallout in the Canadian Arctic is low, and similar to or lower than levels found in the temperate environment. In the Canadian Arctic, human body burdens of cesium, the anthropogenic radionuclide of greatest concern, have declined over the last 30 years from approximately 450 $Bq{\cdot}kg^{-1}$ to 10 $Bq{\cdot}kg^{-1}$. This decline is attributed to the cessation of above ground nuclear weapons testing. Today, people who live in the north receive more than 95% of their radiation dose from naturally occurring radionuclides. Due to bioconcentration in the food chain, people who consume large amounts of traditional foods may be exposed to higher levels of some radionuclides, primarily the naturally occurring radionuclides ^{210}Pb and ^{210}Po. This increased radiation may result in a 10% increased risk of cancer. However, there is a large amount of uncertainty associated with the calculation of these cancer risks, as they are based on extrapolations from high level exposure of nuclear bomb survivors to low level environmental exposures.

In addition to a concern regarding human exposure to contaminants through the consumption of traditional foods, the presence of elevated levels of some POPs in the milk of Inuit women from northern Québec has raised justifiable concern among nursing mothers in the Arctic (Dewailly *et al.* 1989). Substantive health benefits are associated with breast-feeding, such as mother-child bonding, immunological benefits transferred from mother to child, nutritional values and reduced risk of bacterial contamination from poorly prepared formulas. The consequences of abandoning breast-feeding, given the lack of affordable alternatives for nursing women in the north, present a far greater risk to health than the few known and many unknown risks associated with infant exposure to contaminants through human milk (Thomas and Colborn 1992, Dewailly *et al.* 1991).

Determining adverse human health effects due to the presence of contaminants in traditional foods and human milk is extremely difficult as many additional confounding factors contribute to the health of an individual. These include, but are not limited to, the qualitative impacts of being informed of contaminants present in traditionally harvested foods, and several socio-economic factors, such as smoking and alcohol and substance abuse. In fact, Arctic residents are at a significantly higher risk of exposure to cadmium, and its effects, through cigarette smoking than from dietary sources (Benedetti *et al.* 1990). Smoking is also a major risk factor for both coronary heart disease and lung cancer. Such factors cannot be ignored in an assessment of the relative risk of exposure to contaminants through food.

Risk determination for the consumption of traditional foods considers the potential risks from exposure to contaminants, the risks associated with abandoning a diet based on traditional foods, and the known nutritional, social, and economic benefits of a traditional diet. Risk management decisions and organizational structures for the dissemination of information to communities must be developed among communities to reduce exposure to contaminants without sacrificing the many benefits of a diet based on traditional foods. In the short term, this may require reducing the consumption of certain organs or tissues of some animals known to accumulate high levels of contaminants (e.g., caribou liver). A reduction in the consumption of certain foods in the diet, however, must coincide with an increase in consumption of alternative, highly nutritious, locally available foods. In the long term, there is a great need to limit the transport of contaminants to the Canadian Arctic through a reduction in their global manufacture and use.

6.2.2 Ecosystem Health

Evidence for ecosystem effects in the Arctic due to the presence of contaminants is currently derived from several sources including: i) studies of indicator species, such as mink and peregrine falcon; ii) comparisons of levels in Arctic wildlife with the same or similar species in locations where effects have been observed and; iii) studies of biological responses based on biochemical indicators, for example mixed-function oxygenase and metallothionein.

In southern latitudes, certain species such as mink and the peregrine falcon, are effective bioindicators of contamination due to their relative sensitivity to the presence of organochlorines. Mink from the Great Lakes region that consumed fish with elevated tissue contaminant concentrations have suffered adverse reproductive effects (Great Lakes Science Advisory Board 1991, Wren 1991). To date, no such adverse effects have been reported in Arctic mink populations, which generally have lower contaminant levels than other North American mink populations (Poole *et al.* 1995). Populations of tundra peregrine falcons did not suffer the extreme population declines due to DDT experienced by southern populations in the 1960s (Ratcliffe 1967, Peakall 1990). However, eggshell thinning is currently evident. Because peregrine falcons are migratory, the birds are exposed to organochlorines in over-wintering habitats and the contribution to exposure from the Arctic is difficult to determine. However, levels of organochlorines in the eggs, blood plasma, tissue, and the prey species of the tundra peregrine falcon population nesting at Rankin Inlet are within the range that will result in failure of a portion of the reproductive attempts each year.

Research has been conducted under the NCP to identify appropriate biological responses of ecosystem stress in the Arctic (Norstrom 1994, Lockhart and Ferguson 1994, Payne 1994, Wagemann 1994). Biochemical indicators of a biological response, such as mixed-function oxygenase and metallothionein, measure chemical changes at the cellular level caused by the presence of toxic chemicals. The induction of enzymes can be used as an indicator of exposure to contaminants, although the results may not be readily interpreted ecologically. There has been a strong positive correlation of the activity of the cytochrome enzyme, CYP2B, with PCBs and chlordane in polar bears (Norstrom 1996). Similar positive correlations between enzyme activity and OC concentrations have been observed in beluga that were starving due to ice entrapment. Other studies on fish and ringed seals have not shown strong correlations between mixed-function oxygenase activity and PCB levels; nor have they shown the presence of effects related to toxaphene exposure. At present, there is a lack of dose-response data for Arctic animals that limits the ability to interpret the biological significance of the concentrations of contaminants in Arctic wildlife.

Determining effects of contaminants on populations of Arctic biota is difficult in the absence of 'healthy' comparative population data. The baseline population data for many Arctic animals including polar bears, seals, and sea birds, is only available for approximately 20 years, over which time contaminant tissue levels have been relatively constant. Therefore, with the possible exception of the peregrine falcon, the effects of organochlorines on the health of Arctic wildlife, in particular marine mammals, at the individual or population level is inconclusive.

Elevated levels of contaminants in polluted areas in southern Canada, for example the Great Lakes and the St. Lawrence River, have been associated with adverse effects in biota, such as impaired development in prenatal and juvenile life-stages, higher incidences of cancers and mutations, disruption of immune and endocrine systems, and a decrease in resistance to diseases (Colborn *et al.* 1993, Great Lakes Science Advisory Board 1991, Government of Canada 1991). Similarly, observations of the prevalence of skull-bone lesions in seals from the Baltic Sea have been attributed to the presence of industrial chemicals, including OCs (Bergman *et al.* 1992, Mortensen *et al.* 1992). In Arctic marine mammals, concentrations of certain OCs, PCBs and DDT, are 10 to 20 times lower than in the same or similar marine mammal species where such detailed pathological examinations have been conducted.

Immunosuppression is considered to be the most sensitive toxic effect of organochlorines, especially PCBs and TCDD. In Arctic ecosystems, there is a need to link measured biological effects, such as immunosuppression and endocrine disruption in polar bear, beluga, and narwhal with studies on population dynamics. Mean concentrations of TCDD toxic equivalent (TEQ) concentrations in polar bear liver and in walrus blubber from northern Québec are at or above the threshold found for immunosuppression in North Sea harbour seals. However, concentrations of TCDD TEQs in Arctic ringed seal or beluga blubber are 3 to 5 times lower than those associated with impaired immune function (depressed leucocyte activity) in harbour seals. The effects of immunosuppression in Arctic wildlife may only be obvious during times of additional stress, such as exposure to a pathogen, or during periods of starvation when fat reserves are depleted. In the Canadian Arctic the polar bear is probably at greatest risk from exposure to organochlorines due to the consumption of ringed seal tissue. Levels of TCDD TEQ concentrations in the tissue of ringed seal from the Canadian Arctic are at or near Canadian and United States tissue-guideline values established to protect fish-eating wildlife.

Caribou, beluga, and narwhal from the Canadian Arctic are exposed to high levels of heavy metals, especially cadmium. The potential effects, however, are inconclusive. Cadmium in narwhal liver was found to be predominantly associated with metallothionein (a metal-binding protein), suggesting it was in a relatively non-toxic form. Arctic biota may have adapted to relatively high exposure levels to metals and radionuclides, which occur naturally in the Arctic.

6.3 Spatial and Temporal Trends

Much of the information amassed under the NCP over the last six years has focused on determining and comparing contaminant levels in terrestrial, marine, and freshwater ecosystems. Spatial trends of contaminants afford geographic comparisons of the levels of contaminants in wildlife consumed as traditional food. Such comparisons are useful in identifying particular biota or regions that have elevated contaminant levels that may be of concern. They can also be used to support explanations regarding both the natural and anthropogenic, local and long-range, sources of contaminants in the Canadian Arctic. Temporal trends complement this information. Temporal trend data from abiotic media (e.g., the air, glacial ice, and sediment) provide a historical record of the deposition of atmospheric contaminants in the Canadian Arctic. This information, coupled with levels measured in biota over time, is critical in assessing whether contaminant levels in the Canadian Arctic are increasing, decreasing, or remaining the same. Temporal trends will be a useful measure of the success of efforts to mitigate the transport of contaminants originating from sources external to the Canadian Arctic.

6.3.1 Spatial Trends

Differences exist between levels of exposure to contaminants among residents of eastern and western Arctic communities due to variations in the composition of traditional diets and the proportion of the total diet that is based on traditional food. For example, the diet of Inuit communities in the eastern arctic consists mostly of marine mammals. In contrast, Aboriginal peoples of the Mackenzie Delta mainly harvest terrestrial wildlife, particularly large mammals such as caribou and moose, which constitutes a comparatively smaller proportion of their total diet than the traditionally harvested foods consumed by the Inuit (Kuhnlein *et al.* 1995b). Fish is a seasonal staple across the arctic (Coad 1994). Organochlorine levels are generally higher in marine wildlife than in terrestrial wildlife because marine mammals feed at a higher level in the food web and because they tend to accumulate larger fat reserves. In contrast, OCs do not bioaccumulate to the same degree in terrestrial food webs, which is partly due to shorter terrestrial food webs (the air-lichen-caribou food chain) and partly due to relatively smaller fat reserves in terrestrial animals. Consequently, estimated average intake levels of organochlorines are higher in communities that consume marine mammals than in communities that consume terrestrial mammals (Kuhnlein 1995, Kuhnlein *et al.* 1995c). For example, the daily contaminant intake of several OCs by Baffin Island Inuit was higher, by an order of magnitude, than the daily contaminant intake among the Sahtu Dene/Métis (see Chapter 4).

Few estimates of the average daily intake levels of contaminants exist for northern Aboriginal communities. Differences in daily intake levels of traditional foods can vary considerably among communities and depend on several factors, including the proportion of the diet based on traditional food, the preferred wildlife and edible tissue, seasonality of consumption, and age and gender of the consumer (Chan *et al.* 1995, Kuhnlein *et al.* 1995b, Wein 1995). For example, the consumption of traditional foods ranged from 27 kg·year^{-1} in the Wood Buffalo Park area (Dene/Métis) to 292 kg·year^{-1} at Sanikiluaq (Inuit). In addition, levels of contaminants in Arctic biota are variable and dependent on several biological factors, such as species, sex, and age, and on the physicochemical parameters of the pollutant, for example bioavailability and lipophilicity.

The results of the spatial differences in the consumption of different types of traditional foods are reflected in the levels of contaminants, in particular organochlorines, in human tissues samples. Cord-blood studies across the Canadian Arctic indicate that those people who rely on marine mammals as traditional food tend to have higher levels of organochlorines, particularly PCBs, than people who rely on other types of traditional food. Similarly, PCB levels in the cord-blood of people who consume marine mammals are higher than levels in people who do not rely on traditional foods as a significant part of their diet, that is non-aboriginal people who live in both the north and in the south.

The spatial distribution of contaminants in Arctic biota provides additional information on areas or regions where differences in human exposure levels to contaminants might be expected. For example, results from work conducted under the NCP on both the levels of the heavy metals mercury and cadmium, in the tissues of marine and terrestrial mammals and on the levels of mercury and toxaphene in freshwater fish are illustrative of this point.

Among Arctic communities with a similar dietary dependence on marine mammals, differences in exposure to the heavy metals mercury and cadmium might be expected due to west-east differences in the levels observed in marine mammal tissues. For example, polar bears, ringed seals, and beluga from areas bordering the Beaufort Sea all have higher mercury levels than eastern Arctic animals. Mercury levels in the liver of ringed seals ranged from approximately 5 to 60 mg·kg^{-1} in the west and from 5 to 10 mg·kg^{-1} in the eastern Arctic (values expressed on a wet weight basis). Similarly, levels of mercury in beluga liver ranged from 10 to 30 mg·kg^{-1} and 3 to 12 mg·kg^{-1} in the western and eastern Arctic, respectively (Chapter 3). In contrast to this, blood mercury levels among northern Aboriginals are higher in the north and east than in the west. This trend probably reflects the difference in traditional diet composition among Aboriginal Northerners. As previously discussed, the Sahtu Dene/Métis from the western Arctic consume fewer marine mammals than do the Inuit from the eastern Arctic.

In contrast to mercury levels, higher cadmium concentrations were observed in marine biota from the eastern Arctic than from the west. Cadmium levels in the liver of ringed seals ranged from approximately 3 to 7 mg·kg^{-1} in the west, and from 10 to 15 mg·kg^{-1} in the eastern Arctic (values expressed on a wet weight basis). Similarly, levels of cadmium in beluga liver ranged from 1 to 5 mg·kg^{-1} and from 5 to 10 mg·kg^{-1} in the western and eastern Arctic, respectively (Chapter 3). In general, levels of cadmium in the kidney and liver of Canadian Arctic marine mammals are similar to those of terrestrial animals, such as caribou and moose, and are as high as or higher than levels observed in similar species in more temperate areas (Wagemann *et al.* 1983, Dietz *et al.* 1995). Blood cadmium levels are not available for comparison between people from western and eastern Arctic communities.

As discussed in Chapter 4, marine mammal liver is not a large component of a traditional diet. However, relatively high levels of total mercury and methylmercury have been measured in the commonly consumed portions of marine mammals, such as the meat (muscle) and muktuk (Chan *et al.* 1995, Kuhnlein *et al.* 1995a). For example, the average total mercury concentration in muktuk of western Arctic beluga collected in 1993/94 was 0.77 mg·kg^{-1}, based on wet weight. The majority of muktuk samples collected in this period had mercury levels that exceeded the mercury guidelines for edible fish tissue of 0.2 mg·kg^{-1} for subsistence fishing and 0.5 mg·kg^{-1} for the commercial sale of fish. In muktuk, most of the methylmercury is concentrated in the skin, and not in the underlying blubber layer. As previously discussed in section 6.2.1, the risk management decision recommended no change in the amount of muktuk consumed by northern Aboriginals.

Many freshwater fish (e.g., lake trout) harvested by northern residents from specific lakes in the Arctic have mercury levels in edible tissues that exceed the mercury consumption guidelines (Chapter 3). However, there have not been any observed adverse effects associated with these elevated levels either in the fish or among humans. The presence of mercury in the Canadian Arctic is attributed to both industrial and natural sources (Wheatley and Wheatley 1988, Lockhart *et al.* 1995). In the Québec James Bay region, the edible tissue of several subsistence fish, such as whitefish and Northern pike, collected from recently created reservoirs have elevated levels of mercury. The reservoirs were created as result of the flooding caused by large dam projects, such as the Hydro-Québec James Bay project. Flooding results in a change in the biochemical mobilization of mercury due to the presence of decomposing submerged plants (a food source for microbes) that results in an increase in microbial mercury methylation, or biomethylation. The process of biomethylation can transform elemental mercury, naturally present, to the more toxic, bioavailable, organic forms (Jackson 1988). In this case, although the source of mercury to the fish is still natural, the observed elevated levels are a result of anthropogenic perturbations to the ecosystem.

Relatively high levels, that is 10 to 60 mg·kg^{-1}, of cadmium have been observed in the kidney and liver of caribou from the Yukon, NWT, and northern Québec (Crete *et al.* 1989). Higher cadmium levels, such as 20 to 190 mg·kg^{-1}, in caribou from the Tay and Finlayson woodland herds in southeastern Yukon may be the result of bioaccumulation of cadmium from local vegetation that has been exposed to natural soil zones of mineralization rich in cadmium.

