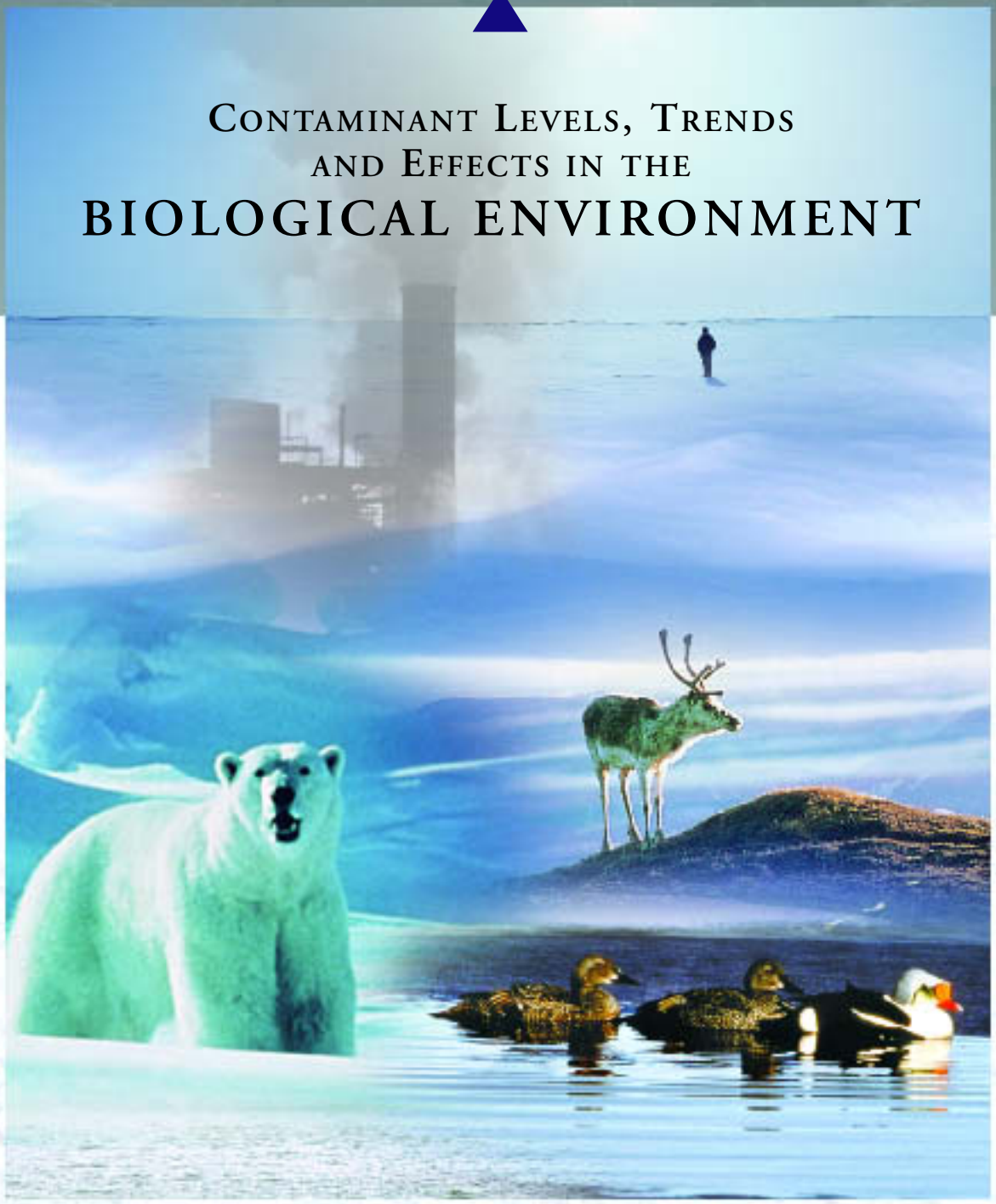
NORTHERN CONTAMINANTS PROGRAM

CANADIAN ARCTIC CONTAMINANTS ASSESSMENT REPORT II

CONTAMINANT LEVELS, TRENDS AND EFFECTS IN THE BIOLOGICAL ENVIRONMENT



Indian and Northern Affairs Canada
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Canada


Chukchi Sea
Arctic Ocean
Beaufort Sea
Sachs Harbour
(Ikaahuk)
Old Crow
Tuktoyaktuk
Aklavik
(Akłarvik)
Inuvik
(Inuuvik)
Holman
(Uluqsaqtuuq)
Fort McPherson
(Teetł'it Zheh)
Tsiigehtchic
(Tsııgehtshık)
Paulatuk
(Paulatuuq)
Colville Lake
(K'áhbamı́tué)
Fort Good Hope
(Rádeyı̨ı̨kǫ́é)
Dawson
Beaver Creek
Keno Hill
Mayo
Pelly Crossing
Norman Wells
(Tłegǫ̀hı̨)
Kugluktuk
(Coppermine)
Iqaluktuutiaq
(Cambridge Bay)
Umingmaktuuq
Faro
Carmacks
Ross River
Déline
(Dél̨ne)
Tulita
(Tulit'a)
Qinguaq
(Bathurst Inlet)
Haines
Junction
Whitehorse
Wrigley
(Pedzéh Kı̨)
Rae Lakes
(Gameti)
Snare Lakes
(Wekweti)
Carcross
Teslin
Wha Ti
(Wha Tı)
Rae-Edzo (Behchokǫ̀-Edzo)
Watson Lake
Fort Simpson (Łíídlı̨ Kúé)
Jean Marie River
(Tthek'éhdélı̨)
Yellowknife (Sǫ̀mbak'è)
Nahanni Butte (Tthenáágó)
Fort Liard
Fort Providence
(Zhahtı Kúé)
Lutselk'e
(Łútsëlk'é)
Hay River
(Xátł'odehchee)
Fort Resolution
(Denınu Kúé)
Fort Smith
(Tthebacha)


Alert
Ausuittuq
(Grise Fiord)
Resolute
(Qausuittuq)
Ikpiarjuk/Tununirusiq
(Arctic Bay)
Mittimatalik
(Pond Inlet)
Kangiqtugaapik
(Clyde River)
Baffin Bay
Taloyoak
Igloolik
Qikiqtarjuaq
(Broughton Island)
Uqsuqtuuq
(Gjoa Haven)
Kugaaruk
(Pelly Bay)
Sanirajak
(Hall Beach)
Pangnirtung
Naujaat
(Repulse Bay)
Iqaluit
Kinngait
(Cape Dorset)
Qamanittuaq (Baker Lake)
Salliq
(Coral Harbour)
Kimmirut
(Lake Harbour)
Igluligaarjuk (Chesterfield Inlet)
Kangiqliniq (Rankin Inlet)
Tikirarjuaq (Whale Cove)
Ivujivik
Salluit
Kangiqsujuaq
Quaqtaq
Akulivik
Kangirsuk
Arviat
Puvirnituq
Aupaluk
Kangiqsualujjuaq
Tasiujaq
Nain
Kuujjuaq
Hudson Bay
Inukjuak
Hopedale
Makkovik
Postville
Rigolet
Umiujaq
Sanikiluaq
Happy Valley/Goose Bay
Kuujjuarapik

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Contaminant Levels, Trends and Effects in the BIOLOGICAL ENVIRONMENT

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Executive Summary

The Northern Contaminants Program (NCP) was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in fish and wildlife species that are important to the traditional diets of northern Aboriginal peoples. Early studies indicated that there was a wide spectrum of substances — persistent organic pollutants, heavy metals, and radionuclides — many of which had no Arctic or Canadian sources, but which were, nevertheless, reaching unexpectedly high levels in the Arctic ecosystem.

Under the first phase of the NCP (NCP-I), research was focussed on gathering the data required to determine the levels, geographic extent, and source of contaminants in the northern atmosphere, environment and its people, and the probable duration of the problem. Results generated through NCP-I were synthesized and published in 1997 in the *Canadian Arctic Contaminants Assessment Report (CACAR)*.

In 1998, the NCP began its second phase (NCP-II), which will continue until March 2003. NCP-II focussed upon questions about the impacts and risks to human health that may result from current levels of contamination in key Arctic food species as well as determining the temporal trends of contaminants of concern in key indicator Arctic species and air. It addressed these issues under a number of subprograms: human health; monitoring the health of Arctic peoples and ecosystems and the effectiveness of international controls; education and communications; and international policy.

NCP Phase II identified three priority areas of study for monitoring of the biological environment: temporal trend studies; monitoring and surveys of traditional/country foods; and wildlife health. The purpose of this, the biological environment technical report in the CACAR-II series, is to review the information that addresses these priority areas and to assess the current state of knowledge on the spatial and temporal trends of contaminants and the biological effects in Canadian Arctic biota. The CACAR-II series consists of a Highlights report and four technical reports: human health, biological environment, physical environment and knowledge in action.

The first Canadian Arctic Contaminants Assessment Report (CACAR) on contaminants in biota (Chapter 3 Ecosystem Uptake and Effects) compiled and assessed contaminant data generated prior to 1997. Most of these data were generated under Phase I of the Northern Contaminants Program (NCP) of the Department of Indian Affairs and Northern Development (DIAND). This report made a series of conclusions and identified knowledge and data gaps on the spatial and temporal trends and biological effects of anthropogenic contaminants in Canadian Arctic biota. These knowledge gaps provided the direction for research under Phase II of the NCP (1997–2003). Highlights of the research conducted on Arctic biota under NCP Phase II include: improved temporal trend data sets of organic and metal contaminants; expanded spatial trends of mercury and organochlorines (OCs) in freshwater and marine biota; new knowledge of food web dynamics of organic and metal contaminants; and the identification and measurement of a suite of new anthropogenic contaminants. However, many knowledge gaps identified in the first CACAR report have received less attention or have remained elusive and new questions and concerns have arisen in the past five years. These include: the cause of high and variable levels of mercury in freshwater fish; the regional and species differences in temporal trends of mercury; the lack of information on spatial and temporal trends of new contaminants and metals other than mercury, cadmium and lead; and the impact of contaminants on the health of Arctic biota.

Approximately 100 "legacy" organochlorines (OCs) [including PCBs, DDTs, chlordanes, dieldrin, hexachlorocyclohexanes (HCHs) and chlorobenzenes (CBz)] have been measured in most studies funded under NCP Phase II. Although some studies have measured up to 25 metals, most studies have focussed on mercury, cadmium and selenium, and information on other metals remains a knowledge gap. The quality of the contaminant data generated under NCP Phase II is generally very good and the quality assurance program is a real strength of the NCP program, providing additional confidence in the results and conclusions of this assessment. All laboratories generating data in this assessment participated in the interlaboratory quality assurance project

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funded by NCP in the past five years. New information on toxaphene has been generated for freshwater fish, seabirds and marine mammals, addressing a data gap identified in the first CACAR report. However, the quality assurance study found greater variability between labs for toxaphene and specific congeners than for other OCs.