In contrast to the observed spatial trends in heavy metals in the tissues of marine and terrestrial mammals, no clear spatial trends of the heavy metals cadmium and mercury were observed in waterfowl from the western and the eastern Arctic (Braune 1994a). Similarly, there were no clear spatial trends of POPs in sea birds (Braune 1994b). These results are possibly confounded by variations in the feeding habits of both waterfowl and sea birds, and due to differences in their overwintering habits and migration patterns.

Spatial trend information has been useful in identifying geographical areas or particular species harvested as traditional food that have contaminant levels that may be of concern. For example, toxaphene levels (the major organochlorine measured in freshwater fish in the Canadian Arctic) in the liver of burbot and the muscle of lake trout from Lake Laberge, Yukon, were as high as 2000 µg·kg^{-1} and 350 µg·kg^{-1}, respectively — 125 times higher than those found in similar species from other Yukon lakes (Kidd and Schindler 1994, Kidd *et al.* 1993, Muir and Lockhart 1993). The high concentrations of toxaphene in burbot liver may be a result of its high fat content. Higher toxaphene levels in burbot and lake trout have been attributed to differences in food web structure among the lakes. Lake trout from Lake Laberge, for example, feed at a higher trophic level than other lake trout, resulting in a greater degree of biomagnification of toxaphene. As previously discussed, public health advisories were issued on Lake Laberge in 1991 in response to elevated toxaphene levels in lake trout muscle and burbot liver. Other species of fish, such as whitefish, from Lake Laberge and all fish from nearby lakes are safe for consumption.

At present, the assessment of the geographic distribution of OC levels in Yukon freshwater fish is quite extensive. However, measurements of OCs in fish from parts of the NWT and the Nunavik region of northern Québec are limited mainly to Arctic char and are insufficient to assess spatial trends.

The major organochlorines present in marine biota are PCBs and toxaphene (Norstrom and Muir 1994). However, unlike the spatial variation observed for the levels of mercury and cadmium in marine mammals, the range in OC concentrations was generally similar among most marine mammals. One exception was the elevated OC levels in polar bears from M'Clure Strait, which is attributed to a different food web structure or some unknown feature of atmospheric or oceanic contaminant transport. Levels of PCBs in beluga and ringed seal blubber and polar bear fat were within two orders of magnitude and ranged from approximately 4 to 6 $mg{\cdot}kg^{-1}$, 1 to 2 $mg{\cdot}kg^{-1}$, and 1 to 70 $mg{\cdot}kg^{-1}$, respectively. Toxaphene levels in beluga blubber were similar to PCB levels and ranged from 4 to 6 $mg{\cdot}kg^{-1}$, while toxaphene levels in ringed seal blubber were slightly lower and ranged from 0.2 to 0.4 $mg{\cdot}kg^{-1}$.

Geographic comparisons among levels of organochlorines in ringed seal and beluga from the Canadian Arctic can be made with recent measurements on the same species from polluted areas. For example, levels of PCBs in female ringed seals from the Gulf of Bothnia (Baltic Sea) sampled in the late 1980s were, on average, 10 times higher than levels in ringed seals from the Canadian Arctic (Blomkvist *et al.* 1992). Similarly, levels of PCBs in male beluga from the St. Lawrence estuary (considered one of the most contaminated populations of beluga in the world), were approximately 12 times higher than male beluga sampled from southern Hudson Bay (Sanikiluaq). This latter difference has declined from a 25-fold difference, observed in the mid-1980s, mainly as a consequence of declining concentrations in St. Lawrence animals and due to slight increases in mean levels in the Hudson Bay animals.

Spatial trends in contaminants among abiotic environmental media, for example snow, air, and sediment, provide insight into the sources and quantities of contaminants transported to the Canadian Arctic. In general, levels of contaminants measured in snow and air are subject to large variations attributed to a combination of factors including: analytical methods, seasonality, and depositional trends.

Since snow comprises most of the wet deposition in the Arctic, work conducted under the AES-NCP has focused on the collection of snow from terrestrial, sea-ice, and lake surfaces (Gregor *et al.* 1995, Peters *et al.* 1995, Gregor 1993, Barrie 1992). Two types of snow measures are typically collected: snowpack and snowfall. The differences between these measures, and the contaminant levels associated with them, are discussed in more detail in Chapter 2. More than 30 snowpack and 9 snowfall sampling stations have been set up across the Canadian Arctic to provide information on the spatial distribution of POPs in snow. Results from the snowfall measurements indicate that HCB and PCBs are the most consistently distributed contaminants in snow samples, with mean concentrations of 0.16 $ng{\cdot}L^{-1}$ and 4.1 $ng{\cdot}L^{-1}$, respectively. A comparison of levels of contaminants in snowfall in the Arctic with levels in wetfall to Lake Superior, Ontario, indicated that for some contaminants, such as HCH, DDT, HCB and PCBs, the Arctic region is as polluted, with respect to snowfall, as regions to the south. These results indicate the importance of atmospheric transport in delivering contaminants to the Arctic.

Four high-volume air samplers at Alert (NWT), Tagish (Yukon), Cape Dorset (Baffin Island) and at the mouth of the Lena River (Russia) have been used to collect particulate and gaseous fractions of airborne OCs and PAHs since 1992. Concentrations of HCHs and PCBs in Arctic air exhibit seasonal fluctuations because of the effect of changes in air pressure and temperature on contaminant transport and deposition. Average concentrations of α-HCH, γ-HCH, and PCBs in the Canadian Arctic in the spring, during the period of peak concentrations, were approximately 85 $pg{\cdot}m^{-3}$, 50 $pg{\cdot}m^{-3}$, and 175 $pg{\cdot}m^{-3}$, respectively (although homologue sub-fractions of total PCBs vary seasonally). These air measures have provided useful insight into several inter-related physicochemical processes in the Arctic. For example, they have aided in interpreting snowpack and snowfall data, have linked atmospheric inputs to levels observed in freshwater sediments, and they have verified global chemical transport models as previously discussed in Chapter 2.

Spatial data for estimating airborne contaminant levels of heavy metals are limited. Therefore, no spatial trends for airborne metals in the Arctic currently exist. The data for airborne concentrations of mercury were collected from Alert only, and levels ranged from 0.67 to 2.8 $ng{\cdot}m^{-3}$, with a mean of 1.5 $ng{\cdot}m^{-3}$. These levels are less than concentrations measured at remote or reference areas in southern Ontario. Levels of other heavy metals such as lead, are discussed later in the context of temporal trends.

6.3.2 Temporal Trends

Limited temporal trend data exist for Canadian Arctic biota. The available information is confounded by a limited number of sampling dates, usually only three or less, advances in analytical techniques, and variations in the types of compounds measured. Tissue bank collections, for example collections with the Canadian Wildlife Service, Ontario and the Freshwater Institute, Manitoba, are one resource for determining historical temporal trends and the continued storage of biota from the Arctic in these tissue banks ensures that there will be a reliable source of data for future comparative studies. Levels of contaminants among abiotic media such as air, snow, water, and soil, demonstrate no consistent temporal pattern. Levels of some contaminants have increased, while other have decreased. In certain cases, changes in levels over time are attributed to changes (either increases or decreases) in source emissions. Future improvements in analytical technologies and an increase in the time periods for which contaminant information is available will aid in determining temporal trends in all media from the Arctic.

Twenty years of OC data are available to determine temporal trends in ringed seal blubber at Holman, NWT and in sea birds at Prince Leopold Island in Lancaster Sound. Both studies show a decline in OC levels from 1970 to 1980, followed by a more recent levelling off. There is evidence that levels of mercury and cadmium in ringed seals and beluga have been increasing over the last decade in the eastern Arctic and over the last 20 years in the western Arctic. These results, however, may be confounded by age differences in the samples analysed. Levels of radionuclides in caribou muscle and in lichen are available for over 30 years. Current levels of cesium in caribou muscle are four to more than ten times lower than levels measured in the 1960s.

Glacial ice and snow-core data provide a history of the deposition of atmospheric contaminants from the pre-industrial revolution era to present day. The majority of ice-core data for the Canadian Arctic have been collected from the Agassiz Ice Cap on Ellesmere Island. Temporal trend data from Arctic ice-core samples for several contaminants — for example PAHs, PCBs, and lead — generally reflect changes in the anthropogenic use and release of these contaminants to the global environment. For example, the background deposition of PAHs to the Arctic, during the pre-industrial era, was approximately 3 to 5 $\mu g \cdot m^{-1} \cdot y^{-1}$. Depositional rates increased and peaked at 70 $\mu g \cdot m^{-1} \cdot y^{-1}$ in 1970 and declined until 1980. From 1980 to 1993, there was a steady increase in PAH deposition from 10 to 20 $\mu g \cdot m^{-1} \cdot y^{-1}$, representing a two-fold increase over the last decade. The historical fluctuations in PAH deposition are attributed to the complex interaction of several factors including: the increase in fossil fuel consumption, the replacement of coal with cleaner fuels such as oil and gas, post-depositional processes, such as volatilization, that affect contaminant levels in the ice, and possible methodological uncertainties.

Temporal trends of PCBs, the least susceptible organochlorine to post-depositional loss from ice, have also been examined. Similar to PAHs, PCB deposition was high prior to 1969, after which a significant decrease in deposition occurred. This may reflect the discontinued production of PCBs in several developed countries. Despite the ban on the production of PCBs, however, they continue to be released into the environment as is reflected in the constant rate of deposition, on average 465 $ng \cdot m^{-1} \cdot y^{-1}$, to the Arctic between 1990 and 1993.

Ice-core data from the Greenland Ice Cap indicate that lead concentrations have been declining since the 1970s, coinciding with a decrease in the use of lead alkyl gasoline additives in Canada, the United States, and Europe (Boutron *et al.* 1995). Over a similar timeframe, lead concentrations in air at Alert, NWT have decreased to a lesser degree compared with concentrations in the Greenland Ice Cap. These results reflect the greater influence of Eurasian emissions compared with North American emissions at Alert.

Similar to the ice and snow-core data, sediment cores are useful indicators of the deposition of contaminants to the Arctic. Sediment cores from eight lakes along a mid-latitudinal transect from 42°N to 82°N were collected to determine latitudinal and temporal variations in contaminant deposition. Highest levels of all OCs measured were found in the top slices of sediment cores, representing the time period of 1960 to present (Muir *et al.* 1995, Muir *et al.* 1996, Lockhart 1996). While relative amounts of OCs from sediment profiles indicate higher deposition rates over the last 50 years, modelling results suggest that sediment profiles from arctic lakes may be limited in predicting absolute loading patterns. Modelling results suggest that arctic lakes act as conduits for chemicals (especially OCs) with a minimal amount of chemical being retained in bottom sediments (Diamond *et al.* 1996).

Currently, sediment core data is also proving useful in determining natural versus atmospheric contributions of heavy metal loadings to Arctic lakes (Lockhart *et al.* 1995, Lockhart 1996). For example, results from sediment core profiles of mercury in YaYa Lake, Mackenzie Delta, indicate that the main source of mercury to this system is geological. Similarly, elevated levels of mercury in surficial sediments from western lakes (Yukon and western NWT) in the Canadian Arctic have been attributed to natural sources. In contrast, elevated levels of mercury in sediment from eastern and far northern NWT lakes are attributed to long-range atmospheric transport. Sources to all of these lakes are determined by comparing concentrations from deep core sediment with surficial sediment. Levels of mercury in the surficial sediments (representing the post-industrial era) from the eastern and far northern NWT have increased three- to seven-fold.

The conflicting evidence from the sediment core data for mercury highlights the difficulty in determining anthropogenic versus natural sources of mercury to the Arctic. Globally, however, atmospheric concentrations of mercury in the Northern Hemisphere have been increasing for the past several hundred years (Slemr and Langner 1992). Anthropogenic releases to the atmosphere have been identified as the cause of an observed three-fold increase in global air and ocean surface water concentrations (Mason *et al.* 1994). The evidence from several Canadian Arctic sediment cores, as previously discussed, supports this conclusion.

Temporal trends for methylmercury in human tissues, discussed in more detail in Chapter 4, provides additional evidence. Recent temporal trends, from 1970 to 1989, in levels of methylmercury in human blood indicate no change in levels among northern Aboriginals. Methylmercury levels in archaeological human hair samples from Greenland, however, are lower than levels in hair samples recently collected from residents of similar regions, suggesting that levels of methylmercury in the north have increased since the beginning of the industrial era.

The majority of atmospheric measures of organochlorines have focused on HCHs, both the α- and γ-HCH isomers, as HCHs are one of the few organochlorine insecticides (lindane) still in large-scale global use. In the Canadian Arctic, recent atmospheric levels of HCHs have been measured at Alert and Resolute Bay (Barrie 1994, Bidleman 1994), although records have been collected since the early 1980s at stations in the Canadian and Norwegian Arctic and in the Bering and Chukchi seas. Concentrations of α- plus γ-HCH in the atmosphere at Alert and Resolute Bay have declined from 880 $\mathrm{pg{\cdot}m^{-3}}$ in 1979 to 100 $\mathrm{pg{\cdot}m^{-3}}$ in 1992/93. A reduction in air concentrations of HCHs is attributed to the recent decrease in the global release of the insecticides to the environment. It has been suggested that a likely consequence of the observed decrease in atmospheric HCH levels will be a reversal in the direction of HCH exchange due to oceanic HCH saturation. Net HCH exchange previously was from air-to-ocean, but is expected to reverse direction to an ocean-to-air exchange, although little change has been observed in the HCH concentrations in the Arctic Ocean to substantiate this hypothesis. The process of HCH exchange between the ocean and the atmosphere and a mass balance of HCH in the Arctic Ocean is discussed in more detail in Chapter 2.

Temporal trend information for the Arctic is limited for most organochlorines and metals because for the biotic media they are based on too few samples, and for abiotic media there is high variance in the results. Long time periods are required to detect statistically significant changes in contaminant levels because of the analytical and natural variability in many of the results. In general, however, results from abiotic (ice cores) and biotic (seals and sea birds) samples indicate that PCB levels declined between the 1970s and the 1980s and have since leveled off. Evidence for overall temporal trends for other organochlorines is conflicting. There is some evidence from abiotic (freshwater and marine sediments) and biotic (seals and beluga) samples that mercury levels have been increasing. This is consistent with the overall perceived trend of increasing global emissions of anthropogenic mercury. There is, however, little temporal trend information for terrestrial and freshwater environments. In comparison, contaminant levels in Lake Ontario lake trout and in various species in northern Sweden are available for the last 15 to 20 years. Continued monitoring of contaminant levels in all Arctic media will aid in determining whether contaminant levels in the Canadian Arctic are increasing, decreasing, or remaining the same.

6.4 Sources

Pollution originating from human activities in industrialized and agricultural regions of the world is the main source of contaminants to the Canadian Arctic. Details on these and additional sources of the contaminants of concern are discussed in Chapters 1 and 2. POPs originate from anthropogenic sources, the majority of which are external to the Arctic. They continue to enter the Arctic as a result of their use and application in both developed and developing countries (Goldberg 1991, Voldner and Li 1995). Accurate and complete information on the emission of these pollutants is required to develop effective international strategies to reduce the input of contaminants to the Canadian Arctic. To date, pesticide emissions from western Europe and North America are more accurately estimated than emissions from Asia and Eastern Europe due to more complete inventories and registration procedures (Pacyna 1995, Voldner and Li 1995). Efforts are needed to better quantify emissions of POPs from the developing countries and from the former Soviet Union and China.

For heavy metals, it is difficult to determine the proportion of anthropogenic versus natural sources to the total contaminant loading to the Arctic. All trace metals are found in association with both airborne particulates and crustal minerals. Mineralogy may be used to indicate local areas (as compared to external sources) expected to contain naturally elevated concentrations of a particular heavy metal. From an economic perspective, the extraction of base and precious metals has, for many years, been important to the economy of the North and nickel, copper, lead, zinc and silver are all currently mined in the Arctic. The by-products of the mining industry may result in elevated levels of trace metals and other contaminants within a localized area that may pose a threat to the health of the local environment. In contrast, localized, naturally elevated concentrations of heavy metals in areas undisturbed by human activities may result in elevated concentrations in resident biota, as previously discussed, but have little consequence for the health of the biota. In general, natural mercury sources are regional, for example in parts of the western Arctic, whereas natural cadmium sources are more widespread across the Canadian Arctic.

Background levels of radioactivity occur naturally, due mostly to the radioactive decay of uranium and thorium in minerals of the region. At present, people living in the north are exposed to greater amounts of natural than anthropogenic radionuclides. Historically, anthropogenic radionuclides of concern in the Canadian North, such as cesium, originated from the atmospheric testing of nuclear and thermonuclear weapons that began in 1955 and ended with the moratorium on this type of testing in 1963. The radioactive fallout from the Chernobyl accident in 1986 also resulted in the transport of cesium isotopes, and to a lesser extent, strontium, to the Arctic. Generally, however, cesium levels have declined in Arctic biota since 1963 and fallout from the latter sources has been buried or consolidated in soil and lake sediment (Thomas *et al.* 1992, Elkin and Bethke 1995). The burn-up of nuclear powered satellites upon reentry through the atmosphere is an additional, albeit small, atmospheric source of radionuclides to the Arctic. Other possible external sources of radionuclides include discharges from nuclear power plants and reprocessing plants, and nuclear waste dumping directly into the Arctic Ocean (Aarkrog 1993, Barrie *et al.* 1992, Yablokov *et al.* 1993). The impact of the ocean disposal of nuclear waste remains unknown. There are indications, however, that dispersion from ocean dump sites in some arctic coastal areas of the former Soviet Union is relatively slow and largely remains localized. Generally, levels of radionuclides in the air, water or soil of the Canadian Arctic are similar to or lower than levels in more temperate areas.

Local sources of pollution that exist within the Canadian Arctic include abandoned distant early warning (DEW) line sites and military sites, northern mining activities and local garbage dumps. Although these sources of pollution are not the principal cause of the widespread distribution of contaminants in the Canadian Arctic, they pose a threat to the health of local ecosystems by emitting a variety of pollutants (including POPs and heavy metals). It is estimated that approximately 0.2% of the Yukon and NWT land mass is contaminated by PCBs from abandoned DEW line sites (Gregor and Reimer 1995). Several sites contaminated from these sources have been inventoried through the AES Action on Waste Program and their restoration remains a priority for the Department of Indian Affairs and Northern Development.

6.5 Pathways and Fate

The atmosphere is the most important transport vector of POPs, heavy metals, and radionuclides to the Canadian Arctic. Work conducted under the AES-NCP, including modelling, spatial distribution of contaminants measured in snow, ice-cores and biota, and the monitoring of air through a circumpolar air-monitoring network, all supports this conclusion.

The transport of airborne contaminants to the Canadian Arctic is dependent on a number of factors including the circulation patterns of global and Arctic air masses, temperature, and a phenomenon known as the "cold-condensation" effect. The Canadian global spectral model has been used to demonstrate the complex atmospheric processes that transport contaminants to the Northern Hemisphere, and more specifically, to the Canadian Arctic (Ritchie 1991, Pudykiewicz and Dastoor 1996). Arctic air masses differ from winter to summer, resulting in seasonal differences in the atmospheric transport of contaminants. Generally, the transport of contaminants to the Arctic is greater in the winter due to a strong flow of air masses from the Eurasian continent into the Arctic and out over the North American continent. In the summer, a weaker south-to-north air flow from North America replaces the transpolar transport from Eurasia.

Temperature is also an important factor for the transport of airborne POPs. An increase in temperature results in a corresponding increase in the potential air mass burden of POPs. Therefore, summer air masses have the potential to carry larger amounts of POPs, although the source region differs from that in the winter. During the summer, additional atmospheric processes, such as increased photo-chemical activity, a build-up in moisture and a consequent increase in rain events, result in the removal of contaminants from the air masses, thus decreasing the extent of transport of contaminants to the Arctic in the summer (Barrie *et al.* 1992, Pacyna 1995).

The physicochemical characteristics of individual compounds, for example solubility in water or lipid, and volatility, are critical to their global transport and deposition, as well as to their exchange among environmental media within the Arctic. For example, OCs of intermediate volatility (that is volatile enough to evaporate substantially following use in temperate and tropical latitudes, but whose volatility decreases sufficiently at lower ambient polar temperatures) will deposit and partition to Arctic terrestrial and aquatic ecosystems (Wania and Mackay 1996). This "cold condensation" effect, is hypothesized to be the mechanism that results in the introduction of certain persistent chemicals in the Arctic. Semi-volatile compounds that are expected to be of significance in this process include chlordane, the lower chlorinated PCB congeners, γ-HCH (lindane) and HCB. In contrast, compounds with very low volatility, for example, DDTs, dieldrin, mirex, or the higher chlorinated PCB congeners, tend to condense closer to source regions and are less available for transport to colder areas (Wania and Mackay 1996).

For many contaminants, air-surface exchange, in particular exchange with water, is an important mechanism for their deposition to the Arctic. In general, gas exchange processes are critical to contaminant loadings as they are also relevant to precipitation scavenging of airborne contaminants. Future efforts to quantify POPs entering the Canadian Arctic will be confounded by distinguishing "new" sources of POPs from those that have been re-volatilized from previous deposition. For example, as previously discussed, some compounds, such as HCHs, may reach saturation levels in the oceans at certain times of the year, resulting in a volatilization from the ocean to the atmosphere (a reversal of flux). They may re-deposit within the Arctic, or be transported out. An estimate of the amount, or flux, of this re-volatilization versus the amount imported to the Arctic from external sources and subsequently re-distributed among environmental media has yet to be identified.

Snow comprises the majority of the wet deposition in the northern Arctic and is the most important medium for depositing contaminants from the atmosphere to terrestrial and aquatic systems. In southern regions of the Arctic (just north of 60°N), rain and snowfall are both equally important in contaminant deposition. Once snow is deposited on land, sea-ice and lake surfaces, melting events result in the release of contaminants, rendering them available for uptake by terrestrial, marine and freshwater organisms. Contaminants may also be further transported via ocean or river currents (Pfirman *et al.* 1995).

Oceans, rivers and sea-ice are additional transport vectors of contaminants to the Canadian Arctic, although relative to these, the atmosphere is the most significant. Contaminants present in the atmosphere reach the Arctic from external sources in a matter of days, whereas contaminants transported by ocean currents and sea-ice may take years or decades. The total amount of contaminant transport into the region via oceans is not well quantified; however, transport via ocean currents is slower than by the atmosphere and oceans act as a reservoir for many contaminants, especially POPs. Contaminants transported to the Arctic Ocean (and in particular to the Canadian Basin or Arctic Interior Ocean) may flow through Fram Strait and the Canadian Archipelago, or be scavenged within the Arctic Ocean by particulates and subsequently be deposited as sediment onto ocean shelves, slopes and basins, or be sequestered and metabolized by biota. Rivers are a potential transport mechanism of terrestrial sources of contaminants, including the heavy metal by-products of mining activities and natural erosion. Rivers may also act as a receiving body for atmospheric deposition to the surrounding watershed. Inputs of PAHs from the Mackenzie River (the largest Canadian freshwater input to the Arctic Ocean) to the Arctic Ocean are relatively high and attributed to natural sources. The high background concentrations of PAHs are below threshold levels known to induce toxic effects in aquatic biota. In general, the overall transport of contaminants to the Arctic Ocean via freshwater is relatively small compared with other loadings, such as those from the atmosphere.

Once transported and deposited in the Arctic, contaminants may become available for uptake by biota through air, food and water. The bioaccumulation of contaminants is discussed in more detail in Chapter 3, and is dependent on several factors, including the physicochemical properties of the contaminant and the biological mechanism of uptake. The fate and distribution of contaminants in animals and humans in the Arctic is primarily a function of food web dynamics. The position of humans at the top of many Arctic food chains renders them particularly exposed to the bioaccumulation and biomagnification of many contaminants.

6.6 Summary

The NCP has generated a considerable body of knowledge on the types and levels of contaminants found in the Arctic environment, wildlife species at most risk and of most concern as human food items, geographic trends in contaminant distribution, and major pathways for transport of contaminants into the Arctic. These findings provide the basis from which further key knowledge gaps can be identified.

Chapter 3 describes contaminant levels and geographic trends in wildlife species and emerging contaminants of interest for future study. It points to the need to improve understanding of mercury speciation and the biological relevance of mercury levels found in northern wildlife. Now that a relatively complete database on contaminants levels in biota exists, a critical question is: Do current levels of contamination have an effect on Arctic wildlife? If so, what are these effects? Associated with this question, is the need to know whether contaminant levels in wildlife are increasing or decreasing. To date, temporal trend data is relatively scarce, and tracking temporal change is important in evaluating potential future effects related to changes in contaminant levels.

The same question applies to the human health implications of current contaminant levels. Are there, or could there be, effects on humans? If so, what are they? The specific knowledge gaps are detailed in Chapter 4, but can be summarized as a need for information on i) the implications of current human exposure through food, and ii) the implications of current human body burdens of contaminants. Effects of individual contaminants (especially toxaphene and chlordane) and effects of mixtures are pressing toxicological questions for both these issues. With respect to management decisions related to exposure through food, it has become apparent that assessment of food safety must, when considering country foods, incorporate the nutritional, cultural and economic benefits of these foods. Knowledge about people's consumption habits is an important factor in evaluating the food-related risks and benefits that they may incur. Similarly, quantification of benefits is important for development of a balanced picture with respect to traditional diets. An approach for quantifying the risk of contaminants in food, while considering the benefits of those foods and the risks associated with abandonment of a traditional diet, is clearly needed if definitive advice is to be provided in future.

. Data on levels of contaminants in human tissues do not cover as wide a geographic area as data for wildlife species. In some areas, studies are ongoing, while in others, communities have expressed an active disinterest in participating in such studies. Little is known about the long-term effects of low-level environmental contaminant exposure on humans, particularly with respect to intergenerational effects. Given the lack of human tissue guidelines for all but mercury, lead, cadmium and PCBs, the need for a better understanding of current human contaminant body burdens is an issue which reflects a lack in the worldwide state of knowledge with respect to low level effects of contaminants. Although current contaminant levels are not considered to be an immediate threat to the health of adults, there are concerns over possible effects on the fetus and infants.

Whether the risks posed by contaminants in the Arctic diminish, increase or stay the same will be determined by changes in the overall contaminant inputs to the Arctic environment. Thus, the key questions in this regard are: Will contaminant levels change? In what way, and how quickly? Research has shown that major source regions, particularly for POPs, are outside the Arctic and mainly anthropogenic. For metals, although evidence indicates that recent increases in environmental levels are likely related to human activity, the proportion of ecosystem contamination that is derived from natural versus anthropogenic sources remains a key unknown. This is an especially important question for mercury, where data suggest that there have been significant increases in environmental levels in recent years.

For all contaminants of concern, global use, release and emissions need to be understood in order to be able to anticipate future changes in the Arctic. Similarly, calls for international action must be followed by detailed protocols that can identify areas to be targeted for change (geographic regions, economic sectors, etc.) This requires quantitative data on sources and emissions. Predictions of change also rely on an understanding of the behaviour of contaminants as they travel north via air and water currents, and their behaviour once they reach the Arctic — particularly with respect to their biological availability and cycling among different environmental media. As described in Chapter 2, progress has been made in developing models and budgets for transport and distribution of HCH to and within the Arctic. The further application of these predictive tools to contaminants of greater biological interest is necessary.

Underlying all of the research is the need to synthesize and interpret the results and make them accessible and useful to Northerners who wish to make their own informed decisions regarding traditional/country food in their diet. Led by the northern Aboriginal organizations, this has been an evolving component of the NCP, progressing to a cooperative process directly involving key community members in the interpretation and dissemination of information. Increasing capacity among the Aboriginal organizations, and in turn at the regional and local level, has been key to the NCP's efforts not only to disseminate program results, but also to involve Northerners actively in the research and planning of the NCP. Capacity at the community level is beginning to develop to the extent that the NCP has funded proposals for scientific research submitted directly to the program by northern communities. This direct community level involvement in the NCP has begun to add a new dimension to program management and implementation.

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Appendix I: Physical and Chemical Characteristics of Contaminants in the Arctic

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Physical and Chemical Characteristics of Contaminants in the Arctic

Organochlorines

Organochlorines (OCs) are stable and persistent in the environment. They have low water solubilities and are highly lipophilic, resulting in uptake and accumulation in the fatty tissues of living organisms. Those that are or have been used as pesticides are meant to be toxic, particularly to invertebrates or fungi, but may also be toxic to fish and mammals. These include DDT, hexachlorocyclohexanes (HCHs), chlordanes (CHLOR), aldrin, dieldrin, mirex, chlorobornanes (CHBs e.g. toxaphene), endosulphan and hexachlorobenzene (HCBZ or HCB).

Structural diagrams of selected OCs are shown in Figure 1. For additional information on physical properties, readers should consult reference texts of Howard (1991) and Mackay *et al.* (1992a,b).

All of the OCs mentioned have been detected in Arctic abiotic or biotic samples and most of them are on lists of priority pollutants established by governments for regulation of environmental contaminants. Many of these contaminants are associated with biological effects observed in more polluted environments such as the Baltic Sea, the North Sea and the Great Lakes. Exposure to certain OCs has been associated with immuno-suppression, endocrine disruption, reproductive failure (lower numbers of young born, unhatched eggs, early death of young), neurobehavioural effects, developmental impairment, and induction of liver detoxification enzymes (mixed function oxidases, cytochrome P450).

The importance of physical characteristics in the distribution of OCs in the Arctic is exemplified in Figure 2 (Norstrom and Muir 1994). The figure shows the relative burden of various OCs in air, snow, seawater, and the marine mammal food chain. In the principle medium of transport, air, the most volatile OCs are the most prevalent. PCBs and DDT, are barely detectable in this medium. The reason for the variation in distribution has to do with the relative volatility and water solubility of various compounds. Incorporation into the bottom of the food chain may be less efficient for compounds with relative lipid/water solubilities in the order of 10^4 or less. This property may be predicted by the octanol water partition coefficient (K_{ow}). HCH has a K_{ow} in the order of 10^4, while the K_{ow} of HCB, chlordanes, and

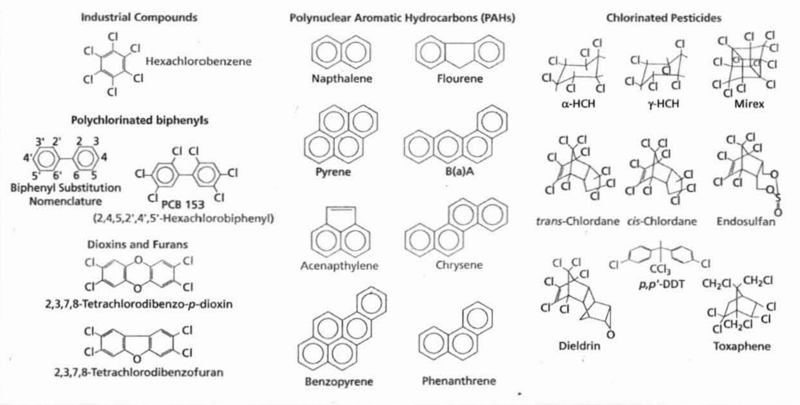

FIGURE 1

Structures of major organochlorine contaminants in this report.

PCBs is 10^5–10^8. In the higher levels of the food chain, OCs with higher K_{ow} values biomagnify to a much greater degree.