Several new chemicals have been found in Arctic biota. The number of new chemicals measured has increased significantly during NCP Phase II. These data have provided more comprehensive coverage for Arctic organisms, including temporal trend data sets, compared with the first CACAR report but the scope is very limited in comparison to legacy OCs. Information on polybrominated diphenyl ethers (PBDEs) has been generated for both freshwater and marine organisms while measurements of short chain chlorinated paraffins and chlorinated naphthalenes (PCNs) were made in beluga and ringed seals. More detailed measurements were also made on the current use pesticides lindane and endosulfan. Investigations showed only very low or non-detectable levels of tributyltin and synthetic musks in eastern Arctic mussels, char and seals. Perfluorinated octane sulfonic acid (PFOS), a chemical that was not expected to be found in the Arctic due to its physical-chemical properties, has also been found in Canadian Arctic biota. The discovery of such a chemical in Arctic biota at significant concentrations raises questions about sources and pathways of these and other polar chemicals. Although concentrations of many of these new chemicals are lower than legacy POPs (PCBs, DDT, etc.) there is concern because PBDEs are increasing in concentrations while PFOS appears to be present at relatively high levels in polar bear liver from Alaska compared to other OCs. Current NCP projects are measuring PFOS in polar bears and other marine biota but results are not yet available. These findings illustrate the need for continued vigilance given that there are thousands of chemicals in commerce in the circumpolar Arctic and only a fraction have been assessed for their persistence, bioaccumulation and long range transport potential. There is a need to assess the PBDEs, PCNs and PFOS, as well as other new contaminants, in a wider range of species, over a broader geographic range, their food web behaviour and possible biological effects.

One of the most significant achievements of NCP Phase II was the development of temporal trend data sets. These now cover a range of species and chemicals (OCs and metals), including new chemicals such as PBDEs. Critical to these studies have been tissue archives, supported under NCP-I, which have proved invaluable and allowed the development of temporal trend data sets for chemicals that have only recently been detected in the Arctic. Many of these studies have provided data sets that stretch from the 1970s to 2000 and have addressed a major data gap identify for both the Canadian and circumpolar Arctic. These world-class data sets provide an excellent comparison to comprehensive long-term studies of contaminants in fish from the Great Lakes and northern Sweden and provide valuable information on the response of Arctic food webs to the banning of chemicals. Continued support of tissues archives and long-term monitoring programs should be a priority.

There is solid evidence that mercury has increased in Canadian Arctic animals from pre-industrial times to the present. For example, mercury has increased from 10–17-fold in beluga in the Mackenzie Bay area from about 1450–2000. However, temporal trend data sets of mercury in these animals over the past 20–30 years show conflicting trends. Mercury has increased 4-fold in the past ten years in beluga from the Beaufort Sea and 2.5-fold western Hudson Bay but not significantly at any other locations. No increases have been observed in cadmium in beluga over the same period. It has been suggested that increased temperatures in the Mackenzie River region have liberated mercury and may explain the increases in mercury concentrations observed in the Beaufort Sea beluga. Mackenzie River burbot are the only freshwater fish species that have shown increasing concentrations of mercury, which supports this hypothesis. Recent changes in climate have been observed in western Hudson Bay and may also play a roll in the increases in mercury observed in beluga from this region. Melting of permafrost and movement of organically bound mercury and its accumulation in the form of methylmercury might explain the observed trends; however, much additional work would have to be done to confirm this.

In contrast to the relatively sudden rise in mercury in Beaufort Sea beluga over the past ten years there has been a slow but steady increase in mercury in seabird eggs over the past 30 years in Lancaster Sound. Mercury has also increased by about three-fold in the liver of ringed seals from Pond Inlet from 1976–2000. On the other hand, no significant increases in mercury in the liver of ringed seals from Holman in the western Canadian Arctic were seen over a 30-year period (1972–2001) although a two-fold increase was observed between 1993–2001. In Great Slave Lake, mercury increased significantly in lake trout over a 22-year period (1979–2001) but declined in walleye, northern pike and burbot over approximately the same period. Mercury temporal trends are thus very species and regionally specific and additional study is needed to understand this variation.

Most "legacy" OCs have significantly declined in Canadian Arctic biota from the 1970s to the 1990s. Significant declines of PCBs and DDT have been observed in all ringed seal populations and in seabird eggs in Lancaster Sound. Increases in the proportion of DDE to DDT in seals, beluga and seabird eggs suggest that the DDT is from weathered or old sources. There have been limited changes in coplanar PCBs and chlorinated dioxin/furan (PCDD/Fs) concentrations in Holman Island ringed seals over 20 years. Levels of PCDD/Fs and coplanar PCBs are, in general, decreasing in seabirds, although PCDFs increased in northern fulmar from 1975–1993. The one exception to this is ΣHCH levels, which have remained relatively constant in most species. Proportions and levels of the toxic β-HCH isomer have actually increased in seabird eggs and in ringed seal blubber. The decline of PCBs and DDT-related compounds was most rapid in the 1970s and 1980s following the banning of OCs in the USA, Canada and Western European countries. However the rate of decrease has slowed or stopped in many species, e.g., polar bears of western Hudson Bay. The rate of decline of OCs also varies between species with the slowest decline in beluga and polar bears and the most rapid in ringed seals and seabirds.

The scope of contaminant data for freshwater and anadromous (sea-run) fish has grown considerably under NCP Phase II. New data for NWT, Nunavik, Northern Labrador and Nunavut address a gap identified in the first CACAR report. Geographic coverage is limited, however, in Baffin Island and the Kitikmeot region of western Nunavut. Data for mercury and OCs are available for lake trout, lake whitefish, walleye, northern pike and Arctic char from a wide geographical area. Mercury data for a number of fish species now include most regions of the Canadian Arctic. For many lakes, concentrations of mercury in freshwater fish exceed guideline levels for subsistence (0.2 μg/g) and/or commercial sale (0.5 μg/g) and there are no geographic trends for these data. A number of studies have found that food web length, age and size of the fish are important variables while lake size or area does not appear to be as important. Yet a definitive answer for these high and variable levels remains elusive and this question remains one of the more important contaminant knowledge gaps for the NCP.

Lake-to-lake variability of OCs in freshwater fish is less than what is seen for mercury but lake trout and burbot in a number of lakes have higher levels. The reason is food web length for some lakes but this does not explain the high levels seen in other lakes. In the Yukon, glacial melt may influence OCs seen in some lakes and this effect could become more important if increasing temperatures continue. A study in Resolute Lake found that higher water temperatures resulted in higher levels of cadmium in Arctic char liver. The influence of climate change remains a relatively unstudied aspect of contaminant dynamics in freshwater biota and should receive attention in the future.

Mercury data have been produced for caribou, ringed seals and beluga from across the Canadian Arctic. Concentrations of mercury in ringed seal livers show no significant geographic trends although levels were higher at Arviat than in other locations. In beluga, concentrations of mercury were 2–10 times higher in Mackenzie Bay/Beaufort Sea animals than in other locations. Regional differences were also found between beluga in Mackenzie Bay and beluga at Paulatuk despite all being in the same southern Beaufort Sea stock. This regional difference within the stock may be related to a subgroup of animals returning to their same feeding grounds each year. It illustrates the large variation that occurs even within a geographically distinct group of beluga. Levels of cadmium in liver and kidney of ringed seals and beluga continue to show large differences between western and eastern Canadian Arctic populations, which is best explained by differences in geology.

Spatial coverage of OCs in ringed seals, beluga and seabirds remains a strength of the Arctic contaminant data set for Canada. Concentrations of PCBs, DDT and chlordane-related compounds, as well as toxaphene in marine biota, are slightly elevated in eastern compared to the western Canadian Arctic, consistent with circumpolar trends. HCHs tend to be higher in the western Arctic because of the influence of their recent use in Asia. New information on the spatial trends of OCs in Arctic fox and marine invertebrates has recently been produced and shows similar patterns. With the exception of HCH, levels of OCs in ringed seals, polar bears,

seabirds, and freshwater fish are lower in the Canadian Arctic compared with the same or similar species from eastern Greenland and the European and Russian Arctic.

A number of comprehensive marine food web studies were carried out recently. These studies have shown that OCs and mercury biomagnify in marine food webs. These studies also highlighted the importance of biomagnification on levels of OCs and mercury in higher trophic level organisms. However, the role of the abiotic-biotic interface at the base of food webs needs to be better understood in both the marine and freshwater environment. In particular, there have been no studies on the uptake of mercury, or other metals, from melt water. Nor have the pathways of oxidized mercury deposited in snow during springtime "polar sunrise" events to freshwater and marine food webs been examined.

The contribution of local contamination by OCs and metals has not been addressed very well in this assessment because of the emphasis of NCP on background sites influenced by long-range transport. Near field — far field comparisons have only been conducted in a few cases, e.g., Saglek Bay (Labrador), Cambridge Bay (Nunavut) and they generally have shown more elevated contaminant levels sources. Near field — far field comparisons conducted in Saglek Bay (Labrador) have shown elevated contaminant levels and related biological effects in seabirds near the military installation where PCBs were used. Monitoring of OCs in marine mussels from Labrador, Nunavik and Frobisher Bay showed elevated PCBs in some harbour sites (Iqaluit and Kuujjuaq). The limited data suggest that sources of contamination of biota from local sources, especially former military sites and harbours, may well be more significant than suggested in the previous assessment and warrant further attention and study.

The first CACAR report concluded that there was limited information and data on biological effects of contaminants in Canadian Arctic biota and it was an important knowledge gap. Since that report there have been few studies on this subject and it remains an important gap. Although the few studies to date have failed to find solid biological effects related to anthropogenic contaminants, and current levels in Canadian Arctic biota are generally below threshold levels of effects, there is still cause for concern and a need for additional research. Biological effects have been observed in polar bears in western Hudson Bay and black guillemots from a locally contaminated site (Saglek Bay). With probable climate warming, burdens of some contaminants may increase and stress associated with current and future exposures may become important. Although the possible effects associated with change in the Arctic clearly goes beyond contaminant effects it is important that they be considered and studied in the future.

A comprehensive study that examined the possible links between trace metal concentrations and health-related biomarkers in sea ducks (common and king eiders) found evidence for a relationship between sea duck health and metal levels. The biomarkers tested included body condition, parasitic infestations and immune function.

TCDD toxic equivalent (TEQ) concentrations in livers of northern fulmars, black-legged kittiwake and thick-billed murre from Lancaster Sound exceeded no-observed-effects-levels and lowest-observed-effects-levels (NOELs and LOELs, respectively) established for fish-eating birds such as herring gulls, bald eagles and cormorants.

Studies of Canadian Arctic and Svalbard (Norway) polar bears have concluded that the immune systems of polar bears are compromised by PCBs and potentially by other OCs and that blood levels of the thyroid hormone, T_4, correlated to levels of PCBs. Svalbard bears had lower thyroid hormone levels than polar bears from the Barrow Strait (Resolute) area. A recently completed study compared immune responses in polar bears from Svalbard and Churchill on western Hudson Bay. Bears with higher levels of PCBs were shown to have altered immune responses based on the production of antibodies when challenged with a virus. The influence of these altered systems on the health of polar bear populations is unknown. Full details of the study were not available in time for this assessment.

Finally, the data sets generated from monitoring of these OC chemicals and heavy metals in NCP Phases I and II, as well as in the pre-NCP program from 1985-1989, are now very large since the same set of contaminants has been monitored in a number of key species for up to 17 years in some cases. There were difficulties accessing all of the contaminants data for this assessment because of varying policies on data access by NCP Partner departments. This constrains the ability to independently assess temporal and spatial trends of contaminants. NCP should consider setting up a database of its own similar to the AMAP data centres to house data that have been paid for under the program.