The intake of OCs by arctic residents consuming large amounts of traditional foods from the freshwater and marine environments are considerably higher than intakes by persons eating primarily store bought foods, since OCs are biomagnified in the food web. OCs are selectively stored in fat and excreted primarily through feces and mothers' milk. Nursing infants are subsequently at risk of exposure to these contaminants. Since OCs accumulate in body tissue, exposure occurring prior to pregnancy can contribute to the overall maternal body burden and result in exposure to the developing fetus during pregnancy.

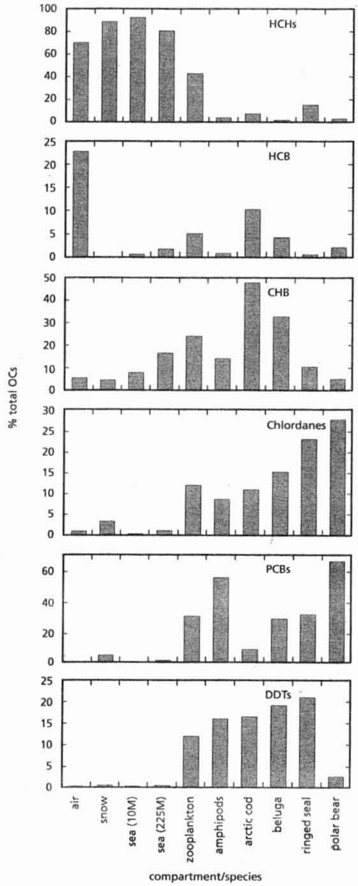

FIGURE 2

Distribution of chlorinated hydrocarbon contaminants (OCs) in Arctic air, snow, seawater, and the marine mammal food chain. Data for the six major classes of chemicals are plotted for each compartment or species as percent of total OCs in that compartment or species. Taken from Norstrom and Muir (1994).

Industrial Organochlorine Chemicals and By-products

Hexachlorobenzene (HBC or HCBZ)

HCB is produced as a by-product in the production of a large number of chlorinated compounds, particularly lower chlorinated benzenes, and in the production of several pesticides. It has been used in small amounts as a fungicide and it has limited use as a pesticide. It is emitted to the atmosphere in flue gases and fly ash generated at waste incineration facilities. HCB has a relatively high bioaccumulation potential because of its high lipophilicity and long-half-life in biota (Niimi 1988). Other chlorobenzenes (tetra- and penta-substituted) are also relatively lipophilic, semi-volatile, and persistent especially in the abiotic environment (Mackay *et al.* 1992).

Animal studies show that ingestion of hexachlorobenzene on a long-term basis can harm the liver, immune system, kidneys, and blood, and produce eruptions and pigmentations of the skin. These studies also suggest that ingestion of hexachlorobenzene on a long-term basis can lead to cancer of the liver and thyroid. There is weak evidence that marginal perturbations of porphyrin metabolism may occur with blood HCB levels in the 2–40 $\mu g \cdot L^{-1}$ range. Such subclinical effects are not expected in association with the blood levels ($<2\ \mu g \cdot L^{-1}$) seen in the general population.

The International Agency for Research on Cancer (IARC) has classified HCB as a possible human carcinogen, based on sufficient evidence in animals, but inadequate evidence of carcinogenicity in humans.

Polychlorinated biphenyls (PCBs)

Commercial PCB mixtures are technical products with widespread uses. PCBs were introduced in 1929 by Monsanto Chemical Corporation and were manufactured in the US, Japan, the former Soviet Union, and eastern and western Europe. They are chemically stable and heat resistant, and were used worldwide as transformer and capacitor oils, hydraulic and heat exchange fluids, and lubricating and cutting oils (Verschueren 1983). Information about their global use and production is given in Chapter 2. Open use is currently banned in all circumpolar countries but large amounts are still in use in closed systems (large capacitors and transformers). Current uses and disposal practices in the developing world are not well documented.

There are 209 chlorinated biphenyl congeners, with different chlorine substitutions on the biphenyl ring (Mackay *et al.* 1992). The number of chlorines, as well as their positioning on the rings, influences the physical properties and biological activity of PCBs. As with other OCs, molecular weight and the K_{ow} relate to volatility, solubility, and the tendency to adsorb to organic surfaces. PCB congeners with 3,4,3′,4′ chlorine substitutions are the most biologically active and are referred to as co-planar or non-ortho PCBs (Ahlborg *et al.* 1994) (Figure 1). They lack chlorine substituents in the 2 and 6 (or ortho) positions and can therefore assume a planar configuration. The toxicity of Aroclor mixtures, commercial PCB formulations in North America, is thought to be due almost entirely to these co-planar PCBs (Kannan *et al.* 1988). Their biological effects include induction of hepatic mixed function enzyme oxidase enzymes, immunotoxicity, teratogenicity, and embryotoxicity (Safe 1990). Some mono-ortho PCBs, which have a single chlorine in the 2-position, have enzyme induction potencies similar to that of the co-planar molecules. Co-planar and mono-ortho PCBs are present at relatively high proportions of total PCBs in the fatty tissues of ringed seal, walrus, narwhal, polar bear, and Arctic char.

Studies of humans exposed to high levels of PCBs, through occupational or accidental exposures, show that acne-like skin lesions and rashes (chloracne) are the primary observed health effect. Longer term occupational exposure may also be associated with reproductive effects and increased incidence of liver cancer.

Polychlorinated-dibenzo-*p*-dioxins and furans (PCDD/PCDFs)

Polychlorinated-dibenzo-*p*-dioxins and furans (PCDD and PCDFs) are byproducts formed in the production of chlorinated substances, combustion processes and in direct chlorination reactions such as chlorine bleaching of paper pulp. The most significant sources are municipal waste and hospital incinerators, which combust wastes that include chlorinated products. An important additional source is motor vehicle fuel combustion in countries where leaded fuel containing chlorine scavengers is still used. Pulp and paper mills using chlorine in the bleaching process have been important sources to the aquatic environment of 2,3,7,8-substituted TCDD. PCDD/Fs are also trace contaminants in chlorophenoxy herbicides and chlorophenol wood preservatives. Tetrachlorinated dioxin (2,3,7,8-TeCDD) is the most toxic among this group of chemicals.

Dioxins and furans are very toxic to some animal species, but the evidence for corresponding toxicity in humans has not been established. The physiological effects of exposure to high levels are similar to PCBs (carcinogenisis in animal studies, immune suppression, hormonal disruption, and developmental impairment). The IARC considers dioxins and furans as possibly carcinogenic to humans.

Chlorinated Pesticides

Chlordane (CHL)

Technical-grade chlordane is a mixture of at least 50 compounds, with the major constituents being *cis-* and *trans*-chlordane, heptachlor, *cis-* and *trans*-nonachlor, α-, β-, and γ-chlordane, and others (Howard 1991, National Research Council of Canada 1974). Because heptachlor is more potent than other chlordanes, efforts were made in the past to produce technical mixtures that were at least 70% heptachlor. Oxychlordane is a toxic metabolite of chlordane. In the past, chlordane was released into the environment primarily from its application as an insecticide and for seed dressings and coatings (CCREM 1987). In the US, it was used extensively before 1983, limited to use for termite control between 1983 and 1988, and cancelled for use after that year. Chlordane is persistent in the environment. Heptachlor is of particular interest since its oxidation product, heptachlor epoxide, is carcinogenic and has been found in the Arctic physical and biological environment.

Although many effects of high level exposure are known, the health effects of long-term exposure to low levels of chlordane in the environment are poorly understood at this time. The IARC has concluded that chlordane is not classifiable as to its carcinogenicity to humans. However, the US EPA (Environmental Protection Agency) has classified chlordane as a probable human carcinogen based on oral studies in animals.

Chlorobornanes (CHBs)

The complex mixture of chlorobornanes (CHBs), known as toxaphene, was widely used in the US on cotton crops until the early 1980s. Use peaked between 1972 and 1975 and declined in the late 1970s and early 1980s. Manufacture was banned in the US in 1982; use was banned in 1986 Similar products are still used in Mexico, central America, eastern Europe, and the former Soviet Union. Toxaphene has been globally dispersed by atmospheric transport to the same extent as DDT and PCBs.

Toxaphene is produced by the chlorination of bornane and can consist of over 300 compounds, substituted with 6 to 10 chlorines, with an average composition of $C_{10}H_{10}Cl_8$. Analysis is difficult because of the mixture's complexity, because it occurs in the presence of other interfering OCs (PCBs, DDTs, HCHs), and because of the lack of standards. For this reason, the levels and effects of toxaphene in the Arctic are not well studied even though it is a major concern.

Toxaphene is one of the major contaminants detected in arctic biota (particularly fish and marine mammals) (Andersson *et al.* 1988, Kawano *et al.* 1988, Muir *et al.* 1988, Norstrom *et al.* 1988, Lockhart *et al.* 1989). Lockhart and co-workers noted that in some locations consumption of fish and their livers by native people could lead to an intake of CHBs that exceed the US National Academy of Sciences acceptable daily intake of 1.25 $\mu g \cdot kg^{-1} \cdot day^{-1}$ (Bidleman *et al.* 1989).

Exposure in humans from breathing, eating or drinking high levels of toxaphene for brief periods has been reported to cause damage to the lungs, nervous system, liver, and kidneys, and can result in death. Gastrointestinal discomfort and seizures as well as chromosomal aberrations have been reported in humans exposed occupationally to toxaphene. Limited data suggest that chronic exposure to low environmental levels of toxaphene may also result in some effects on the liver, kidneys, lungs, and nervous system.

There is no conclusive evidence available to link toxaphene exposure with cancer in humans. The IARC has classified toxaphene as possibly carcinogenic to humans. Based on animal studies (thyroid and hepatic carcinomas), toxaphene has been classified as a probable human carcinogen by the US EPA. Toxaphene has recently been observed to have estrogenic effects on human breast cancer estrogen-sensitive cells. Xenoestrogens have been hypothesized to have a role in human breast cancer. In addition to potential carcinogenic effects, toxaphene may also cause disruption of the endocrine system due to its estrogenic activity.

Hexachlorocyclohexane (HCH)

Technical HCH *(1,2,3,4,5,6-*hexachlorocyclohexane*)* consists of a number of isomers: 55–70% α-HCH, 5–14% β-HCH, 10–18% γ-HCH (also called lindane), δ-HCH and impurities (Iwata *et al.* 1993). Technical HCH is still used in China as an insecticide on hardwood logs and lumber, seeds, vegetables and fruits. Lindane (γ-HCH), the most biologically active insecticidal isomer, is the only form of HCH currently used in North America, Japan and Europe (Bidleman *et al.* 1989), where it is used mainly in seed treatment. Other isomers have been banned for use in the US and most other circumpolar countries since the late 1970s.

Levels of exposure to HCH that produce harmful effects on human health are generally much higher than normally received by the general population. Of the three HCH isomers, γ-HCH is the most toxic but it does not bioaccumulate to any significant extent. Long term exposure can affect the liver, the kidney, the reproductive system, and perhaps the immune system. The IARC has classified HCH as possibly carcinogenic to humans.

Mirex

Mirex is of interest because of its high K_{ow} and its persistence. It was used as an insecticide and fire retardant, mainly in the US and Canada until it was banned in 1978. High levels of mirex were found in Lake Ontario and St. Lawrence river biota as a result of past industrial releases. In these areas, levels are declining in lake trout and coho salmon, but not in herring gulls and beluga whales. Its presence in Arctic biota at low levels is somewhat surprising, but may be explained by exposure of migratory species to this contaminant when feeding at southern latitudes.

Information on the human health effects of mirex are limited. Chronic exposure is related to renal effects, decreases in body weight or body weight gain greater than 10%, non-precancerous lesions of the liver, and cancer (an increased incidence of hepatocellular adenomas was noted, but only in animals having hepatotoxicity). The IARC has classified mirex as a possible human carcinogen, based on sufficient evidence in animals, but inadequate evidence of carcinogenicity in humans.

Endosulfan

Endosulfan is a very toxic insecticide still widely used against a variety of insects especially on high value crops and particularly in tropical countries. Isomers of endosulfan are detected as contaminants in air, water, sediments, soil, fish, other aquatic organisms, and food. It has been measured and detected in the Arctic in a limited number of studies. It is a concern mainly because of its high volume usage at more southern latitudes.

Little is known of the effects of long-term exposure to low levels of endosulfan in humans. Animal studies have shown effects on the kidneys, testes, developing fetus, and liver from long-term exposure to low levels of endosulfan. The ability of animals to fight infection was also lowered. Animal studies have not shown that endosulfan causes cancer and no human studies are available. Neither the IARC nor the US EPA have classified endosulfan as a human carcinogen.

DDT

DDT was introduced in 1945 as an insecticide. Its use has been restricted in Canada, the US and Europe for more than two decades, however, it is still manufactured and used as a pesticide in southern Asia, Africa, and Central and South America (Voldner and Ellenton 1988, Voldner and Li 1995). Levels of DDT and its principal metabolite, DDE, have decreased in the environment since the 1970s. DDT is still entering the Arctic ecosystem through atmospheric transportation and deposition.

Target organs for DDT, DDE and DDD toxicity include the nervous system, the reproductive system, and the liver. DDE is stored in fatty tissues in biota. DDE was identified as the cause of eggshell thinning in many birds and has since been found to disrupt endocrine hormone metabolism and change the activity of liver enzymes. An association between exposure to DDT and the development of cancer has not been found in studies of occupational exposure to DDT. Based on animal studies (oral exposure), the US EPA has concluded that DDT, DDE and DDD are probable human carcinogens.

Recent data indicate that current blood and adipose tissue concentrations of DDT in humans are well below levels associated with clinical manifestations. However, there may be instances where DDT intake may surpass guideline levels in breast-fed infants (corresponding to 1 $mg{\cdot}kg^{-1}$ total DDT in milk, fat basis). Suppression of reflexes in neonates appear to be associated with levels of DDE in breast milk exceeding 4 $mg{\cdot}kg^{-1}$ (fat basis), however, it has not been substantiated if DDE is the causative factor. Elevated levels of DDE in human breast milk (3.0 $mg{\cdot}kg^{-1}$, fat basis) have been correlated with a shortening of breast-feeding duration; inhibition of lactation was hypothesized. Blood concentrations have shown a dependence on alcohol consumption and serum triglyceride levels, while duration of breast-feeding and parity influence milk concentrations.

Dieldrin

Dieldrin is no longer manufactured in the US, and its use is now restricted for termite control. It was used mainly as a soil insecticide and was also released as the major degradation product of aldrin (also no longer in use). Manufacture in Europe, especially for export to developing countries, continued until the late 1980s. It is extremely persistent in soil and has a long half-life in biota.

Studies with animals fed dieldrin have shown liver damage and immuno-suppression. However, long-term occupational exposure to low levels of aldrin and dieldrin has not been documented as resulting in any demonstrable adverse effects. The IARC has concluded that evidence for carcinogenicity of aldrin and dieldrin is inadequate. The US EPA considers aldrin and dieldrin to be probable carcinogens based on sufficient evidence in animals.

Polynuclear Aromatic Hydrocarbons (PAHs)

Polynuclear aromatic hydrocarbons (PAHs) are predominantly byproducts of high temperature thermal reactions. The majority of environmental PAHs are formed by the incomplete combustion of organic materials such as fossil fuels, wood, tobacco, the incineration of garbage, and by steel and coke production, coal liquification, and gasification. Soot can be a rich source of PAHs and other organic compounds (Daisey *et al.* 1981). Small amounts are formed naturally from the transformation of organic material by micro-organisms.

Studies of the coastal zones of industrialized regions have described elevated PAH concentrations in surface sediments (Hites *et al.* 1980, Bouloubassi and Saliot 1991) and documented biological effects as a consequence (Malins and Hodgins 1981). In the Arctic, PAH levels in the abiotic environment are low compared with industrialized areas. They are not persistent in biota.

The greatest sources of exposure to PAHs for the general population are inhalation of tobacco smoke (active or passive), wood smoke and contaminated air, and the ingestion of PAHs through smoked food. The total daily potential exposure of carcinogenic PAHs for adult males is estimated to be 3 µg and may be as high as 15 µg. For smokers, exposure levels may be twice as high. For non-smokers, food is the main source of PAH exposure (up to 99% of total). The main excretory routes are feces, urine, and breast milk.

The toxicological significance of PAHs is found in their potential to form mutagenic and carcinogenic diols and epoxides that react with DNA, thus leading to mutagenesis and cancer (Zedeck 1980, Malins and Hodgins 1981, Stein *et al.* 1990). Benzo[a]pyrene is the most studied of PAHs and is classified as probably carcinogenic to humans by IARC. Several other PAHs, including benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenzo[a,h]anthracene, and indeno[1,2,3-cd]pyrene have caused tumours in laboratory animals when they were exposed via ingestion, inhalation, or dermally for long periods of time. Lung cancer following inhalation exposure, and skin cancer following dermal exposure, have been reported in workers exposed occupationally to PAHs. Additional effects reported from occupational exposure to PAHs include chronic bronchitis, dermatitis, cutaneous photosensitization, and pilosebaceous reactions.

Inorganic Contaminants

Heavy Metals

Heavy metals are identified by their grouped position on the Periodic Table, and exhibit some similar physical and biological properties. Unlike OCs, heavy metals occur naturally in the earth's crust, and are bound and released by normal physical processes. Heavy metals may occur as free ions, inorganic complexes or bound to organic material.

There are also anthropogenic sources of heavy metals (e.g. mining, smelting, fossil fuel and waste combustion). Increased deposition of metals on the earth coincided with the industrial revolution. Data from ice cores from Greenland confirm that Pb, Cd, Zn, and Cu concentrations increased from approximately 200 years ago to about the mid-1960s (Boutron *et al.* 1995). In the past two decades, there has been a decrease of some atmospheric metals due to the control of industrial emissions.

Heavy metals compete for active biological sites with other essential non-toxic cations such as calcium, iron and selenium. These same cations may decrease the uptake of toxic metals into an organism. Because of similar physical properties, the effects of metals may be additive or synergistic. For example, the acute joint toxicity of copper, cadmium, nickel, and zinc, can usually be described with models with linear additive functions of concentrations (EIFAC 1980). The relationship between levels of metals in organisms and levels of metals occurring in the environment is often unpredictable (Presley 1994). Metals can be taken up through food, however they generally do not accumulate up the food chain as organic contaminants do. There are a few studies which demonstrate biomagnification of heavy metals and studies generally show inconsistent results. (Mance 1987).

The three heavy metals of greatest concern in the Arctic are lead, mercury, and cadmium, all of which have been found near or above recommended levels in major species of interest. Other metals, some with similar properties to the above, but either less prevalent or less toxic, include copper, zinc, aluminum, nickel, arsenic, iron, manganese, and tin.

Lead

Lead is one of the most toxic heavy metals known to affect the nervous system, reproductive system, hemoglobin synthesis and the cardiovascular system. These effects occur at lead levels slightly above those found in the general population. Intake of lead from non-food sources (water, dust and soil) are often the most important human concern (National Research Council 1978).

In the past, most environmental lead was derived from lead additives in gasoline. In 1981, lead-alkyl additives were banned in gasolines and have decreased in the environment since that time.

The effects of lead are the same regardless of whether it enters the body through inhalation or ingestion. Susceptibility to lead toxicity is influenced by dietary levels of calcium, iron, phosphorus, vitamins A and D, dietary protein and alcohol.

Fetuses, infants, children up to six years of age, and pregnant women are at greatest risk of adverse health effects from exposure to lead. The central and peripheral nervous systems are the principal targets of lead toxicity. Studies indicate that exposure to even low levels of lead prior to birth, or during infancy and early childhood can potentially cause reduced birth weight, impairment to intellectual development (decrease in IQ), behavioral disturbances, decreased size, speech and hearing impairment and effects on peripheral nerve function. Exposure to high levels of lead can cause damage to the brain and kidneys of adults and children and has been linked to increased incidence of spontaneous abortion, miscarriages and stillbirths in women and damage to the male endocrine and reproductive system (decreased sperm production). Lead interferes with a hormonal form of vitamin D, which affects multiple processes in the body, including cell maturation and skeletal growth.

In adults, health effects from chronic exposure to lead include muscle weakness, anorexia, paresthesia in the lower limbs, poor performance on cognitive tests and visual-motor coordination, impaired verbal reasoning ability, anemia, abdominal pain and constipation, fatigue, and sleeplessness. Lead exposure may increase blood pressure in middle-aged men but it is not known if it affects blood pressure in women.

Current evidence suggests possible adverse health effects on children at blood levels of 10 $\mu\cdot dL^{-1}$. The Canadian Federal-Provincial Committee on Environmental and Occupational Health has adopted this blood level as the first criterion of intervention in both adults and children. In 1990, it was estimated that for the general population, approximately 5% of children exceeded a mean blood lead level of 10 $\mu\cdot dL^{-1}$.

Cadmium

Cadmium (Cd) is an extremely toxic element for plants, animals, and man. Cd emissions from various high-temperature sources, (smelting of ores, incineration, combustion of fossil fuels) and the use of cadmium in some domestic products such as batteries, pigments, plastic stabilizers, have lead to continuing concern about the health effects of Cd. It does not biomagnify along food chains, however its high availability and toxicity, and tendency to accumulate in kidney tissue (National Research Council 1979a) requires that this element be carefully evaluated.

Aerial pollution from Russian industrial sources has been related to increased cadmium in blood and hypertensive diseases in reindeer herders in Arctic Finland (Luoma *et al.* 1995). Cadmium is often a complicating factor in lead poisoning, and effects of the two metals have been confused. The main sources of cadmium in the Canadian diet are organ meats such as liver and kidney, aquatic animals such as molluscs and crustaceans, rice and wheat.

The main long-term effects of high level exposure to cadmium are kidney disease, chronic obstructive lung disease such as emphysema (limited to occupational exposure), and bone disease (osteomalacia, osteoporosis, spontaneous fractures and severe pain). A disease called *itai-itai* was first noticed in Japan where it has been associated with consumption of foods with elevated levels of cadmium over the long-term (compared with reference areas) and is characterized by bone disease.

Chronic exposure to high levels of cadmium has been reported to cause mild anemia, anosmia (loss of smell), yellowing of teeth, and occasionally, liver damage. High blood pressure has been observed in animals exposed to cadmium but it is not clear whether cadmium plays an important role in human hypertension. Tissues reported to be injured by cadmium exposure in animals or humans include the testes, the immune system, the nervous system, and the blood.

Cadmium's carcinogenic effects have been demonstrated in animal studies (lung tumours in rats inhaling cadmium compounds). Epidemiologic studies of occupationally exposed workers suggest a possible association between cadmium inhalation and lung cancer. The IARC has classified cadmium as carcinogenic to humans.

Mercury

Mercury is a non-essential natural element and is a ubiquitous contaminant in air, water and food. It is the only heavy metal that has a high vapour pressure and occurs in the atmosphere in the vapour state. Mercury bioaccumulates in the form of methylmercury. Food, particularly fish and seafood, is the major source of exposure to mercury for humans and other carnivorous species. Government action has led to the reduction of mercury release from a number of industrial processes after a number of cases of serious mercury toxicity in humans. Methylmercury has been cited as the possible cause of a neurological disease in some native communities which consumed large amounts of contaminated fish.

In humans, the main effects of exposure to mercury are neurological and renal disturbances. Long-term exposure to either organic or inorganic mercury can result in damage to the brain, kidneys, and developing fetus. The form of mercury (organic, elemental, inorganic mercury salts) and the route (ingestion, inhalation) of exposure determine the severity of these health effects.

Effects from acute and chronic exposure through ingestion are known primarily from large populations affected by the ingestion of methylmercury contaminated fish in Japan (Minimata Bay, 1956 and Niigata, 1965), and through the ingestion of methylmercury contaminated seed consumed as bread (Iraq 1971–72). In Canada, some Native and Inuit communities consuming large amounts of methylmercury contaminated fish and wildlife have reported symptoms which suggested mild poisoning. The health effects are known as "Minimata Disease" characterized by progressive nervous system effects.

Prenatal exposure to organic mercury affects the development and maturation of the central nervous system and results in psychomotor retardation. Such exposure can occur at levels at which no maternal effects are noted (maternal hair mercury levels >10-20 $\mu g{\cdot}g^{-1}$). Delays in walking and speech of over 12 months may occur (maternal hair mercury levels 50 $\mu g{\cdot}g^{-1}$). Retarded walking may occur at maternal hair levels of 6–10 $\mu g{\cdot}g^{-1}$, with a threshold at 5 $\mu g{\cdot}g^{-1}$. Neurobehavioural dysfunction may occur at maternal hair levels of >6 $\mu g{\cdot}g^{-1}$. Seizure and features resembling cerebral palsy often arise. Mild neurologic and developmental delays have been reported in infants exposed to methylmercury through breast milk.

Copper

Copper is a biologically necessary trace element, however increased levels in water or in tissues have detrimental effects. There are many data on acute lethal effects, usually obtained from experiments with high levels of copper, but relatively few data on chronic effects (National Research Council 1979b).

In general, copper enters the aquatic environment primarily from soils and mineral deposits by the erosional activity of water. Most natural waters are capable of removing copper from the water column. The chemical form of copper in water, and hence its toxicity, is related to pH, alkalinity, salinity, and complexation with ligands, clays, inorganic particulates, hardness (Ca^{++} and Mg^{++}) and metal oxides. Certain chemical forms exhibit varying degrees of absorption and toxicity. Because of this, there is much uncertainty in identifying detrimental concentrations under different circumstances. Bottom-dwelling organisms that feed by particulate filtration may be especially susceptible to adsorbed copper. Whole body copper concentrations tend to decrease as the trophic levels increase, presumably because consuming organisms are capable of metabolic regulation.

In humans, copper is necessary for good health. However, very large single or daily intakes of copper can be harmful to health. Drinking water that contains higher than normal levels of copper may cause vomiting, diarrhea, stomach cramps, and nausea. Very young children are sensitive to copper, and long-term exposure to high levels of copper in food and water may cause liver damage and death. Copper is not known to cause cancer or birth defects in humans. The seriousness of the effects of copper can be expected to increase with both level and duration of exposure.

Radionuclides

The largest doses and risks of radionuclides in the Arctic are from natural radionuclides, predominantly ^{210}Po (Polonium), ^{210}Pb (Lead), ^{228}Th (Thorium) and ^{232}Th. Among the radionuclides derived from anthropogenic sources, ^{137}Cs (Cesium) is the major contributor to dose but represents only 2% to 3% of the total dose and derives from land-mammal consumption.

Anthropogenic radionuclides of interest and concern in the Arctic environment are:

^{137}Cs: This Cs isotope is produced during fissioning (splitting of atoms) of uranium and plutonium fuels. It is long lived (radiation half-life of 30 years) and is found in the environment as a result of worldwide fallout from atmospheric weapons tests and nuclear power production. It is used in industry as a sealed gamma source for measuring the thickness of materials and in medicine as a sealed source for therapy and as a tracer substance.

^{90}Sr: Strontium has 6 radioisotopes which are direct fission products of uranium. ^{90}Sr is the most important of these because of its long radiation half-life of 29 years. In medicine, it is used to treat cutaneous lesions that are only a few millimetres in depth. In industry, it is used in thickness gauges, as a source for static dust elimination by air ionization, a compact heat source, and a thermoelectric source in devices where a long-lived, independent power source is needed. ^{90}Sr is found in the environment as a result of nuclear fallout during atmospheric weapons tests. It accumulates in bones and teeth.

Measurable levels of some radionuclides still remain in temperate and Arctic ecosystems after the weapons testing era (1952–1981). In northern ecosystems, higher levels of the longer lived fission products (^{137}Cs (half-life 30.2 years) and ^{90}Sr) are still present due to slower turnover rates in a cold and dry climate. The 1986 Chernobyl accident released additional ^{137}Cs into the atmosphere, and increased loading in boreal Canada by approximately 5% (Paliouris *et al.* 1995). They are also transported by ocean currents from industrialized countries of Europe.

^{131}I: Radioactive iodines, especially ^{131}I, ^{132}I, and ^{129}I, are fission products from nuclear weapons tests and nuclear reactors. Once released to the atmosphere, radioactive iodine may return via precipitation to land used for pasture, thereby contaminating vegetation and, ultimately, the food and milk supply. Radioiodine is important in terms of its selective irradiation of the thyroid.

^{239}Pu: Plutonium is a man-made element used as fuel in nuclear power reactors and as an explosive in nuclear weapons. ^{239}Pu has a radiation half-life of 24 390 years. Nuclear weapons testing programs have placed more than 5 000 kilograms of plutonium, mostly as insoluble particles of oxide, into the stratosphere, which has resulted in worldwide deposition. Actual concentrations in the environment are extremely low.

^{222}Rn: Radon results from the radioactive decay of radium, a ubiquitous element in rock and soil derived from the decay of uranium. Radon gas seeps from soil into buildings primarily through sump holes, dirt floors, floor drains, cinder-block walls, and cracks in foundations and concrete floors. When trapped indoors, it can accumulate to significant levels.

Tritium ($^3H_2$): Tritium is a hydrogen atom with a nucleus containing two neutrons. It has a radiation half-life of 12.5 years. Tritiated water (^3H_2O) is the most familiar form of tritium. It is formed when heavy water (^2H_2O) absorbs neutrons during the process of moderating nuclear fission in a reactor. Tritiated water has also been produced in the atmosphere as a result of the release of tritium gas from nuclear reactors. By far, the greatest part of the tritium found in the environment is due to atmospheric nuclear weapons tests conducted prior to 1963.

^{238}U: Naturally occurring uranium consists of ^{238}U (99.27%), ^{235}U (0.72%) and ^{234}U (0.0054%). ^{235}U is extracted or concentrated from natural uranium for use in nuclear power reactors or nuclear weapons. The uranium remaining after ^{235}U has been removed is referred to as "depleted uranium," which continues to be a radiation as well as a chemical hazard. In general, chemical damage is more important than radiological effects. High levels of uranium have been detected in well water in various parts of Canada.

Human Health

Chronic low level exposures are of most concern in cases of exposure to environmental levels of radiation. Early injuries result from acute external radiation exposure with consequent damage to tissues which have a rapid turnover, such as bone marrow and the gastrointestinal mucosa. Late or delayed effects of ionizing radiation may not appear for several decades after exposure, and can result either from massive doses that have caused early effects, or from relatively low exposures received over an extended period of time.

The health effects caused by exposure to ionizing radiation may appear as either hereditary or somatic effects. Hereditary effects result from exposure of the reproductive organs, which may induce mutations in the genetic material of an exposed individual. The results of these mutations may appear in the offspring of the persons exposed and may range from minor disorders to serious defects. No conclusive effects, attributable to exposure from either natural or artificial radiation, have been found in human offspring. Somatic effects are the most common consequence of radiation exposure. *Early* somatic effects appear in the individual within days or weeks after a significant external exposure and may include nausea, loss of hair, sore throat, hemorrhage and diarrhoea. The main *late* somatic hazards from exposure to environmental levels of radiation are the development of leukemia and other cancers, for example, in the bone, thyroid or lung, as well as cataracts of the eye. The probability that a malignant disease will develop following exposure is dependent on the radiation dose received. The severity of the disease is independent of the exposure, and radiation-induced cancers are indistinguishable from those that occur from other reasons.

Some radionuclides have a tendency to concentrate in certain tissues as a result of their interaction with normal physiologic processes. For example, cesium and strontium isotopes tend to congregate in bone, whereas the thyroid gland selectively concentrates iodine and radioiodine.