Résumé

Le Programme de lutte contre les contaminants dans le Nord (PLCN) a été créé en 1991 en réponse aux préoccupations concernant l'exposition des humains à des concentrations élevées de contaminants dans les poissons et les espèces sauvages qui sont importants dans l'alimentation traditionnelle des Autochtones du Nord. Les premières études avaient mis en évidence une vaste gamme de substances — les polluants organiques persistants, les métaux lourds et les radionucléides — dont beaucoup n'avait pas de sources dans l'Arctique ni au Canada, mais qui se retrouvaient à des concentrations anormalement élevées dans l'écosystème de l'Arctique.

Dans la première phase du PLCN (PLCN-I), la recherche était concentrée sur la collecte des données nécessaires pour déterminer les concentrations, l'étendue géographique et les sources des contaminants dans l'atmosphère, l'environnement et les habitants du Nord, et la durée probable du problème. Les résultats de cette phase ont été colligés et publiés en 1997 dans le *Rapport d'évaluation des contaminants dans l'Arctique canadien (RECAC)*.

En 1998, le PLCN a entrepris sa deuxième phase (PLCN-II), qui se poursuivra jusqu'en mars 2003. Cette phase se concentre sur les impacts et les risques pour la santé humaine que pourraient induire les concentrations actuelles de contaminants dans les espèces alimentaires clés de l'Arctique, et sur la détermination des tendances temporelles des contaminants dans les espèces arctiques qui sont des indicateurs clés, ainsi que dans l'air. Elle examine ces questions dans un certain nombre de sous-programmes : santé humaine, surveillance de la santé des populations et écosystèmes de l'Arctique et de l'efficacité des contrôles internationaux, éducation et communications, et politique internationale.

La phase II du PLCN a identifié trois domaines d'étude prioritaires pour surveiller la composante biologique : les études sur les tendances temporelles; la surveillance et les enquêtes sur les aliments traditionnels; et la santé des animaux sauvages. Le rapport technique sur l'environnement biologique de la série RECAC-II a pour but d'examiner l'information visant ces domaines prioritaires et d'évaluer l'état actuel des connaissances sur les tendances spatiales et temporelles des contaminants, et leurs effets biologiques sur le biote de l'Arctique canadien. La série CACAR-II est constituée d'un rapport de synthèse et de quatre rapports techniques sur les sujets suivants : santé humaine, environnement biologique, environnement physique et application des connaissances.

Le premier Rapport de l'évaluation des contaminants dans l'Arctique canadien (RECAC) sur les contaminants dans le biote (chapitre 3, Absorption des contaminants par les écosystèmes et leurs effets) compilait et évaluait les données sur les contaminants obtenues avant 1997. La plupart de ces données ont été recueillies dans la phase I du Programme de lutte contre les contaminants dans le Nord (PLCN) du ministère des Affaires indiennes et du Nord canadien (MAINC). Ce rapport présentait une série de conclusions et identifiait les lacunes dans les connaissances et les données sur les tendances spatiales et temporelles et les effets biologiques des contaminants anthropiques dans le biote de l'Arctique canadien. Ce sont ces lacunes dans les connaissances qui ont orienté les recherches dans la phase II du PLCN (1997–2003). Les points saillants de la recherche effectuée sur le biote arctique dans la phase II du PLCN sont notamment des ensembles de données améliorés sur les tendances temporelles des contaminants organiques et métalliques, l'élargissement des tendances spatiales de la dynamique des contaminants organiques et métalliques, et l'identification et la mesure d'un ensemble de nouveaux contaminants anthropiques. Toutefois, nombre des lacunes dans les connaissances qui ont été identifiées dans le premier RECAC ont reçu moins d'attention ou n'ont pas pu être comblées, et de nouvelles questions et préoccupations se sont manifestées dans les cinq dernières années. Entre autres, ce sont la cause de niveaux élevés et variables de mercure dans le poisson d'eau douce, les différences régionales et les différences entre espèces dans les tendances temporelles du mercure, l'absence de tendances spatiales et temporelles pour les nouveaux contaminants et les métaux autres que le mercure, le cadmium et le plomb, et l'impact des contaminants sur la santé du biote arctique.

Une centaine de composés organochlorés (CO) « classiques » [dont les BPC, le DDT, le chlordane, la dieldrine, les hexachlorocyclohexanes (HCH) et les chlorobenzènes (CBz)] ont été mesurés dans la plupart

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des études subventionnées par la phase II du PLCN. Bien que jusqu'à 25 métaux aient été mesurés dans certaines études, la plupart de celles-ci se sont concentrées sur le mercure et le cadmium et le sélénium, et on continue de manquer d'information sur les autres métaux. La qualité des données sur les contaminants recueillies dans la phase II du PLCN est en général très bonne, et le programme d'assurance de la qualité est vraiment un point fort du PLCN, qui autorise une plus grande confiance dans les résultats et les conclusions de cette évaluation. Tous les laboratoires qui ont fourni des données dans cette évaluation ont participé au projet d'assurance de la qualité interlaboratoire financé par le PLCN ces cinq dernières années. De nouvelles données sur le toxaphène ont été obtenues pour le poisson d'eau douce, les oiseaux de mer et les mammifères marins, ce qui contribue à combler le manque de données identifié dans le premier RECAC. Toutefois, l'étude sur l'assurance de la qualité a révélé une variabilité entre les laboratoires plus grande pour le toxaphène et ses congénères spécifiques que pour les autres CO.

Plusieurs nouvelles substances chimiques ont été détectées dans le biote arctique. Le nombre des nouvelles substances mesurées a augmenté de façon significative au cours de la phase II du PLCN. Ces données fournissent une couverture plus exhaustive que le premier RECAC pour les organismes arctiques, dont des ensembles de données sur les tendances temporelles, mais leur portée reste très limitée comparativement aux CO classiques. Des données sur les éthers diphényliques polybromés (EDP) ont été obtenues pour les organismes d'eau douce et les organismes marins alors qu'on a mesuré des paraffines chlorées courtes et des naphtalènes chlorés dans les bélugas et les phoques annelés. Des mesures plus détaillées ont également été effectuées sur des pesticides utilisés présentement, le lindane et l'endosulfane. Les enquêtes n'ont révélé que des concentrations très faibles ou non détectables de tributylétain et de muscs synthétiques dans les moules, l'omble et les phoques de l'est de l'Arctique. L'acide sulfonique, une substance que l'on ne s'attendait pas à trouver dans l'Arctique à cause de ses propriétés physiques ou chimiques, a également été détecté dans le biote arctique canadien. La découverte de cette substance dans le biote arctique à des concentrations importantes soulève des questions quant aux sources et aux voies de transport de ces substances et d'autres substances des régions polaires. Bien que les concentrations de plusieurs de ces nouvelles substances chimiques soient inférieures à celles des POP classiques (BPC, DDT, etc.), la situation est préoccupante parce que les EDP sont détectés à des concentrations de plus en plus grandes, alors que les AS semblent présents à des concentrations relativement élevées dans le foie de l'ours blanc de l'Alaska comparativement aux autres CO. Les projets actuels du PLCN mesurent les AS dans l'ours blanc et autres organismes marins, mais les résultats ne sont pas encore disponibles. Ces résultats montrent qu'il faut continuer d'être vigilant étant donné qu'il y a des milliers de substances chimiques commercialisées dans l'Arctique circumpolaire et que la persistance, la bioaccumulation et le potentiel de transport à grande distance n'ont été évalués que pour un petit nombre d'entre elles. Il faut évaluer les ECP, les naphtalènes chlorés et les AS, ainsi que d'autres nouveaux contaminants, dans une gamme d'espèces plus large et sur une plus grande étendue géographique, en ce qui concerne leur comportement dans le réseau trophique et leurs effets biologiques possibles.

L'une des réalisations les plus importantes de la phase II du PLCN est l'élaboration d'ensembles de données sur les tendances temporelles. Ceux-ci couvrent maintenant une gamme d'espèces et de produits chimiques (CO et métaux), dont certaines nouvelles substances comme les EDP. Les archives de tissus, qui ont été financées par la phase I du PLCN, se sont révélées cruciales pour ces études; elles ont été précieuses et ont permis de créer des ensembles de données sur les tendances temporelles pour des substances qui n'ont que récemment été détectées dans l'Arctique. Nombre de ces études ont fourni des ensembles de données qui vont des années 1970 à 2000, et ont comblé une lacune de données majeure pour l'Arctique canadien et circumpolaire. Ces ensembles de données mondiaux offrent une excellente comparaison pour des études exhaustives à long terme des contaminants dans le poisson des Grands Lacs et du nord de la Suède, et fournissent des renseignements précieux sur la réponse du réseau trophique dans l'Arctique de l'interdiction de substances chimiques. La poursuite de l'appui aux archives de tissus et aux programmes de surveillance à long terme devrait être une priorité.

On a des indications solides que les concentrations de mercure ont augmenté dans les animaux de l'Arctique canadien entre l'ère préindustrielle et l'époque actuelle. Par exemple, le mercure a augmenté de 10 à 17 fois dans le béluga de la baie du Mackenzie entre les environs de 1450 et 2000. Cependant, les ensembles de données sur les tendances temporelles du mercure dans ces animaux montrent des tendances divergentes durant les 20 ou 30 dernières années. Le mercure a augmenté d'un facteur 4 au cours des 10 dernières années dans le béluga de la mer de Beaufort, et d'un facteur 2,5 dans l'ouest de la baie d'Hudson, mais pas de façon significative à aucun autre endroit. Aucune augmentation du cadmium dans le béluga n'a été observée au cours de la même période. Il a été suggéré que la hausse de températures dans la région du Mackenzie a libéré le mercure, ce qui pourrait expliquer l'augmentation des concentrations de ce métal observée dans le béluga de la mer de Beaufort. La lotte du Mackenzie est la seule espèce de poisson d'eau douce où l'on a constaté une augmentation de la concentration de mercure, ce qui supporte cette hypothèse. Des changements climatiques ont récemment été constatés dans l'ouest de la baie d'Hudson; ils pourraient également jouer un rôle dans l'augmentation de la concentration de mercure observée dans le béluga de cette région. Le dégel du pergélisol, et le déplacement du mercure lié aux matières organiques et son accumulation sous forme de méthylmercure pourraient expliquer les tendances observées, mais il faudrait encore beaucoup de recherches pour le confirmer.

Contrairement à l'augmentation relativement soudaine du mercure chez le béluga de la mer de Beaufort au cours des 10 dernières années, on n'a constaté qu'une augmentation lente, mais constante, de ce métal dans les œufs d'oiseaux de mer depuis 30 ans dans le détroit de Lancaster. Le mercure a également augmenté environ d'un facteur trois dans le foie du phoque annelé de Pond Inlet entre 1976 et 2000. Par contre, aucune augmentation importante du mercure dans le foie du phoque annelé de Holman dans l'ouest de l'Arctique canadien n'a été constatée sur une période de 30 ans (1972–2001), bien que le mercure ait doublé entre 1993 et 2001. Dans le Grand lac des Esclaves, le mercure a augmenté de façon importante dans le touladi sur une période de 22 ans (1979–2001), mais a diminué dans le doré jaune, le grand brochet et la lotte durant à peu près la même période. Les tendances temporelles du mercure varient donc considérablement avec les espèces et les régions, et des études supplémentaires sont nécessaires pour comprendre cette variation.