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Appendix II: Interlaboratory Quality Assurance and Quality Control Program for the Northern Contaminants Program

Jiping Zhu

1. Introduction

The research and monitoring projects that form the Canadian Arctic Contaminants Assessment Report (CACAR) were funded mainly by the Arctic Environmental Strategy's (AES) Northern Contaminants Program (NCP). Individual sampling as well as analytical methods have been described in various references cited in previous chapters (see Chapters 2, 3 and 4). Descriptions are also available in the annual *Synopsis of Research Conducted Under the Northern Contaminants Program* (Environmental Studies Nos. 68, 70, 72 and 73, published by the Department of Indian Affairs and Northern Development). This appendix provides information on the overall quality of the NCP's analytical data, with focus on issues such as data comparability and interlaboratory variability of analytical data used. The information contained in this appendix is largely the result of the Interlaboratory Quality Assurance Program for Analytical Data (QA Program) produced by NCP/AES (Zhu 1994, 1995a, 1996).

The QA Program was an on-going program for the duration of the NCP aimed at establishing acceptable levels of accuracy and precision of analytical results produced for the NCP, and at providing information on data comparability among many projects of the NCP. The QA Program was also designed to improve the performance of participating laboratories by establishing laboratory quality assurance and quality control (QA/QC) guidelines, providing recommendations on sampling and chemical analysis, and communicating with laboratories including site visits and laboratory inspections.

The QA Program started with a survey on laboratory QA/QC programs. The survey demonstrated that all NCP laboratories meet the minimum QA/QC requirements of conducting chemical analysis such as valid analytical methods, qualified personnel, adequate laboratory equipment and instruments, and function of internal QA/QC program etc. (Zhu 1993). Each NCP laboratory was visited by a QA/QC officer at least once during the monitoring program (Garfield 1991). These visits established an effective means of communication between the laboratory and the officer, enabling the officer to discuss many quality assurance related issues and analytical protocols with laboratory personnel on site.

The major activities of the QA Program were the series of interlaboratory comparisons on many contaminants monitored under the NCP. Data comparability and knowledge of analytical data variation (both within and between laboratories) are important in order to consolidate analytical data from various projects for the NCP. Besides providing knowledge on the interlaboratory variability of chemical measurements, which is critical for the comparison of analytical results from different research groups, interlaboratory comparison results also reveal procedural problems such as misidentification of compounds, concentration change in standard solutions over time, and un-optimized instrument conditions, which can be quickly corrected (Maier *et al.* 1993).

TABLE 1

Interlaboratory comparison exercises and the participating laboratories.

No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Lab 1	X	X	X	X	X	X			X		X			
Lab 2	X	X	X	X	X	X	X	X	X		X			X
Lab 3	X	X	X	X	X	X	X	X		X			X	X
Lab 4	X	X	X	X	X	X	X	X						X
Lab 5			X	X	X	X	X	X					X	X
Lab 6			X	X	X	X	X	X	X	X		X		
Lab 7			X	X		X			X					
Lab 8	X	X	X	X	X	X	X	X	X	X	X	X	X	
Lab 9			X	X	X	X	X	X						
Lab 10				X	X	X	X	X	X					
Lab 11									X*	X		X		
Lab 12		X	X											
Lab 13														X

* *Participated in HCB and HCHs only.*

1. *PCBs in polar bear (92/93)*
2. *OCs in polar bear (92/93)*
3. *PCBs in solution (93/94)*
4. *OCs in solution (94/95)*
5. *PCBs in seal blubber (94/95)*
6. *OCs in seal blubber (94/95)*
7. *PCBs in solution (95/96)*
8. *OCs in solution (95/96)*
9. *Co-planar PCBs in solution (94/95*
10. *Co-planar PCBs in solution (95/96)*

11 *PCDD/PCDFs in solution (94/95)*

12. *PCDD/PCDFs in solution (95/96)*
13. *PAHs in sediments (94/95)*
14. *Heavy metals in kidney (92/93)*

2. Interlaboratory Comparison Exercise

A list of interlaboratory comparisons and their participants is given in Table 1. Based on the sample matrices they routinely handle, the laboratories (Table 2) can be roughly divided into the following groups:

i) Laval University and Health Canada (handling human samples mainly blood and milk),
ii) Freshwater Institute, McGill University and Canadian Wildlife Service (terrestrial and marine biological samples),
iii) National Water Research Institute, Institute for Ocean Science and Atmospheric Environmental Services (abiotic samples), and
iv) contract laboratories such as AXYS Analytical Laboratories, Great Lakes Institute (for organic analysis), and Elemental Research Inc. and Analytical Services Laboratories (for inorganic analysis).

A complete list of all participating laboratories is provided in Table 2.

TABLE 2

Laboratories participating in AES interlaboratory QA/QC program.

1. Research Section
National Wildlife Research Centre
Environment Canada
100 Gamelin Blvd.
Hull, Quebec K1A 0H3

2. Laboratory Service
National Wildlife Research Centre
Environment Canada,
100 Gamelin Blvd.
Hull, Quebec K1A 0H3

3. Freshwater Institute
Department of Fisheries and Oceans
501 University Cres.
Winnipeg, Manitoba R3T 2N6
Freshwater Institute has three analytical groups, dealing with persistent organochlorines, PAHs and metals, respectively.

4. Centre de Toxicologie du Quebec
CHUL
2705 Boul. Laurier
Ste-Foy, Quebec G1V 4G2

5. Laboratory
National Water Research Institute
Environment Canada
867 Lakeshore Road
Burlington, Ontario L7R 4A6.
NWRI has four analytical groups, dealing with PCBs/OCs, PCDD/PCDFs, PAHs and metals, respectively.

6. Great Lakes Institute
University of Windsor
401 Sunset Avenue
Windsor, Ontario N9B 3P4

7. Atmospheric Environment Service
Environment Canada
4905 Dufferin Street
Downsview, Ontario, M3H 5T4

8. AXYS Analytical Services Ltd.
2045 Mills Road
Sidney, B.C.V8L 3S8

9. Centre for Nutrition and Environment of Indigenous Peoples
McGill University
21–111 Lakeshore
Ste-Anne-de-Bellevue, Que H9X 3V9

10. Medical Services Branch
Health Canada
Building 17
Tunney's Pasture
Ottawa, Ontario K1A 0L3

11. Institute of Ocean Sciences
Department of Fisheries and Oceans
9800 West Saanich St.
Sidney, B.C. V8L 4B2

12. Bedford Institute of Oceanography
Department of Fisheries and Oceans
Dartmonth, Nova Scotia B2Y 4A2

13. Elemental Research Inc.
#309–267 West Esplanada
North Vancouver, B.C. V7M 1A5

To include as many laboratories as possible for the intercomparisons, both standard solutions and real environmental samples were used as check samples. The use of standard solutions gave the participants a common base regardless of the types of samples they routinely handle and analyse. The standard solutions, however, do not reflect the matrix effects and the interferences encountered in the real samples. This was supplemented by the partially cleaned environmental samples such as polar bear fat extract and ringed seal blubber extract, as well as sediments and kidney samples.

Environmentally persistent chlorinated organic compounds such as polychlorobiphenyls (PCBs), organochlorine pesticides (OCs) and polychlorodibenzo-*p*-dioxins and polychlorodibenzofurans (PCDD/PCDFs) were the major chemical contaminants in the Arctic environment and its ecosystem (Barrie *et al.* 1992, Lockhart *et al.* 1992, Muir *et al.* 1992, Thomas *et al.* 1992, Kinloch *et al.* 1992). Most of the NCP projects were undertaken to study these contaminants in the Arctic (see Chapters 2, 3, and 4). The interlaboratory comparisons were therefore conducted mainly on these chlorinated organic contaminants, especially PCBs and OCs. Other interlaboratory comparisons conducted by the QA Program included polycyclic aromatic hydrocarbons (PAHs) in sediments, and heavy metals in caribou kidney. The interlaboratory comparisons conducted by the QA program did not include radioactive contaminants (RACs) and biomarkers as studies on RACs and biomarkers are carried out by only a small number of groups.

3. Results of Interlaboratory Comparisons

In all interlaboratory comparisons, participants were permitted to use their own analytical methods, and values were reported as the concentrations in the sample solutions provided, except for PAHs (in sediments) and for heavy metals (in kidney) where the sample weight was used. Relative standard deviations (%RSD), also called coefficient of variance (CV), are used in the following discussion (Taylor 1987).

Only the summary results of the intercomparisons are discussed in this Appendix. Detailed reviews and analyses of each interlaboratory comparison can be found in various QA Program reports submitted to the Department of Indian Affairs and Northern Development (DIAND 1997).

PCBs and OCs

PCBs and OCs are the two major groups of contaminants studied under the NCP, with large number of laboratories analysing these contaminants. Interlaboratory comparisons on two standard solutions, one OC solution and the other PCB solution, were conducted in 1994/95. The laboratories were asked to directly inject the solutions six times after proper dilution over three weeks' time (Sol: 94/95-dir), and perform a triplicate analysis (Sol: 94/95-prep). Another standard solution containing both OCs and PCBs was analysed in 1995/96 (Sol: 95/96). In addition, extracts of polar bear fat (Bear: 93/94) and ringed seal blubber (Seal: 94/95) were also analysed for interlaboratory comparison.

The %RSDs of the four intercomparisons (Sol: 94/95-prep, Sol: 95/96, Bear: 93/94, and Seal: 94/95) for OCs and for PCBs are summarized in Figure 1 and Figure 2, respectively. The average %RSD value of each intercomparison for OCs and for PCBs is about 20% to 30% (Figures 1 and 2). With a few exceptions, %RSDs were all below 40%, the majority between 15% and 30%. Minor components in matrix samples (extracts from polar bear and ringed seal) had %RSDs of up to 100%. %RSDs greater than 80%, such as PCB 149 (105%) and PCB 74 (112%) in polar bear extract, are not shown in these Figures. Dieldrin and oxychlordane in polar bear extract had %RSDs of 45% and 61%, respectively. The results of these two compounds were improved in seal extract a year later (21% and 30% respectively).

Two interlaboratory studies on PCBs and OCs, one for the Integrated Atmospheric Deposition Network (IADN) (Cussion 1992a,b, 1993a,b) and the other for the Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME) (de Boer and Wells 1995), were conducted at about the same time as this QA Program. The PCB and OC results obtained from these intercomparisons are comparable to those from the QA Program. IADN demonstrated %RSDs of 20% to 40% for the interlaboratory variations for PCBs and OCs in standard solutions, while QUASIMEME achieved a better data comparability among its member laboratories with %RSDs of PCBs and OCs at 10%–15% for the standard solutions, 20%–30% for biological tissues and 30%–60% for the sediments samples.

Replicate analyses were performed on the 1994/95 standard solutions (Sol: 94/95-dir and Sol: 94/95-prep) to determine both pooled within-laboratory standard deviation S_p and between- laboratory standard deviation S_b. Triplicate analysis (Sol: 94/95-prep) on the two standard solutions (PCBs and OCs) showed that within laboratory variation for PCBs and OCs in the standard solution was relatively small, often less than 10% with a few exceptions. The

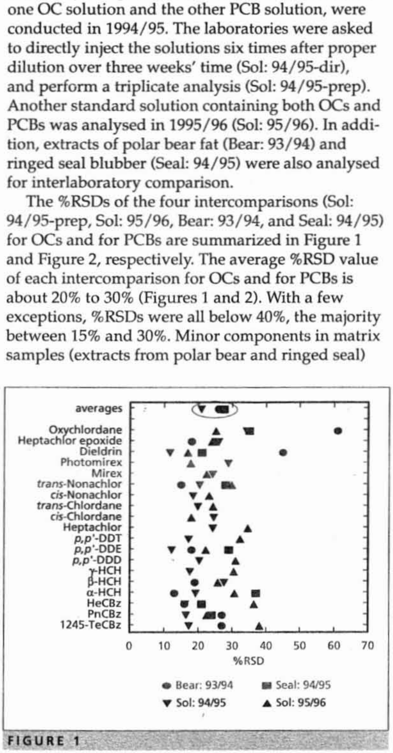

FIGURE 1

Relative standard deviations (%RSDs) of OCs in four intercomparisons. Sol: 94/95 — triplicate analysis results from intercomparison on OC standard solution in 1994/95; Sol: 95/96 — analytical results from intercomparison on PCB/OC standard solution in 1995/96; Bear: 93/94 — analytical results from intercomparison on polar bear fat extract in 1992/93; and Seal: 94/95 — analytical results from intercomparison on ringed seal blubber extract in 1994/95. All check samples were analysed in the same way as laboratory routine samples.

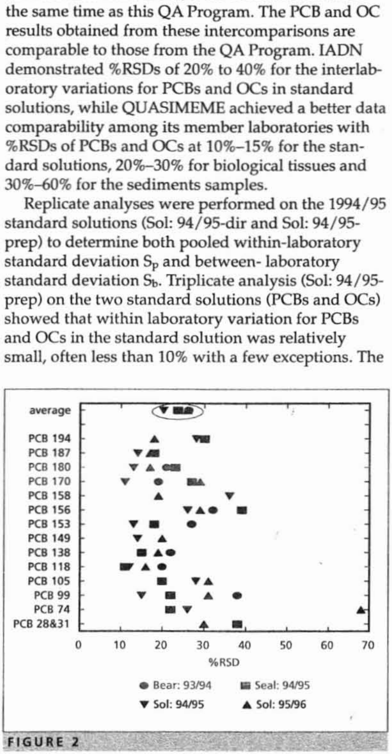

FIGURE 2

Relative standard deviations (%RSDs) of PCBs in four intercomparisons. Sol: 94/95 — triplicate analysis results from intercomparison on PCB standard solution in 1994/95; Sol: 95/96 — analytical results from intercomparison on PCB/OC standard solution in 1995/96; Bear: 93/94 — analytical results from intercomparison on polar bear fat extract in 1992/93; and Seal: 94/95 — analytical results from intercomparison on ringed seal blubber extract in 1994/95. All check samples were analysed in the same way as laboratory routine samples.

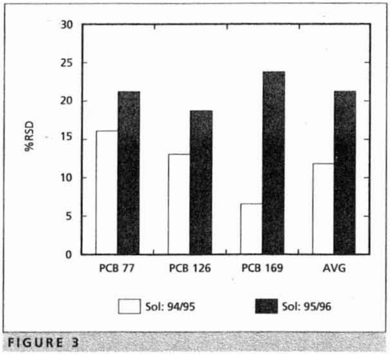

FIGURE 3

Relative standard deviations (%RSDs) of co-planar PCBs in two intercomparisons. Sol: 94/95 — analytical results from intercomparison on co-planar PCB standard solution in 1994/95; Sol: 95/96 — analytical results from intercomparison on co-planar PCB and PCDD/PCDF standard solution in 1995/96.

between-laboratory standard deviation S_b was close to the total interlaboratory variation S_t, and was about 2 to 5 times greater than within-laboratory standard deviation S_p in most cases (Table 3). Six replicate analyses in which the check solutions were injected directly into the instrument (Sol: 94/95-dir) produced a similar results (Table 4).

TABLE 3

Comparison of between-lab precision S_b and within-lab precision S_p for PCBs and OCs in solutions (triplicate analyses).

	Conc.	$S_{(t)}$	$S_{(b)}$	$S_{(p)}$	$S_{(b)}/S_{(p)}$
PCB 28	2.46	0.279	0.240	0.248	0.97
PCB 31	1.14	0.177	0.153	0.153	1.00
PCB 28/31	3.60	0.995	0.992	0.144	6.90
PCB 74	1.30	0.301	0.295	0.102	2.90
PCB 99	1.34	0.176	0.166	0.100	1.66
PCB 118	1.60	0.172	0.159	0.113	1.40
PCB 149	1.24	0.180	0.169	0.105	1.61
PCB 105	1.42	0.331	0.324	0.119	2.73
PCB 153	1.70	0.193	0.183	0.106	1.72
PCB 138	1.58	0.119	0.109	0.084	1.30
PCB 158	0.38	0.118	0.117	0.027	4.30
PCB 187	1.28	0.138	0.125	0.100	1.26
PCB 156	1.10	0.268	0.260	0.111	2.34
PCB 180	1.28	0.162	0.151	0.103	1.46
PCB 170	0.66	0.078	0.067	0.068	0.99
PCB 194	1.36	0.301	0.295	0.103	2.87
Tetrachlorobenzene	1.74	0.261	0.260	0.031	8.27
pentachlorobenzene	2.58	0.358	0.338	0.207	1.63
hexachlorobenzene	5.16	0.674	0.653	0.292	2.23
α-HCH	3.16	0.494	0.482	0.184	2.63
β-HCH	3.10	0.694	0.689	0.142	4.85
γ-HCH	2.16	0.329	0.324	0.102	3.17
p,p'-DDD	2.04	0.354	0.348	0.116	3.00
p,p'-DDE	5.22	0.554	0.537	0.235	2.28
p,p'-DDT	1.70	0.286	0.284	0.063	4.52
heptachlor	1.82	0.389	0.383	0.116	3.29
cis-chlordane	2.82	0.549	0.545	0.107	5.07
trans-chlordane	3.60	0.603	0.598	0.139	4.30
cis-nonachlor	1.60	0.241	0.227	0.138	1.64
trans-nonachlor	7.50	1.210	1.197	0.304	3.94
mirex	1.46	0.307	0.305	0.066	4.61
photomirex	3.68	0.829	0.824	0.158	5.23
dieldrin	2.84	0.307	0.300	0.112	2.68
heptachlor epoxide	5.72	1.168	1.163	0.180	6.47
oxychlordane	5.04	1.327	1.324	0.161	8.22

Co-planar PCBs and PCDD/PCDFs

In response to the concerns about the data comparability of these highly toxic contaminants, two interlaboratory comparisons on co-planar PCBs and PCDD/PCDFs using standard solutions have been conducted. In the first round (1994/95), the co-planar PCBs (at about 1 ng·µL^{-1} per compound) and PCDD/PCDFs (at about 40 pg·µL^{-1}) were in separate solutions; in the second round (1995/96) these chemicals were in a single solution at about 1 to 2 pg·µL^{-1} per compound. In 1994/95, the check sample was injected directly onto instruments with proper dilutions, while in the 1995/96 intercomparison the sample was treated through sample preparation procedures.

The %RSDs of the co-planar PCBs are shown in Figure 3. The average %RSDs of co-planar PCBs increased from about 12% in 1994/95 to 21% in 1995/96. The increased variability may be due to lower concentration level and sample preparation process in the second year. On the other hand, the average %RSDs of PCDD/PCDFs decreased from about 21% in 1994/95 to 9% in 1995/96 (Figure 4).

TABLE 4

Comparison of between-lab precision S_b and within-lab precision S_p for PCBs and OCs in solutions (six direct injection replicate analyses).

	Conc.	$S_{(t)}$	$S_{(b)}$	$S_{(p)}$	$S_{(b)}/S_{(p)}$
PCB 28	2.46	0.297	0.294	0.109	2.69
PCB 31	1.14	0.131	0.130	0.043	3.05
PCB 28/31	3.60	1.100	1.099	0.091	12.04
PCB 74	1.30	0.343	0.342	0.076	4.49
PCB 99	1.34	0.220	0.218	0.077	2.82
PCB 118	1.60	0.203	0.200	0.082	2.43
PCB 149	1.24	0.220	0.218	0.074	2.95
PCB 105	1.42	0.320	0.318	0.083	3.82
PCB 153	1.70	0.207	0.203	0.103	1.97
PCB 138	1.58	0.166	0.163	0.075	2.17
PCB 158	0.38	0.120	0.120	0.019	6.47
PCB 187	1.28	0.189	0.187	0.070	2.66
PCB 156	1.10	0.259	0.257	0.078	3.30
PCB 180	1.28	0.216	0.214	0.075	2.86
PCB 170	0.66	0.092	0.091	0.035	2.59
PCB 194	1.36	0.222	0.212	0.165	1.28
Tetrachlorobenzene	1.74	0.244	0.240	0.108	2.22
pentachlorobenzene	2.58	0.372	0.371	0.078	4.74
hexachlorobenzene	5.16	0.803	0.797	0.235	3.39
α-HCH	3.16	0.762	0.760	0.139	5.48
β-HCH	3.10	0.797	0.794	0.163	4.86
γ-HCH	2.16	0.322	0.320	0.084	3.83
p,p'-DDD	2.04	0.343	0.340	0.103	3.31
p,p'-DDE	5.22	1.231	1.228	0.207	5.92
p,p'-DDT	1.70	0.333	0.330	0.113	2.93
heptachlor	1.82	0.396	0.388	0.192	2.02
cis-chlordane	2.82	0.500	0.498	0.104	4.77
trans-chlordane	3.60	0.578	0.576	0.128	4.50
cis-nonachlor	1.60	0.245	0.241	0.105	2.30
trans-nonachlor	7.50	1.034	1.025	0.342	3.00
mirex	1.46	0.331	0.330	0.072	4.55
photomirex	3.68	0.841	0.840	0.091	9.20
dieldrin	2.84	0.253	0.245	0.157	1.56
heptachlor epoxide	5.72	0.947	0.937	0.335	2.80
oxychlordane	5.04	0.548	0.529	0.351	1.51

Combining the results from these two intercomparisons, we concluded that a range of %RSD at about 10%–20% exists for the interlaboratory analytical variation of co-planar PCBs and PCDD/PCDFs among the laboratories. The %RSDs for the real environmental samples could be larger due to matrix effects and interferences of other chemicals in the sample. The interlaboratory %RSDs of co-planar PCBs in spiked matrix from other programs were 20%–40% (de Voogt *et al.* 1994). Results of two interlaboratory comparisons on PCDD/PCDFs in fly ash and in egg extracts showed a %RSD of 20%–30% for the total toxicity equivalency in three fly ash extracts (van Bavel, *et al.* 1994), and about 15% in the eggs (Malisch *et al.* 1994).

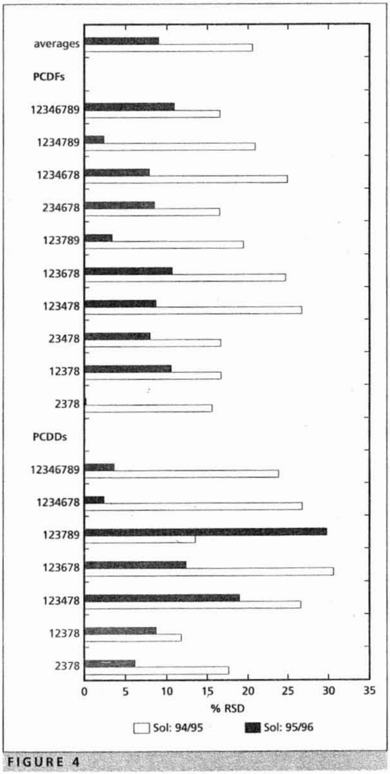

FIGURE 4

Relative standard deviations (%RSDs) of PCDD/PCDFs in two intercomparisons. Sol: 94/95 — analytical results from intercomparison on PCDD/PCDF standard solution in 1994/95; Sol: 95/96 — analytical results from intercomparison on co-planar PCB and PCDD/PCDF standard solution in 1995/96.

PAHs in sediments

Many NCP projects dealt with PAHs in various abiotic matrices such as sediments, air and water. An interlaboratory comparison on PAHs in sediments was conducted among four laboratories analysing abiotic samples. The study was aimed at evaluating the interlaboratory variability associated with PAH data. The mean values and %RSDs are shown in Figure 5. The %RSDs ranged from 25%–50% with exception of anthracene (%RSD = 62), which had a very low concentration level in the sample. The average %RSD was 41%. In comparison, the average interlaboratory %RSDs of PAHs in solutions were around 20% (Law and Klungsøyr 1995) and 24% (Cussion 1992c) from other programs.

Toxic heavy metals

Besides interlaboratory comparisons on various organic contaminants, efforts were also made to evaluate the interlaboratory variation of toxic heavy metals for the NCP. The intercomparison focused on three most common toxic trace elements in the Canadian Arctic: cadmium, mercury, and lead. A homogenized caribou kidney sample was chosen as the check sample. The results showed that the interlaboratory variation of the three elements was satisfactory (Figure 6). For the element with the highest concentration (cadmium) the %RSD was 4.5%. The variability increased when the concentration decreased (%RSD was 11% for mercury (7.6 ppm) and 36% for lead (0.6 ppm)). The results were

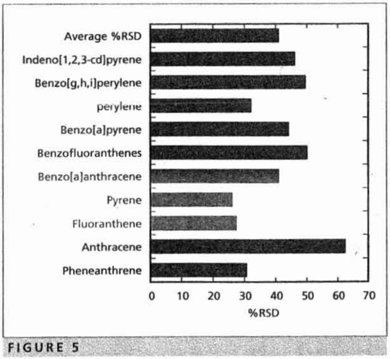

FIGURE 5

Relative standard deviations (%RSDs) of PAHs in sediments check sample. The concentration of anthracene in the sample was close to the detection limit for most laboratories.

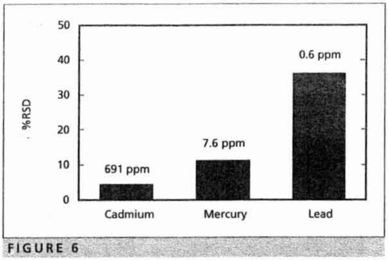

FIGURE 6

Relative standard deviations (%RSDs) of three trace metals in caribou kidney check sample. The concentration of lead in the sample was close to the detection limit for a few laboratories.

comparable to the study conducted by Cussion, which had interlaboratory %RSDs less than 10% for high and medium concentration and up to 70% for the low concentrations of metals in standard solutions (Cussion 1992d, 1993c).

4. Other Interlaboratory Comparisons

Besides participating in QA program, and its interlaboratory comparisons, many laboratories have been engaged in various other interlaboratory comparison exercises and in sample exchanges among laboratories. The participation in other intercomparison programs ensures the data comparability of the NCP to other related studies.

The first round of the interlaboratory comparison of toxaphene in cod liver oil (NIST SRM 1588), involving 15 laboratories from four countries, was conducted by Health Canada in 1993 (Andrews *et al.* 1995). A mean value of 4 ppm with 50% %RSD was found for the total toxaphene level in the oil. Lab 3, Lab 8 and Lab 10 were part of the participating laboratories.

A series of interlaboratory comparisons of PCBs, OCs, PAHs, and trace metals was conducted on a yearly basis using standard solutions to support the Integrated Atmospheric Deposition Network (IADN). Six laboratories from Canada and the US participated in the comparisons, of which three participated in the QA (Labs 3, 5 and 7) (IADN 1997).

Another important international interlaboratory study among marine laboratories worldwide has been conducted by the International Council for the Exploration of the Sea (ICES). The study was conducted in different steps, starting with standard calibration solutions, followed by spiked solutions and finally the marine matrices (de Boer and Wells 1995). Some of the QA program participants are also engaged in the ICES interlaboratory comparisons (Labs 3, 8, and 12).

5. Overall NCP Analytical Data Quality

Analytical data quality can be affected by all three stages of a measurement: sampling, analysis and data reporting. Under the current NCP funding structure, projects were managed by individual project leaders. Each project leader/management was responsible for the project design, sampling plan, analytical work and data validation/reporting that were either conducted in house or contracted to a third party. Analytical data were independently reviewed in some cases by the QA/QC officer if the analytical work was contracted out to a third party.

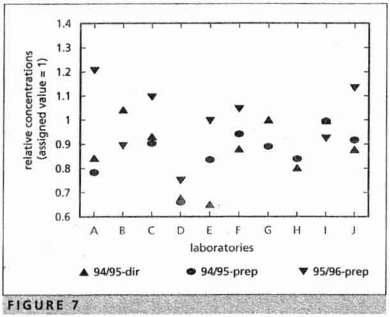

FIGURE 7

Relative concentration of PCBs among participants. Reported values from each laboratory were normalized against the assigned values for all three intercomparisons. 94/95-dir: — mean values of six replicate direct injections of the PCB standard solution in 1994/95. 94/95-prep: — mean values of triplicate analysis of the PCB standard solution in 1994/95. 95/96-prep: — mean values of analytical results of the PCB/OC standard solution in 1995/96.

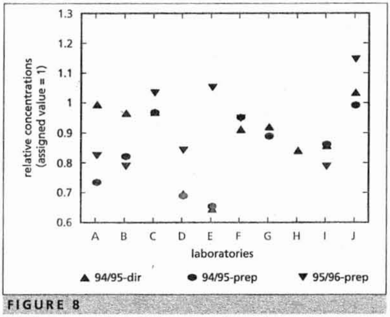

FIGURE 8

Relative concentration of OCs among participants. Reported values from each laboratory were normalized against the assigned values for all three intercomparisons. 94/95-dir: — mean values of six replicate direct injections of the OC standard solution in 1994/95: 94/95-prep: — mean values of triplicate analysis of the OC standard solution in 1994/95. 95/96-prep: — mean values of analytical results of the PCB/OC standard solution in 1995/96.

Sampling procedure includes sampling plan, sample collection, on-site sample handling and storage, sample transportation to the laboratory, sample treatment if necessary, and storage in the laboratory prior to analysis. Intercomparison on sampling procedures among different research groups was not conducted for the NCP. However, a survey on sampling procedures undertaken for the NCP (Zhu 1995b) showed that most abiotic samples were collected by scientists themselves and/or under their strict supervision. Field duplicates were collected in a few cases. Collections of biological samples such as fish, mammals, birds and human blood were largely the result of cooperation of hunters and helpers living in the north. Field workers were instructed on how to collect and handle samples. Biological variations were controlled in a few cases by careful design of sampling programs for age, sex, season, time, location and health condition of each individual sample.

All laboratories participated in the QA Program have well-documented analytical procedures. Contract laboratories in the QA program are accredited by the Canadian Association for Environmental Analytical Laboratories Inc. With few exceptions, all analytical data for organic contaminants and metals were generated by laboratories participating in the QA Program.

The interlaboratory variations have been discussed in the previous section. In general, the interlaboratory %RSDs of 20% or less can be estimated for analytical data of co-planar PCBs and PCDD/PCDFs, and about 10% for toxic heavy metals. PAH data had relatively large interlaboratory variation (%RSDs around 30%–50%). Interlaboratory %RSDs for PCB and OC data were mostly around 15%–30%, with some spreading to 10% and 40% respectively. Compounds with low concentration in the samples had larger variation.

In addition to these general findings, the following cautions may be taken when using PCB and OC data. PCBs and OCs data from Lab D were consistently biased low compared with others (Figures 7 and 8). The OC data from Lab D were improved in the 1995/96 intercomparison (95/96-prep, Figure 8). PCB values in 1994/95-dir and OC values in 1994/95 intercomparisons from Lab E were also low (Figures 7 and 8).

6. Analytical Data for Temporal and Spatial Trends Studies

The comparison of analytical data in a time dimension (temporal trends) or geographic dimension (spatial trends) is always challenging, and sometimes rather difficult because of different analytical methods, as well as sampling methods that are employed over time by the same laboratory or among different laboratories.

To solve the data comparability problems, most temporal studies in this report were conducted by analysing the archived samples collected over years. Other temporal studies were concluded by reanalysing a few achieved samples to confirm the comparability of "old" and "new" data.

Spatial studies on contaminants in a particular species or matrix (e.g., polar bear) were based mainly on a single laboratory's data. However, in a few cases, some studies were based on analytical data from several laboratories. If the laboratories all participated in the QA Program, the interlaboratory variations discussed above can be applied when interpreting the analytical results. There is insufficient information to estimate the data comparability of a study that linked the analytical data generated by NCP laboratories and data from other laboratories/programs.

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Appendix III: Human Health Risk Assessments

TABLE 1

Summary of Human Health Risk Assessments conducted in the Canadian North, based on data collected from 1990 to 1996 (HC=Health Canada, PAH=polyaromatic hydrocarbon, RMWI=recommended maximum weekly intake (based on lifetime intake), wk=week)*.