La plupart des CO « classiques » ont diminué de façon importante dans le biote de l'Arctique canadien des années 1970 aux années 1990. Des baisses importantes des BPC et du DDT ont été observées dans toutes les populations de phoques annelés et dans les œufs des oiseaux de mer du détroit de Lancaster. L'augmentation du rapport du DDE au DDT dans les phoques, les bélugas et les œufs d'oiseaux de mer porte à croire que le DDT provient de sources altérées ou anciennes. Il y a

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eu des changements limités des concentrations de BPC coplanaires et de dioxine chloré/furanne (PCDD/F) dans le phoque annelé de l'île de Holman sur une période de 20 ans. En général, les concentrations de PCDD/F et de BPC coplanaires sont à la baisse dans les oiseaux de mer, bien que les concentrations de PCDF aient augmenté dans le fulmar nordique de 1975 à 1993. Une exception : la concentration de ΣHCH est restée pratiquement la même dans la plupart des espèces. La proportion et la concentration de l'isomère toxique βHCH ont en fait augmenté dans les œufs des oiseaux de mer et le lard du phoque annelé. La baisse des composés liés aux BPC et au DDT a été le plus prononcée dans les années 1970 et 1980 après l'interdiction des CO aux États-Unis, au Canada et dans les pays d'Europe de l'Ouest. Toutefois, la décroissance s'est ralentie ou s'est arrêtée dans de nombreuses espèces, p. ex. l'ours blanc de l'ouest de la baie d'Hudson. Le taux de décroissance des CO varie également d'une espèce à l'autre, la décroissance la plus faible se trouvant dans le béluga et l'ours blanc, et la plus rapide, dans le phoque annelé et les oiseaux de mer.

La quantité de données sur les contaminants pour les poissons d'eau douce et les poissons anadromes a considérablement augmenté dans la phase II du PLCN. De nouvelles données pour les T.N.-O., le Nunavik, le nord du Labrador et le Nunavut comblent une lacune identifiée dans le premier RECAC. Toutefois, la couverture géographique est limitée sur la terre de Baffin et la région de Kitikmeot de l'ouest du Nunavut. Des données sur le mercure et les CO sont disponibles pour le touladi, le grand corégone, le doré jaune, le grand brochet et l'omble chevalier dans une grande région géographique. Les données sur le mercure pour un certain nombre d'espèces de poissons couvrent maintenant la plupart des régions de l'Arctique canadien. Dans de nombreux lacs, les concentrations de mercure dans les poissons d'eau douce dépassent les concentrations recommandées pour la subsistance (0,2 μg/g) et/ou la vente commerciale (0,5 μg/g), et on ne dispose pas de tendances géographiques pour ces données. D'après un certain nombre d'études, la taille du réseau trophique et l'âge et la taille du poisson sont des variables importantes, tandis que la taille des lacs ou la région semble être des facteurs plus secondaires. Malgré cela, l'explication finale de la hauteur et de la variabilité de ces concentrations continue de nous échapper, et cette question reste l'une des plus importantes lacunes des connaissances sur les contaminants pour le PLCN.

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La variabilité des CO dans les poissons d'eau douce d'un lac à l'autre est moindre que celle du mercure, mais on en trouve de fortes concentrations dans le touladi et la lotte dans un certain nombre de lacs. La raison en est l'étendue du réseau trophique pour certains lacs, mais cet élément n'explique pas les fortes concentrations observées dans d'autres lacs. Au Yukon, la fusion glaciaire peut influer sur les CO détectés dans certains lacs, et cet effet pourrait devenir important si la hausse de la température se maintient. D'après une étude menée dans le lac Resolute, la concentration de cadmium dans le foie de l'omble chevalier augmente avec la température de l'eau. L'influence du changement climatique reste un aspect relativement non étudié de la dynamique des contaminants dans le biote dulcicole et devrait être examinée dans l'avenir.

Des données sur le mercure ont été recueillies pour le caribou, le phoque annelé et le béluga dans tout l'Arctique canadien. Les concentrations de mercure dans le foie du phoque annelé ne révèlent aucune tendance géographique significative, bien que les concentrations soient plus élevées à Arviat qu'ailleurs. Pour le béluga, les concentrations de mercure sont de 2 à 10 fois plus élevées dans les animaux de la baie du Mackenzie ou de la mer de Beaufort qu'aux autres endroits. On constate également des différences régionales entre le béluga de la baie du Mackenzie et celui de Paulatuk, bien qu'ils fassent partie de la même population du sud de la mer de Beaufort. Cette différence régionale dans la population pourrait s'expliquer par le retour de sous-groupes d'animaux aux mêmes aires d'alimentation année après année. Elle illustre la grande variation même dans des groupes géographiquement distincts de bélugas. Les concentrations de cadmium dans le foie et les reins de phoques annelés et de bélugas continuent de manifester de grandes différences entre les populations de l'ouest et de l'est de l'Arctique canadien, ce qui s'explique surtout par des différences géologiques.

La couverture spatiale des CO dans le phoque annelé, le béluga et les oiseaux de mer reste un point fort de

l'ensemble de données sur les contaminants arctiques pour le Canada. Les concentrations de BPC, de DDT et de composés liés au chlordane, ainsi que du toxaphène dans le biote marin, sont légèrement plus élevées dans l'est de l'Arctique canadien que dans l'ouest, ce qui est conforme aux tendances circumpolaires. Les concentrations de HCH sont généralement plus élevées dans l'ouest de l'Arctique parce qu'on a commencé à les utiliser en Asie. De nouvelles données sur les tendances spatiales des CO dans le renard arctique et les invertébrés marins ont été obtenues récemment et montrent des répartitions similaires. À l'exception des HCH, les concentrations de CO dans le phoque annelé, l'ours blanc, les oiseaux de mer, et les poissons d'eau douce de l'Arctique canadien sont plus faibles que chez les mêmes espèces ou des espèces semblables de l'est du Groenland et des régions arctiques d'Europe et de Russie.

Un certain nombre d'études exhaustives sur le réseau trophique marin ont été menées récemment. Elles montrent que les CO et le mercure se bioamplifient dans les réseaux trophiques marins. Elles mettent également en évidence l'importance de la bioamplification sur les concentrations de CO et de mercure dans les organismes de niveau trophique élevé. Toutefois, le rôle de l'interface abiotique-biotique à la base des réseaux trophiques doit être mieux compris pour les milieux marins et les milieux dulcicoles. En particulier, il n'y a pas eu d'études sur la fixation du mercure ou d'autres métaux provenant des eaux de fonte. Les voies de transfert du mercure oxydé déposé dans la neige aux réseaux trophiques d'eau douce et marin n'ont pas non plus été examinées.

La contribution de la contamination locale par les CO et les métaux n'a pas été examinée très bien dans cette évaluation à cause de l'importance accordée par le PLCN aux sites de fond influencés par le transport à grande distance. La comparaison des champs proche et lointain n'a été effectuée que dans quelques cas, par exemple à la baie de Saglek (Labrador) et à Cambridge Bay (T.N.-O.), et elle a généralement révélé des sources plus contaminées. Les comparaisons effectuées à la baie de Saglek (Labrador) ont révélé des niveaux élevés de contaminants et des effets biologiques connexes dans les oiseaux de mer au voisinage de l'installation militaire où des BPC étaient utilisés. La surveillance des CO dans les moules marines du Labrador, du Nunavik et de la baie Frobisher révèle des concentrations de BPC élevées dans certains ports (Iqaluit et Kuujjuaq). Les rares données disponibles portent à croire que la contamination du biote par des sources locales, particulièrement d'anciens sites et ports militaires, pourrait être beaucoup plus importante que ne le suggérait la précédente évaluation et qu'elle devrait être surveillée et étudiée de façon plus approfondie.

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Le premier RECAC concluait qu'on n'avait que des informations et des données limitées sur les effets biologiques des contaminants dans le biote de l'Arctique canadien et que cela constituait une lacune importante dans les connaissances. Depuis la publication de ce rapport, il y a eu peu d'études sur ce sujet et la lacune reste importante. Bien que les quelques études effectuées jusqu'ici n'aient pas mis en évidence d'effets biologiques certains liés aux contaminants anthropiques et que les concentrations actuelles dans le biote de l'Arctique canadien soient généralement inférieurs aux seuils des effets, la situation reste préoccupante et des recherches plus approfondies sont nécessaires. Des effets biologiques ont été observés dans l'ours blanc de l'ouest de la baie d'Hudson et le guillemot à miroir d'un site contaminé local (baie de Saglek). Étant donné que le climat va probablement se réchauffer, la charge de certains contaminants pourrait augmenter et les perturbations liées aux expositions présente et future pourraient devenir importantes. Bien que les effets possibles liés au changement dans l'Arctique ne soient pas limités aux effets des contaminants, il est important que ceux-ci soient examinés et étudiés dans l'avenir.

Une étude exhaustive des liens possibles entre les concentrations de métaux-traces et les biomarqueurs liés à la santé dans le canard de mer (canard commun et eider à tête grise) a mis en évidence des indications de liens entre la santé du canard de mer et les concentrations de métaux. Les biomarqueurs testés étaient l'état corporel, les infestations parasitaires et la fonction immunitaire.

Les concentrations d'équivalents toxiques du TCDD dans le foie du fulmar boréal, de la mouette tridactyle et du guillemot de Brünnich du détroit de Lancaster dépassaient la concentration sans effets observés et la concentration minimale avec effets observés (CSEO et CMEO, respectivement) établies pour des oiseaux ichtyophages

comme le goéland argenté, le pygargue à tête blanche et le cormoran.

Des études sur les ours blancs de l'Arctique canadien et du Svalbard (Norvège) ont conclu que le système immunitaire de l'ours blanc est affaibli par les BPC et peut-être d'autres CO, et que la concentration sanguine de l'hormone thyroïdienne T_4 est corrélée à la concentration de BPC. Les ours du Svalbard avaient des concentrations d'hormones thyroïdiennes plus faibles que les ours blancs du détroit de Barrow (région de Resolute). Une étude terminée récemment comparait les réponses immunitaires des ours blancs du Svalbard et de Churchill (région de l'ouest de la Baie Hudson). Les ours ayant de plus fortes concentrations de BPC montraient une réponse immunitaire modifiée, d'après leur production d'anticorps quand ils étaient exposés à un virus. L'influence de cette modification sur la santé des populations d'ours blancs est connue. L'exposé intégral de l'étude n'était pas disponible à temps pour la présente évaluation.

Finalement, les ensembles de données résultant de la surveillance de ces CO et métaux lourds dans les phases I et II du PLCN, ainsi que le programme préliminaire au PLCN de 1985 à 1989 sont maintenant très considérables car le même ensemble de contaminants a été surveillé chez un certain nombre d'espèces clés, jusqu'à 17 ans dans certains cas. Nous avons eu de la difficulté à obtenir toutes les données sur les contaminants pour cette évaluation en raison des différences dans les politiques d'accès aux données des ministères participants au PLCN. Cela limite la capacité d'évaluer indépendamment les tendances temporelle et spatiale des contaminants. Le PLCN devrait examiner la possibilité de créer sa propre base de données, semblable à celle du PSEA, pour y recueillir les données payées par le programme.

Pat Roach

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PART I

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Introduction

1.1 Background

The Northern Contaminants Program (NCP) was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in fish and wildlife species that are important to the traditional diets of northern Aboriginal peoples. Early studies indicated that there was a wide spectrum of substances — persistent organic pollutants, heavy metals, and radionuclides — many of which had no Arctic or Canadian sources, but which were, nevertheless, reaching unexpectedly high levels in the Arctic ecosystem.

Under the first phase of the NCP (NCP-I), which took place from 1991–1997, research was focussed on gathering the data required to determine the levels, geographic extent, and sources of contaminants in the northern atmosphere, environment and its people, and the probable duration of the problem. The data enabled us to understand the spatial patterns and temporal trends of contaminants in the North, and confirmed our suspicions that the major sources of contaminants were other countries. Information on the benefits from continued consumption of traditional/country foods was also used to carry out assessments of human health risks resulting from contaminants in those foods. Results generated through NCP-I were synthesized and published in 1997 in the Canadian Arctic Contaminants Assessment Report (CACAR) (CACAR, 1997).

In 1998, the NCP began its second phase (NCP-II), which will continue until March 2003. NCP-II focusses upon questions about the impacts and risks to human health that may result from current levels of contamination in key Arctic food species as well as determining the temporal trends of contaminants of concern in key Arctic indicator species and air. A large amount of data on contaminants in the Canadian Arctic has been produced since the first CACAR report was published. The "legacy" POPs, as well as mercury, cadmium and selenium have continued to be the focal point of the measurement and monitoring of contaminants in the Canadian Arctic. However, new chemicals, such as polybrominated diphenyl ethers (PBDEs) have warranted significant attention. CACAR-II was prepared in parallel with the international Arctic Monitoring and Assessment Programme (AMAP) II Assessment.

Phase II of the NCP identified three priority areas of study for monitoring the biological environment: Temporal Trend Studies; Monitoring and Surveys of Country/Traditional Foods; and Wildlife Health (NCP BluePrint 2000–01). The purpose of this chapter is to review the information that addresses these priority areas and to assess the current state of knowledge on the spatial and temporal trends of contaminants and biological effects in biota in the Canadian Arctic. Most of the data are drawn from the "Biotic Monitoring" subprogram of the Northern Contaminants Program (1997–2001), although published and unpublished results have been included from other contaminant studies conducted in the Canadian Arctic during the 1997–2001 period. Results from the early 1990s which appeared in the previous assessments of the biological environment (1985–1996) (Lockhart *et al.*, 1992; Thomas *et al.*, 1992; Muir *et al.*, 1992a; 1997; 1999a) are not included except where needed for comparison with newer data. In keeping with the NCP mandate, the results are mainly from studies conducted in Yukon, NWT, Nunavut, and

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in coastal Nunavik and Labrador. Contaminant measurements in freshwater biota in Nunavik and Labrador or in northern Ontario and western provinces are not included.

1.2 Study design

The "Biotic Monitoring" program was designed with a focus on "priority contaminants" identified under the Persistent Organic Pollutant and Heavy Metal Protocols of the United Nations Economic Commission for Europe Convention on Long-Range Transboundary Air Pollution that were under negotiation in 1997. The program was also designed to conduct preliminary studies on "other" contaminants, not currently identified in the "protocols" and having physical/chemical properties which give reason for concern in that they may pose a risk to northerners (e.g., persistent, toxic, bioaccumulative, and prone to long-range transport and deposition).

The "Biotic Monitoring" program was designed to address the large number of "knowledge gaps" for contaminants in the biological environment which were identified in the previous assessment. These knowledge gaps include:

1. Temporal trends in biota. The lack of temporal trend information for most contaminants was identified as the most significant knowledge gap. Temporal trend data were very limited for most persistent organochlorines (OCs) and metals because they were based on two or at most three sampling times and some comparisons were limited because of changes in methodology.

   Temporal trend data for mercury were judged to be particularly important given evidence of increasing concentrations in marine mammals. There were essentially no data on temporal trends in mercury concentrations for freshwater and anadromous fish in the NWT.

2. Priority chemicals. While the list of priority substances monitored in biota in the Canadian Arctic was substantial there were a number of chemical contaminant groups for which information was quite limited. These groups included PCDD/Fs, as well as non-*ortho* polychlorinated biphenyls (PCBs), toxaphene, other planar OCs such as chlorinated naphthalenes, chlorinated and brominated diphenyl ethers, current-use pesticides, as well as other species and isotope ratios of metals in biota.

3. Geographic coverage of contaminant measurements in biota. In freshwater fish spatial coverage was found to be very good in Yukon, but is much less detailed in the NWT and northern Québec. Contaminant analysis of freshwater fish in the Nunavut region of the NWT and the Nunavik area of northern Québec was limited mainly to Arctic char (*Salvelinus alpinus*) and was insufficient to assess spatial trends. Further sampling of other subsistence fish in Nunavut and Nunavik was recommended as a future priority.

   Geographic coverage of marine mammal and seabird populations was found to be very good. All major beluga, ringed seal and polar bear stocks along with several major seabird colonies were sampled for OC and heavy metal contaminants during the 1990–1995 period.

4. Marine fish and food webs. There was little information on contaminant levels and very limited geographic coverage for marine fish and invertebrates. This gap is significant because of its importance in understanding of the pathways of bioaccumulation of the contaminants. It was also noted that the marine food web data are important because some animals are harvested by northerners such as mussels and amphipods, or fished commercially (turbot, prawns).

5. Impact of local pollution sources. There was evidence at some locations in the Canadian Arctic of localized PCB contamination of terrestrial plants, soils and nearshore sediments and biota due to pollution from military radar facilities and other activities. When considered on a broad regional scale, there is a need to determine whether marine mammals and seabirds

frequenting the waters within the general area of these sites as well as terrestrial mammals, such as caribou and Arctic fox feeding within the affected areas, have elevated PCB and lead (Pb) contamination.

6. Lack of modelling of chemical fate and bioaccumulation. Lack of integration of chemical measurement data with well established models of chemical fate and food chain bioaccumulation was identified as a major gap in the 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.
7. Biological effects. The limited data on biological effects indicators, especially immunosuppression in mammals at high trophic levels, was identified as a major gap which should be addressed in future. A research program on polar bear and beluga immunology and immunosuppression was proposed as the top priority given the fact that these species exhibit the highest levels of PCBs and other OCs such as chlordane and toxaphene than most other species.

   The biological implications of very high levels of mercury (Hg) and cadmium (Cd) in seabirds, marine mammals and caribou were identified as a significant knowledge gap.

One of the goals of this assessment of contaminant levels and trends in the biological environment is to determine if these knowledge gaps have been addressed.

INAC

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Pathways and Processes of Delivery of Contaminants to Marine, Freshwater and Terrestrial Ecosystems

2.1 Inputs of airborne and seaborne contaminants

Pathways of transport of the OCs, heavy metals, radionuclides, and hydrocarbon contaminants to the Arctic include transport in the troposphere in gas phase and adhered to particles, as well as via ocean currents (Macdonald *et al.*, 2000). The processes that deliver contaminants to marine, freshwater and terrestrial environments where they can become available for uptake by biota are considered herein. Processes for the marine and terrestrial environments are illustrated in Figure 2.1.1.

Airborne contaminants are removed from the atmosphere by gas absorption, precipitation and dry deposition. Many

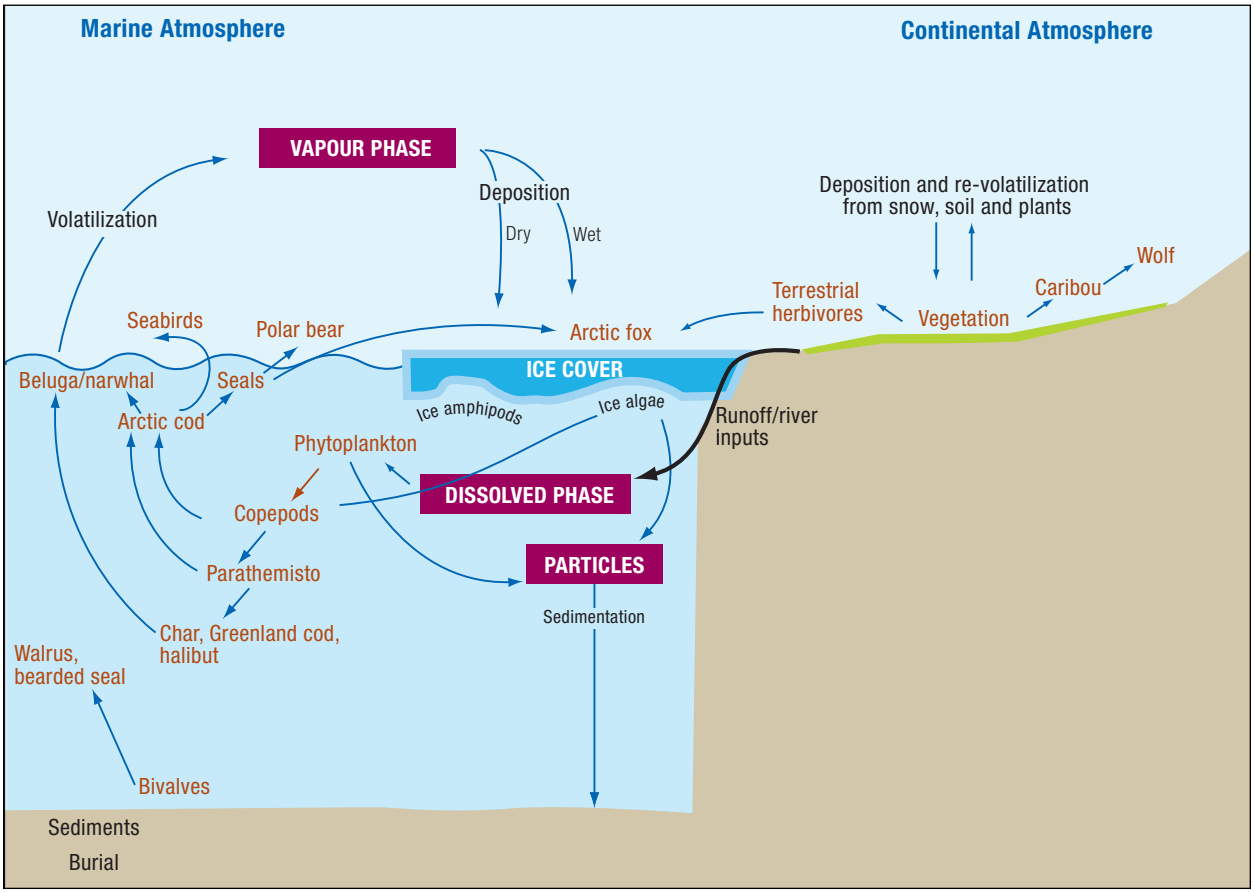

FIGURE 2.1.1
Bioaccumulation pathways of contaminants in Arctic food webs.

chlorinated organics are present as gases even at low temperatures and are absorbed from the gas phase by water, snow and plant surfaces. Precipitation scavenging of gas 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. For the more water soluble, less volatile contaminants, transport via ocean currents may be more important than the airborne route (Li *et al.*, 2002).

In the water column, more highly chlorinated OCs such as DDT and PCBs (Cl_4–Cl_{10}) are associated with particles while others are found mainly in the dissolved phase (hexachlorocyclohexanes or HCH, and toxaphene).

Waterborne 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.

2.2 Bioaccumulation processes

Hydrophobic organics and heavy metals such as mercury, 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 depend on the properties of the chemical and on the physical, chemical and biological environment into which it is released.