Location (indicates physical area or community closest to sampling site)**	Species (Tissue)**	Contaminant(s)	Consumption Advice*	HC Assessment Date	Sampling Date(s)	Initiating Department/ Agency
Inuvik Region, NWT						
Lac Ste Thérèse, Deline (Fort Franklin)	Lake Trout, Lake Whitefish, Longnose Sucker, Northern Pike, Walleye (kidney, liver, muscle)	metals	Advice was issued to Deline (Fort Franklin) residents at a community meeting in the form of RMWIs based on levels of mercury in the kidney, liver, muscle for all fish. There was little concern in the community due to the fact few people fish the lake.	1 March 1993	1975, 1980, 1992	1
Keller Lake, Lac Tache, Lac Ste Thérèse, Tseepantee Lake (Fort Norman)	Walleye, Lake Whitefish, Northern Pike, Lake Trout (liver, muscle)	metals	Advice was issued in the form of RMWIs based on levels of mercury in muscle (m) and liver (l) from: *Lac Ste Therese* Lake Trout m=180 g/wk (6 oz/wk), l=55 g/wk (2 oz/wk) Northern Pike m=225 g/wk (7 oz.wk), Walleye m=140 g/wk (5 oz/wk) *Tseepantee Lake* Walleye m=210 g/wk (7 oz/wk) All other fish species and tissues from this health assessment would not pose a health concern to consumers.	13 Jan. 1995	1992, 1993	2
Beaufort Sea	Greenland Halibut (muscle, skin)	organochlorines	No consumption advice as levels of organochlorines in fish tissues would not pose a health concern to consumers.	11 Dec. 1995	1994	1, 2
South Beaufort Sea (Paulatuk)	Beluga (blubber, kidney, liver, muktuk, muscle)	organochlorines, metals	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1993-1994	1, 2
South Beaufort Sea (Tuktoyaktuk)	Arctic Char, Whitefish (muscle, skin)	organochlorines	No consumption advice as levels of organochlorines in fish tissues would not pose a health concern to consumers.	11 Dec. 1995	1990	1, 2
Mackenzie Delta	Beluga (blubber), Ringed Seal (kidney, liver, muscle)	organochlorines	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1983, 1987-1988, 1993	1, 2
East Whitefish Station	Beluga (blubber, kidney, liver, muktuk, muscle)	organochlorines, metals	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1989-1990, 1993-1994	1, 2
Kendall Island	Beluga (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1989-1990	1, 2
Hendrickson Island	Beluga (blubber, kidney, liver, muktuk, muscle)	organochlorines, metals	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1993-1994	1, 2
Shingle Point	Beluga (kidney, liver, muktuk, muscle)	metals	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1993-1994	1, 2

continued on next page

Location (indicates physical area or community closest to sampling site)**	Species (Tissue)**	Contaminant(s)	Consumption Advice*	HC Assessment Date	Sampling Date(s)	Initiating Department/ Agency
Mackenzie Region, NWT						
Slave River (Fort Smith), Alexie/Chitty Lake, Leland Lake	Burbot (liver), Walleye (whole fish)	organochlorines	No consumption advice issued as levels of organochlorines in all fish tissues would not pose a health concern to consumers.	3 Feb. 1992	1990, 1991	1, 2
Slave River (Fort Smith), Alexie/Chitty Lake, Leland Lake	Burbot (liver), Walleye (whole fish)	organochlorines	Advice was issued in the form of a general recommendation to people who consume burbot liver from the Fort Smith area to limit consumption to no more than one burbot liver per week, or 52 livers per year, based on the RMWI for toxaphene levels in burbot liver.	30 Oct. 1992	1991-1992	1, 2
Slave River (Fort Smith), Alexie/Chitty Lake, Leland Lake	Burbot, Walleye, Northern Pike, Whitefish (muscle, liver, bile, whole fish)	organochlorines, metals, PAHs	Assessment not complete.	incomplete	1989-1994	1, 2, 3
Kam Lake	Walleye, Lake Whitefish, Northern Pike, Trout, Longnose Sucker, Burbot (muscle)	metals	No consumption advice as levels of metals in fish muscle would not pose a health concern to consumers.	March 1994	1990-1991	1, 2
Trout Lake	Walleye, Lake Whitefish, Northern Pike, Trout, Longnose Sucker, Burbot (muscle)	metals	No consumption advice as levels of metals in fish muscle would not pose a health concern to consumers.	21 March 1994	1990-1991	1, 2
Giauque Lake, Thistlewaite Lake	Lake Trout, Lake Whitefish, Northern Pike, Longnose Sucker, and round Whitefish (muscle, liver, kidney)	metals	Advice was issued in the form of RMWIs based on levels of mercury in muscle (m), liver (l), kidney (k) from: *Giauque Lake* Lake Trout m = 85 g/wk (3 oz/wk), l=42 g/wk (2 oz/wk), k=48 g/wk (2 oz/wk) Lake Whitefish m=226 g/wk (8oz/wk), l=63 g/wk (2 oz/wk), k=80 g/wk (3 oz/wk) Longnose sucker m=217 g/wk (8 oz/wk), l=263 g/wk (9 oz/wk), k=371 g/wk (12 oz/wk) Northern Pike m=123 g/wk (4 oz/wk), l=88 g/wk (3 oz/wk), k=129 g/wk (4 oz/wk) Round Whitefish m= 500 g/wk (16 oz/wk), l=370 g/wk(12 oz/wk), k=298 g/wk (10 oz/wk) *Thistlewaite Lake* Lake Trout m=561 g/wk (19 oz/wk), l=394 g/wk (13 oz/wk), k=249 g/wk (8 oz/wk) Northern Pike m=525 g/wk (18 oz/wk) All other fish species and tissues from this health assessment would not pose a health concern to consumers.	April 1994	1992	2

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Location (indicates physical area or community closest to sampling site)**	Species (Tissue)**	Contaminant(s)	Consumption Advice*	HC Assessment Date	Sampling Date(s)	Initiating Department/ Agency
Great Slave Lake (Fort Resolution)	Walleye, Northern Pike, Trout, Lake Whitefish, Burbot, Longnose Sucker (muscle)	metals	No consumption advice was issued as levels of metals in fish muscle would not pose a health concern to consumers.	16 Feb. 1994 and 10 May 1994	1992, 1993	2
Deer Pass Bay (Port Radium/Echo Bay)	Lake Trout, Lake Whitefish, Arctic Grayling (muscle, liver)	metals, radionuclides	No consumption advice was issued as levels of metals and radionuclides in fish tissues would not pose a health concern to consumers.	23 March 1995	1993	1, 2
Liard River (Fort Liard)	Walleye, Northern Pike, Mountain Whitefish, Longnose Sucker (muscle, whole fish)	organochlorines, PAHs	No consumption advice was issued as levels of organochlorines and PAHs in fish tissues would not pose a health concern to consumers.	8 Aug. 1994	1991-1993	1, 2
Liard River (Fort Liard)	Walleye, Northern Pike, Mountain Whitefish, Longnose Sucker, Burbot (liver, whole fish)	metals, organochlorines, PAHs	Assessment not complete.	ongoing	1991-1993	1, 2
Lac a Jacque (Fort Good Hope)	Walleye, Northern Pike, Lake Whitefish	metals	Assessment not complete.	assessment not complete	1994	1, 2
Yellowknife Bay, Back Bay of Great Slave Lake	Whitefish, Walleye, Northern Pike, Longnose Sucker (muscle)	metals	No consumption advice as levels of metals in fish tissue would not pose a health concern to consumers.	30 July 1996	1992-1994	1, 2
Hay River	Walleye, Northern Pike	metals, PAHs	Assessment not complete.	ongoing	1994-1995	1, 2, 3, 4
Inuvik and Kitikmeot Regions, NWT						
Beverly (Fort Smith), Victoria Island (Cambridge Bay), Bluenose (Inuvik), North Baffin (Pond Inlet), Boothia (Taloyoak)	Caribou (fat, kidney, liver, muscle)	organochlorines, radionuclides, metals	Advice was issued to limit consumption to one caribou kidney per week, or up to 50 per year, based on the RMWI for cadmium in caribou kidney.	26 April 1995	1993-1994	3, 5
Keewatin Region, NWT						
Baker Lake (Lutselk'e/Snowdrift)	Caribou (kidney, liver, muscle)	radionuclides	No consumption advice was issued as levels of radionuclides in caribou tissues would not pose a health concern to consumers.	14 Dec. 1993	1991-1992	5, 6
West Hudson Bay (Arviat)	Beluga (blubber), Ringed Seal (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1986, 1993-1994 and 1994	1, 2
Peter Lake (Rankin Inlet)	Charr and Lake Trout	organochlorines, metals	Assessment not complete.	upcoming	1995-1996	2
Repulse Bay (Eastern Arctic)	Beluga (kidney, liver, muktuk, muscle)	metals	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1993-1994	1, 2

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Location (indicates physical area or community closest to sampling site)[**]	Species (Tissue)[**]	Contaminant(s)	Consumption Advice[*]	HC Assess-ment Date	Sampling Date(s)	Initiating Depart-ment/ Agency
Keewatin and Baffin Regions, NWT						
South Baffin (Cape Dorset, Lake Harbour), Southampton Island (Coral Harbour), Qamanirjuaq (Arviat)	Caribou (kidney, liver)	organochlorines, metals	Advice was issued to limit consumption to one caribou kidney per week, or up to 50 per year, based on the RMWI for cadmium in caribou kidney.	17 May 1993	1992	3
Amituk Lake (Resolute)	Arctic Char (liver, muscle)	metals	No consumption advice was issued as levels of metals in fish tissues would not pose a health concern to consumers.	21 July 1993	1992	1, 2
Baffin Region, NWT						
South Baffin (Lake Harbour)	Caribou (fat, kidney, liver, muscle)	organochlorines,	No consumption advice was issued as levels of organochlorines in caribou tissues would not pose a health concern to consumers.	18 Jan. 1994	1992	3
Frobisher Bay, Baffin Island (Iqaluit)	Iqaluit Clam	organochlorines, metals, PAHs	No consumption advice was issued as levels of organochlorines, metals, and PAHs would not pose a health concern for consumers.	8 Aug. 1996	1995	4
Broughton Island, Pond Inlet, Resolute, Lake Hazen (Ellesmere Island)	Arctic Char (muscle, skin)	organochlorines	No consumption advice as levels of organochlorines in fish tissues would not pose a health concern to consumers.	11 Dec. 1995	1987, 1989, 1990	1, 2
Eureka, Ellesmere Island	Ringed Seal (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.[†]	11 Dec. 1995	1994	1, 2
Resolute, Cornwallis Island	Ringed Seal (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.[†]	11 Dec. 1995	1994	1, 2
Cumberland Sound	Greenland Halibut (muscle, skin)	organochlorines	No consumption advice as levels of organochlorines in fish tissues would not pose a health concern to consumers.	11 Dec. 1995	1994	1, 2
Arctic Bay (Baffin Island)	Ringed Seal (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.[†]	11 Dec. 1995	1994	1, 2
Cumberland Sound (Pangnirtung)	Beluga (liver, blubber), Narwhal (liver, blubber), Ringed Seal (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.[†]	11 Dec. 1995	1983-1986, 1993-1994	1, 2
Hudson Bay (Sanikiluaq)	Beluga (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.[†]	11 Dec. 1995	1994	1, 2

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Location (indicates physical area or community closest to sampling site)**	Species (Tissue)**	Contaminant(s)	Consumption Advice*	HC Assessment Date	Sampling Date(s)	Initiating Department/ Agency
Northwest Territories						
Southampton Island, Beverly, Victoria Island, Porcupine, Banks Island	Caribou (kidney, liver), Muskoxen (kidney, liver)	metals	Advice was issued to limit consumption to one caribou kidney per week, or up to 50 per year, based on the RMWI for cadmium in caribou kidney.	14 Dec. 1992	Various years for each herd: 1978, 1985-1990	3, 4, 6
Arviat, Holman, Sanikiluaq, Taloyoak, Tuktoyaktuk	Canada Goose, Lesser Snow Goose, King Eider, Common Eider (muscle)	organochlorines metals	No consumption advice was issued as levels of organochlorines in bird tissues would not pose a health concern to consumers. Lead shot should be removed from birds as soon as possible.	18 Jan. 1994	1985-1991	4
NWT	waterfowl and other game	organochlorines metals	No consumption advice was issued as levels of organochlorines and metals in waterfowl and other game would not pose a health concern to consumers.	7 Jan. 1997	1993	1, 4
Yukon Territory						
Lake Laberge	Burbot (liver), Lake Trout (muscle, skin)	organochlorines	Advice was issued to limit consumption of lake trout to 2 meals per month and refrain from eating burbot liver based on the RMWIs for toxaphene in Trout muscle and Burbot liver.	11 Dec. 1995	1990, 1992	1, 4, 7
Atlin Lake	Burbot (liver)	organochlorines	Advice was issued to refrain from eating burbot liver based on the RMWI for toxaphene in Burbot liver.	11 Dec. 1995	1992	1, 4, 7
Yukon Territory	Caribou herds- Bonne Plume, Nahanni, Porcupine, Forty-Mile, Wolf Lake, Finlayson, Tay (kidney, liver, muscle)	organochlorines metals	Advice was given in the form of a chart with the maximum number of organs recommended for consumption per year. Finlayson and Tay herds recommended maximum consumption of 8 and 7 kidneys and 5 and 4 livers per year due to cadmium levels. No consumption advice for meat.	1993-1995	1993-1995	1, 4, 7
Yukon Territory	moose (kidney, liver, muscle)	organochlorines metals	Advice was issued recommending a maximum consumption of 1 kidney and 1 liver per year based on the RMWIs for cadmium in kidney and liver of moose. No consumption advice for meat.	1993-1995	1993-1995	1, 4, 7
Yukon Territory	sheep, goat, beaver, porcupine, snowshoe hare (kidney, liver, muscle)	organochlorines metals	Advice was given, based on the levels of cadmium, in the form of a chart with the recommended maximum number of organs that can safely be consumed per year. No consumption advice for meat.	1993-1995	1993-1995	1, 4, 7
34 Yukon Lakes/Rivers††	Trout, whitefish, grayling, pike, and salmon where available (muscle)	organochlorines	No consumption advice was issued as the levels of organochlorines in fish tissues would not pose a health concern to consumers.	1992-1995	1992-1995	1, 4, 7
Quebec Region						
East Hudson Bay (Inukjuak)	Ringed seal (blubber), Walrus (blubber)	organochlorines	No consumption advice was issued based on risk management considerations.†	11 Dec. 1995	1993	1, 2

* Compiled from information provided by GNWT-Health and Social Services and Yukon Territories Contaminants Committee.

** In general, the species and locations listed for the NWT were collected and chosen opportunistically by a number of agencies to study the health of Arctic wildlife populations. Because these species/tissues are also consumed by Northerners, the contaminant results from these collections were submitted to Health Canada for assessment.

† Health Canada's recommendations were reviewed by federal and territorial agencies, and aboriginal organizations. Based on insufficient information about the risks and the significant known benefits of traditional foods, people were not advised to alter their current diets. Further details on this case are provided in CACAR 4.4.

†† Lakes: Atlin, Aishihix, Bennett, Big Kalzas, Breaburn, Dezdeash, Dragon, Ethel, Fish, Fox, Francis, Hanson, Jackfish, Kathleen, Kluane, Klukshu, Kusawa, Laberge, Little Atlin, Little, Salmon, Mayo, Marsh, Quiet, Schwatka, Tagish, Tatlmain, Teslin, Watson, Yukon/Klondike, Coal, Big Whitefish; Rivers: Porcupine, Yukon/White, Klukshu.

1-Department of Indian Affairs and Northern Development, 2-Fisheries and Oceans Canada, 3-Government of Northwest Territories-Renewable Resources, 4-Department of the Environment, 5-Health Canada, 6-University of Saskatchewan, 7-Yukon Territory Government.

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