Persistent organochlorines accumulate in organisms due to a high affinity for lipids and, most importantly, high biological inertness of the parent chemical or metabolites. For metals, differences in uptake due to speciation of the element (which may be influenced by water hardness, salinity, 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 elimination of metal into the bile following uptake.

ITK/Eric Loring

Chemicals are accumulated by fish and invertebrates via three major route of exposure: gills, diet, and dermal. For organic chemicals, the relative importance of these exposure routes varies with the physical-chemical properties of the chemical. Dermal is generally not considered an important exposure route for organic chemicals in fish but may be important for some soft-bodied invertebrates such as oligochaetes. Dissolved chemicals, both organics and metals (in ionic form), enter the organism by diffusion across the gill membrane or other respiratory surfaces into the blood (Connell, 1988; Heath, 1987). Accumulation from water is termed *bioconcentration*. For organochlorines, such as PCBs, equilibrium is approached and sometimes established between the water and lipid phase of the organisms. However, dietary accumulation plays a large role in the concentrations observed in biota. A discussion on food web transfer follows. For metals, equilibrium is established between dissolved forms in water and dissolved or protein-bound forms in blood and other tissues. OCs and heavy metals also adsorb to particle surfaces and are then available for uptake in food by grazing animals (zooplankton).

Dietary accumulation is often the dominant route of exposure for organic contaminants and methylmercury. This varies with the hydrophobicity of the chemical. Dietary accumulation makes up a greater percentage of the exposure with increasing hydrophobicity. This is due to relatively low concentrations in water (typically in the picograms/L in the Arctic) and low rates of elimination of these chemicals by the organism. Therefore, concentrations in food tend to be higher than what is found in water, with the difference becoming greater with each step up the food web. Organisms that feed higher in the food web tend to get almost all of their exposure to hydrophobic chemicals via food. For example, it has been estimated that piscivorous lake trout get 99% of their PCB load from their diet (Thomann and Connolly, 1984). On the other hand, for chemicals such as HCH, uptake via the gills may be relatively important even for lake trout.

Many factors such as organic carbon content of soils and sediments, pH and kinetic limitations influence the amount of a contaminant that can be released from food particles in the gut or dissolved into sediment porewaters, and therefore reduce the environmental bioavailability. 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.

Transfer within the food web through food ingestion is the dominant pathway for uptake of persistent organic chemicals and heavy metals in larger marine and freshwater organisms, and in terrestrial food webs (Figure 2.1.1). These pathways, coupled with the slow rate of excretion and metabolism, lead to *biomagnification*, the increase in concentration of a chemical from prey to predator. Biomagnification factors are calculated by dividing the concentration found in the predator by the concentration found in the prey (both on a lipid basis). The food requirements of an organism are controlled by metabolic rate and production (growth, lipid deposition, reproduction). Thus metabolic rate is linked to the rate of uptake of contaminant. Homeotherms (mammals, birds) have greater energy requirements for the same body weight than poikilotherms (fish) because of the requirement to maintain body temperature; thus the metabolic rate and their caloric requirements are higher. As a result the largest biomagnification factors, the concentration of contaminants in predator divided by concentration in prey, are usually observed between fish and marine mammals or seabirds for higher chlorinated PCBs, OC pesticides and mercury, in Arctic food webs (Muir *et al.*, 1992a; Fisk *et al.*, 2001a).

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GNWT/RWED

Contaminants in Biota — Levels and Spatial Trends

3.1 Spatial trends of contaminants in freshwater biota

Freshwater fish are an important dietary item in many communities of the Canadian Arctic and in some cases are a commercial resource through sport fishing and the sale of fish for consumption. Consumption of freshwater fish is generally limited to Yukon, western NWT and northern Québec, but land-locked Arctic char (*Salvelinus alpinus*) are available in some Nunavut communities. Concerns about the health of freshwater fish have been raised in the past. For example, the liver of burbot (*Lota lota*) was considered at one point to be of poor quality in the Mackenzie River (Lockhart *et al.*, 1988). As well, many of the Arctic lakes are susceptible to high levels of contaminants in top trophic level fish due to the characteristics of the lake (i.e., cold with long food webs) and fish (i.e., high lipid content and slow growth rates) (Figure 3.1.1).

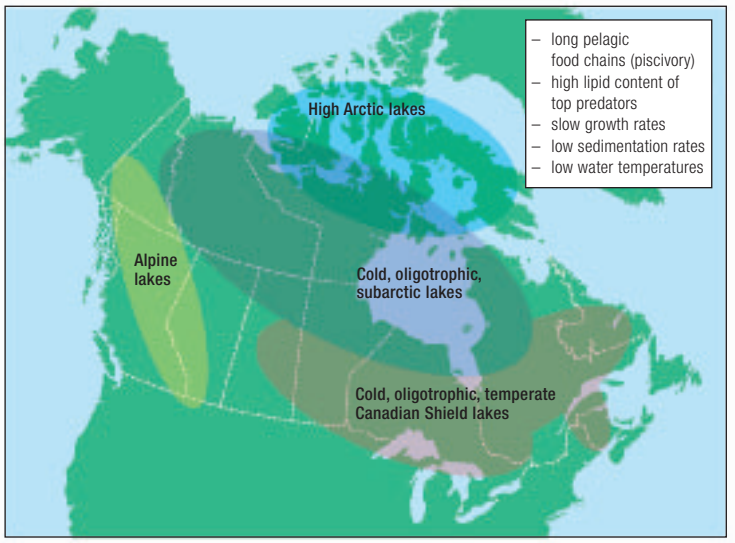

FIGURE 3.1.1

Lakes susceptible to high levels of organochlorines in top trophic level fish.

A fairly large data set of OC and mercury in freshwater fish was available for the first Canadian Arctic Contaminants Assessment Report (CACAR) (Muir *et al.*, 1997). From this report, it was concluded that mercury was the one contaminant that consistently exceeded guideline limits for subsistence consumption or commercial sale. Concentrations of toxaphene and PCBs were also found to be significant in some lakes. Mercury and OCs were shown to biomagnify through Arctic lake food webs (Kidd *et al.*, 1995; 1998) with the highest concentrations in the top predators, such as lake trout (*Salvelinus namaycush*) and northern pike (*Esox lucius*). Other variables such as fish length and age were also found to have a significant influence on contaminant concentrations.

The first CACAR concluded that coverage of spatial trends of contaminants in Yukon was considered adequate but that information on the NWT, Nunavut and northern Québec was insufficient (Muir *et al.*, 1997). Levels of mercury and OCs in fish were shown to vary spatially between Arctic lakes (Muir *et al.*, 1997). OCs in fish tended to be similar between lakes with some notably high levels in some lakes, such as Lake Laberge (Kidd *et al.*, 1995). Variability in spatial trends of mercury and OCs was attributed to a combination of differences in the food webs of each lake, biological characteristics of the fish, and potential sources of mercury (anthropogenic versus natural) but research has continued on this issue.

Since the first CACAR a large number of fish have been analyzed for contaminants. Most, however, have been for mercury only. These analyses have also provided information on variables that influence contaminant levels in freshwater fish and provide more comprehensive spatial coverage. Recently, efforts have been made to measure and assess the concentrations of new chemicals in freshwater fish, mainly brominated flame retardants.

3.1.1 Spatial trends of metals in freshwater fish

Most of the data produced on metals in freshwater organisms are on mercury in fish, due mainly to concerns about levels exceeding human consumption guidelines. Most mercury values have been determined as total mercury (inorganic + organic) but most mercury found in freshwater fish is predominantly in the form of methylmercury (MeHg). There are smaller amounts of data produced for selenium (Se), cadmium, arsenic (As)

and lead, although levels of these chemicals are rarely of concern for consumption guidelines. No new data have been produced for metals in invertebrates.

3.1.1.1 Factors influencing concentrations of metals in freshwater fish

Concern about high mercury concentrations in predator species of Arctic Canadian freshwater fish is geographically extensive. Fish from Yukon to northern Nunavut have been found to contain high levels of Hg, e.g., lake trout from Nunavut have an unusually high proportion of values exceeding the limit for marketing recommended by Health Canada (Lockhart *et al.*, 2001). Large spatial variations in Hg concentrations have also been found in predatory fish in the NWT (Jensen *et al.*, 1997), particularly in lakes within the Mackenzie River Basin where, in many instances, Hg levels exceed 0.5 μg/g. The reasons for the large regional variations between lakes in this area are not yet fully understood.

Size and age

Mercury levels are influenced by age and size of fish (Hermanson and Brozowski, 1993; Lockhart *et al.*, 2000a; Muir *et al.*, 2000a; Riget *et al.*, 2000), however, larger fish do not always contain higher levels of Hg than smaller fish (Amundsen *et al.*, 1997; Evans and Lockhart, 2001; Skotvold *et al.*, 1997). Evans and Lockhart (2001) concluded that fish had high Hg concentrations in some Arctic Canadian lakes because of slow growth rate. The same authors found that predatory fish start to approach Hg concentrations of 0.5 μg/g at about 10–12 years of age when they are eating more small forage fish. Examples of the variation in fish mercury levels versus length are given in Figure 3.1.2 (Evans and Lockhart, 2001). Stern *et al.* (2001a) studied mercury, arsenic and selenium in burbot from Fort Good Hope and did not find any significant correlation between length and mercury concentration in muscle and liver in either sex. For selenium, weak negative correlations exist between concentration and length for liver and muscle in male fish.

Trophic position

Comparison of mercury concentrations in lake trout, Arctic char, lake whitefish (*Coregonus clupeaformis*), walleye, northern pike, burbot and white sucker (*Catostomus commersoni*) from a data set consisting of > 2,000 samples based on analyses by the former Fish Inspection Service of DFO, previous and ongoing NCP studies (Lockhart *et al.*, 2000a; 2001; Muir and Lockhart, 1996; 1997; Evans and Lockhart, 2001) allows some inferences about the importance of the trophic position of the fish. Elevated Hg in this large fish data set is relatively specific to predator species like walleye, northern pike and lake trout. Species that feed lower in the food chain like lake whitefish have lower levels of Hg. It is known that the body burden of Hg in fish increases with the trophic level of their prey (Bruce and Spencer, 1979; Kidd *et al.*, 1995). Therefore, predatory fish like northern pike and walleye (*Stizostedion vitreum*) have generally higher mercury concentrations than species feeding on lower trophic levels, like whitefish and Arctic char, which mainly feed on zooplankton and benthos (Lockhart *et al.*, 2001).

Water chemistry and dissolved organic content

Wetland area and changes in these areas such as during instances of flooding or reservoir establishment have been found to be an important variable affecting Hg concentrations in fish in some regions (Frick, 1992; St Louis *et al.*, 1994). So far, however, studies have not revealed any strong relationships between Hg levels in fish and factors such as lake size, drainage basin area and wetland area (Evans and Lockhart, 2001). Preliminary findings from Mackenzie River Basin lakes indicate that the humic content of the water may also play a role in the uptake of mercury in fish, leading to higher mercury uptake in coloured streams than in clear ones (Evans and Lockhart, 2001a). The higher humic content favours the transport and retention of Hg in the water phase, as organic complexes, which may increase the availability of Hg for uptake in the food chain. Earlier studies of acidified lakes in Sweden and Norway indicated that the process of acidification increased mercury accumulation in fish, especially where the food web had been damaged, i.e., roach extinct by acidification (Andersen *et al.*, 1986; Björklund *et al.*, 1984). Methylation is also known to be favoured by increasing acidity and temperature (Evans and Lockhart, 2001; and references therein), i.e., trends that would lead to low concentrations of mercury in cold temperatures and alkaline bedrock areas such as occur in large parts of Arctic Canada.

Sediments

Mercury concentrations in lake sediments do not correlate strongly with Hg concentrations in fish. For example, the mercury concentrations in sediment in the lake at position 69°26′N 139°36′W in Ivvavik National Park, NW Yukon, were 20–40 times higher than in the other lakes in the Canadian Arctic visited during the Tundra North West Expedition in 1999, but the levels in fish muscle were still not elevated compared to the rest of the lakes (Borg, 2001). Painter *et al.* (1994) showed maps of sediment concentrations of mercury in SE Yukon and a recent collection of a few lake trout from Itsi Lake in that area revealed only low levels of mercury in the fish (Annex Table 1).

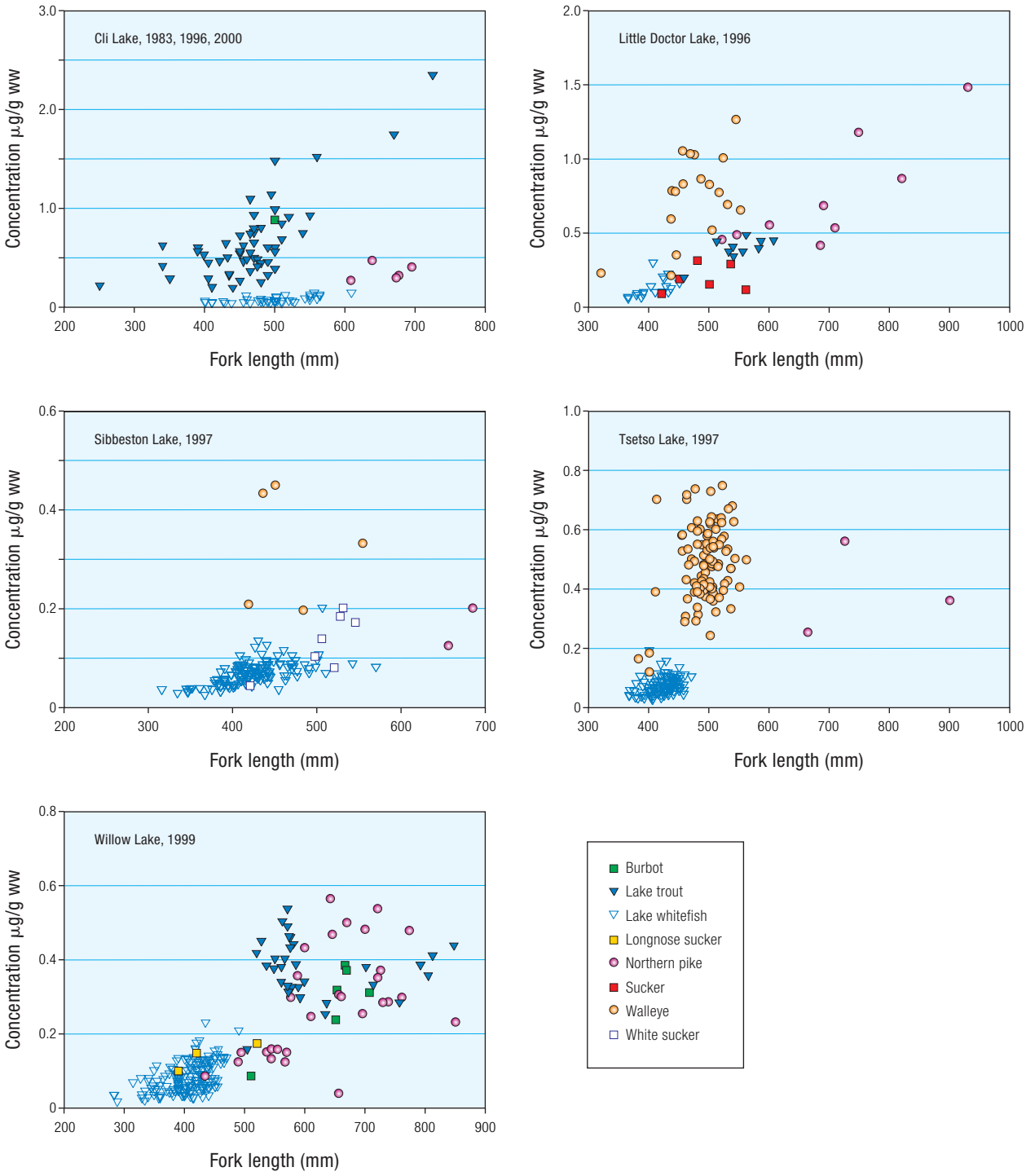

FIGURE 3.1.2

Mercury — length relationships in fish from lakes in the Mackenzie River basin (Lockhart *et al.*, 2001).

Migration and piscivory in char

Arctic char especially anadromous (sea-run) populations generally have low mercury levels [0.1 µg/g wet weight (ww)] Higher levels are found in non-migratory char (Lockhart *et al.*, 2001). About three-fold higher levels of Hg were found in land-locked char from a lake near Kangiqsujuaq than in sea-run char from the same area (Muir *et al.*, 2000a). Land-locked char have been collected over a number of years from lakes near the community of Qausuittuq (Resolute) in the Canadian High Arctic (Muir *et al.*, 2000b; 2001a). The effect of diet and trophic level on mercury and organochlorine levels was

investigated using stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). A few fish had $\delta^{15}N$ values that were up to 3.8 parts per thousand (‰) higher than others. This is associated with a change of trophic level, because concentrations of the heavier isotope of nitrogen, ^{15}N, are progressively enriched from prey to predator an average of 3 to 5‰ (Peterson and Fry, 1987). Land-locked char populations are known to develop a behavioural dichotomy wherein some individuals at a certain "escape size" adapt to a cannibalistic feeding habit, while other individuals may not. In Char Lake, a system containing only char, Hobson and Welch (1995) associated $\delta^{15}N$ values of 13.7 ‰ with piscivory in char. They also found a significant increase in the $\delta^{15}N$ of these fish with size that they attributed to cannibalism within the population. There were no significant correlations of $\delta^{15}N$ with length or weight in Resolute Lake char; however, mercury was significantly correlated with $\delta^{15}N$ in char from Resolute Lake collected in 1997 and 1999 (Figure 3.1.3). No other metals were correlated with $\delta^{15}N$, which suggests that biomagnification of mercury is occurring within the char population due to the presence of piscivorous char. The Hg concentrations in char from Resolute Lake and nearby lakes were higher than at the Canadian sites visited during the Tundra North West Expedition (Borg, 2001) but this may be partly explained by a larger average size of the analyzed individuals from Resolute Lake compared with the Expedition samples (about double). A correlation between Hg concentration and length and weight were found in the char but the scatter was large. The size influence on Hg was found to vary between the different lakes and in some there seemed to be no correlation (Borg, 2001).

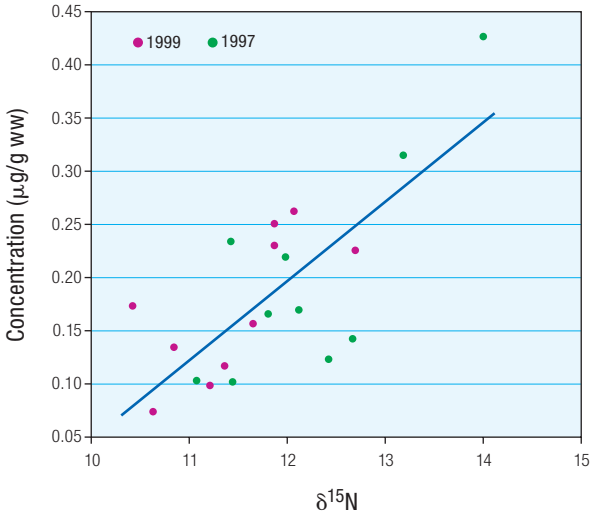

FIGURE 3.1.3

Relationship of mercury and $\delta^{15}N$ in land-locked char from Resolute Lake (1997 and 1999) (Muir *et al.*, 2001a). A significant relationship was found indicating that char with high mercury levels are feeding at a higher trophic level.

Comparison of metals in different fish tissues

Metal concentrations were investigated in selected fish [pike, burbot, walleye, and inconnu (*Stenodus leucichthys*)] from the Resolution Bay area (1996) and cisco (*Coregonus artedii*) (1995) from the East Arm of Great Slave Lake (Evans *et al.*, 1996) to examine differences in concentration between tissues. Overall, there was no evidence from this small study of highly elevated metal concentrations in the fish. This study showed, however, that fish tissues and the species of fish with the highest concentrations varied with the type of metal. In general, mercury concentrations were higher in muscle than in liver and below the 0.05 µg/g guideline set for the commercial sale of fish (Annex Table 1). Similarly, Stern *et al.* (2001a) found that mercury concentrations were higher in muscle than in liver of burbot from Fort Good Hope. The highest concentrations were observed in pike and walleye, while concentrations were substantially lower in burbot. Arsenic and cadmium concentrations tended to be higher in liver than in muscle, and cadmium was not detected in muscle. Cisco had the highest arsenic concentrations and cadmium concentrations were highest in walleye liver. Copper concentrations were higher in liver than muscle and particularly high in pike liver. Zinc concentrations also tended to be higher in liver than in stomach with remarkably high concentrations in pike stomach.

3.1.1.2 Spatial trends of metals in freshwater fish

Mercury

Data for mercury levels in freshwater fish of the Canadian Arctic are presented in Annex Table 1 and in maps for lake trout, northern pike and walleye (Figure 3.1.4). Most of these data have not been published and the information herein was derived from raw data tabulations; they represent the analyses of several thousand fish.

Levels of mercury have been measured in fish from northern Canada since the early 1970s, mainly in an effort to determine whether the fish should be marketed commercially. Studies for that purpose conducted by the Fisheries Inspection Service relied on small numbers of samples from an array of lakes and species. The size ranges of the fish were usually quite small — being those that would normally be taken in a commercial fishery. Species were usually limited to those of commercial interest. More recent studies have attempted to obtain larger numbers of samples and a broader range of sizes

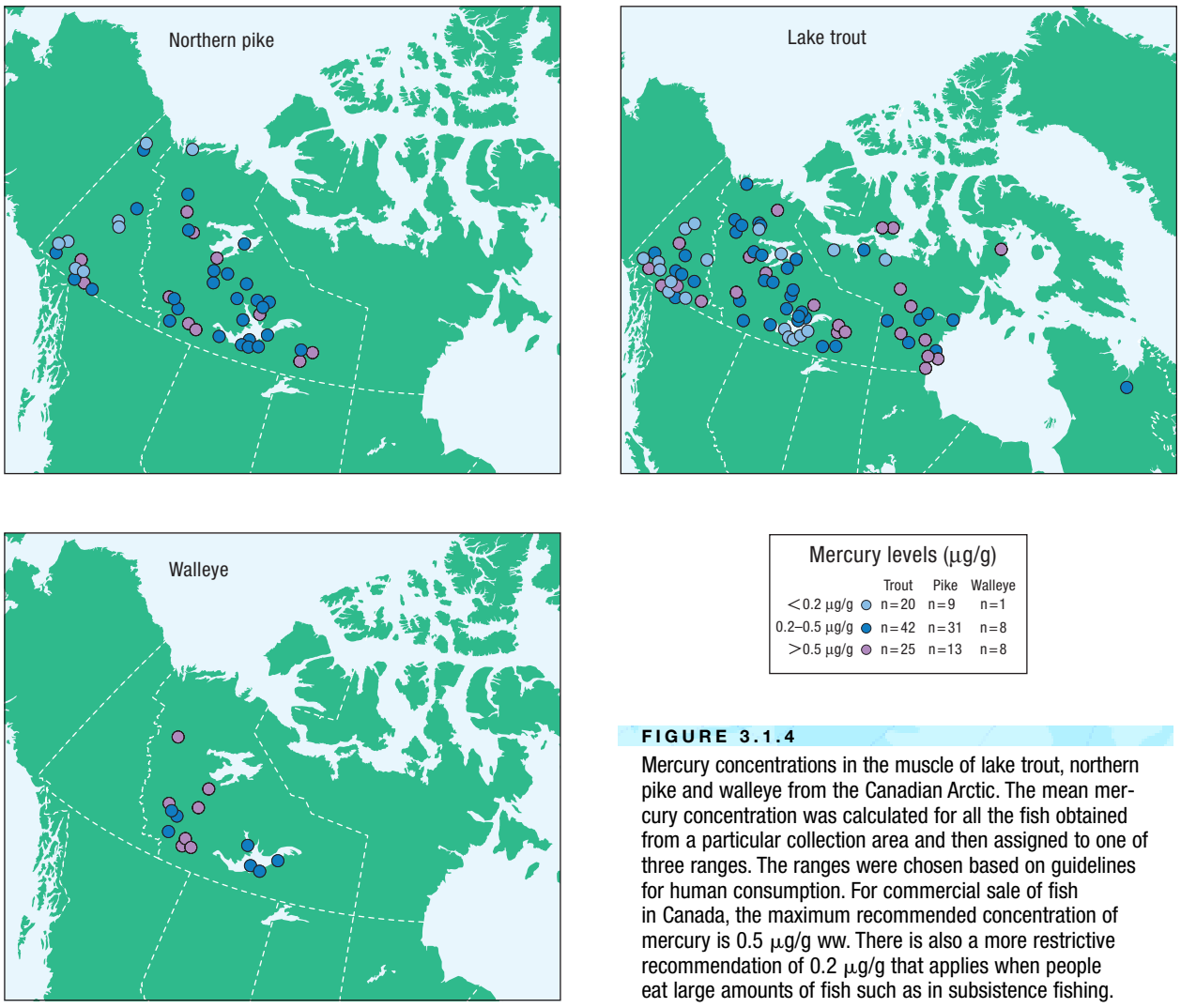

FIGURE 3.1.4

Mercury concentrations in the muscle of lake trout, northern pike and walleye from the Canadian Arctic. The mean mercury concentration was calculated for all the fish obtained from a particular collection area and then assigned to one of three ranges. The ranges were chosen based on guidelines for human consumption. For commercial sale of fish in Canada, the maximum recommended concentration of mercury is 0.5 µg/g ww. There is also a more restrictive recommendation of 0.2 µg/g that applies when people eat large amounts of fish such as in subsistence fishing.

from fewer locations, but the overall composition of the data reflects the larger fish of commercial and subsistence interest. In most instances, especially for the earlier collections, fish ages were not determined and so no attempt has been made to adjust for age. Normally fish lengths were recorded and have been tabulated as a descriptor to compare among lakes.

The maps in Figures 3.1.4 and 3.1.5 show points where fish were collected as one of three colours for a single species important in subsistence or commercial fishing. The mean mercury concentration was calculated for all fish obtained from a particular collection area and then assigned to one of three ranges. The ranges were chosen based on guidelines for human consumption. For commercial sale of fish in Canada, the maximum recommended concentration is 0.5 µg/g ww. There is also a more restrictive recommendation of 0.2 µg/g that applies when people eat large amounts of fish such as in subsistence fishing. There are additional data not shown for other species for which more limited data have been collected [suckers, inconnu, grayling (*Thymallus arcticus*), cisco].

No adjustment for the sampling time or fish size has been made in Figure 3.1.4 and Figure 3.1.5 so that the means calculated include any errors that would be introduced by systematic changes in levels over time and by differing mean sizes of the fish. The question of temporal change can be approached for lakes from which fish have been sampled on several occasions. Evidence suggests that temporal change would be a relatively small error component in the regional survey data (see Section 4.1.1). The years fish were sampled and mean fish lengths are shown for mercury and selenium levels in muscle in Annex Table 1. A more serious source of error is variation in fish age from lake to lake since mercury levels usually increase with increasing age. Examination of fish lengths in the tables, however, suggests relatively small differences in average fish length from one lake to

another, and so this variable too probably represents a relatively small component of error in the mean mercury levels and not one likely to produce systematic regional errors in interpretation.

There is no clear geographic pattern evident in mercury concentrations in lake trout, northern pike and walleye (Figure 3.1.4). High and low levels of mercury in the fish were found throughout the Arctic and this likely reflects the variability between lake systems. Factors that influence mercury in fish are discussed in Section 3.1.1.1. Efforts to explain geographic variability using larger scale geological information are inconclusive. For example, superimposing the levels of mercury in the fish over several types of regional geological maps failed to reveal obvious correlative patterns that might indicate that levels in the fish were controlled by the geological settings of the lakes. Lake area and shoreline (perimeter) lengths (ignoring islands) for a number of the lakes were estimated but no significant relationships between these measures and levels of mercury in lake trout were found. The two very large lakes (Great Slave and Great Bear), however, had some of the lowest levels of mercury in the fish. Removing the two very large lakes from the data set increased the slope of the regression of mercury on lake area and shoreline length but still yielded insignificant relationships.

The question of the relative contributions of natural within-drainage geological mercury and mercury imported to a drainage via atmospheric deposition (both anthropogenic and natural) still has no definitive answer. The extent of geological information on mercury in lakes and streams in northern Canada is very limited. There is no doubt that mercury from local pollution sources can result in very high levels in the fish in the North (e.g., Giauque Lake, Discovery gold mine), but it is not clear whether the inputs of atmospheric mercury are sufficient to drive concentrations in the fish. Improved geological information, larger sample numbers of fish to allow for statistical compensation for size/age differences, and experimental work are required to accomplish that distinction.

Studies of lake trout, inconnu, burbot and round and broad whitefish in the Dogrib region surrounding Rae Lakes and Peel River, NWT supported the above conclusion for lake trout, and found that in burbot and round and broad whitefish the levels of mercury were generally low (Snowshoe, 2001). Inconnu had mercury levels in the 0.19–0.53 μg/g range that were in the same range as land-locked Arctic char (Snowshoe, 2001). It was found that in 2000 the mean concentrations of total Hg in northern pike muscle of (~ 1.50 μg/g) and Arctic grayling (~ 0.25 μg/g) from Yukon were about three times higher than in the same species from the Kuskokwim, Alaska (Naidu and Kelley, 2001). The difference for northern pike in the two rivers may be attributable to differences in age/size, which also influences the mercury concentration in this species (Lockhart *et al.*, 2001).

Arctic char

The broadest geographic perspective on mercury in fish in the Canadian Arctic comes from studies with Arctic char. Combined results from Fish Inspection Data and previous NCP studies (Muir and Lockhart, 1994; 1996; 1997; Muir *et al.*, 2000a; 2001a) are plotted in Figure 3.1.5 using ranges chosen based on guidelines for human consumption as discussed previously. All sea-run char have low mercury levels while some land-locked char have higher concentrations although in only one case, Amituk Lake on Cornwallis Island, were mean concentrations greater than 0.5 μg/g ww. Results for sea-run char are discussed further in Section 3.3.1.1.

In a separate study, 21 Canadian lakes were visited during June–September 1999 as part of the expedition Tundra North West 99 (Borg, 2001). The expedition covered the area from Baffin Island in the east to Ivvavik National Park in NW Yukon in the west, and Ellef Ringnes Island as the northernmost site (78 49′ N). Samples collected included water, suspended particles, bottom sediment profiles, zooplankton, and Arctic char. To date, metals have been analyzed in five individuals (Arctic char females) from six lakes and in lake sediments. The Arctic char were small (generally 20–100 g weight) and were not sea-run or came from lakes with stationary populations. The highest median mercury values were found in char from a lake on the Ungava Peninsula and from a lake in southern Ellesmere Island (near Muskox Fiord), but the variations of Hg concentrations between lakes were relatively small. Median concentrations of total mercury in the char from the six lakes ranged from 0.55–0.09 μg/g ww (Borg, 2001). Mercury concentrations in sediment in the lake in Ivvavik National Park were 20–40 times higher than in the other lakes, but the levels in fish muscle were still not elevated compared to the rest of the lakes.

Other metals

There are limited data on levels of metals other than mercury in freshwater fish. Some experimental evidence suggests that selenium can ameliorate the toxicity of mercury for at least some mammals (e.g., Eaton *et al.*, 1981) and for that reason selenium was measured in many of the fish analyzed for mercury (Annex Table 1). There was no clear relationship between levels of

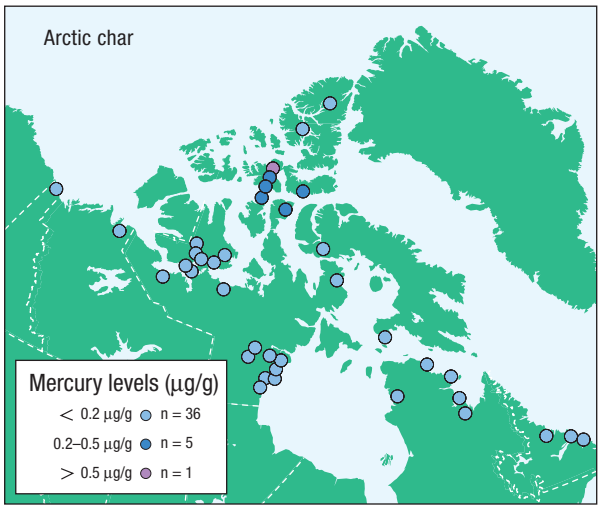

FIGURE 3.1.5

Mercury in both sea-run and land-locked Arctic char (Muir *et al.*, 2001a). Land-locked char are from high Arctic sites and western Ungava Bay.

mercury and those of selenium found in the fish. Stern *et al.* (2001a) included arsenic and selenium in studies of metals in burbot from Fort Good Hope. Arsenic and selenium were below levels of concern.

Land-locked char (from Kangiqsujuaq) had significantly higher levels of selenium than all sea-run char studied in Nunavik and Labrador (Muir *et al.*, 2000a). Age and size of fish had no significant effect on selenium or arsenic levels in muscle.

Muir *et al.* (2000b; 2001a) analyzed mercury and 22 metallic elements (by ICP-MS) in land-locked Arctic char from Resolute and Char Lakes near Qausuittuq and in three other lakes of the region (Annex Table 1). Statistical analysis showed that concentration data for most elements in char muscle from Resolute, Boomerang, and Sapphire Lakes were normally distributed. Results for Hg and other metallic elements in land-locked char from North, Sapphire and Boomerang Lakes were similar to those in Char and Resolute Lakes.

Mercury was strongly correlated with fish length and weight in all four lakes. Mercury was also significantly correlated with thallium (Tl), rubidium (Rb) and selenium in all lakes, but not consistently with any of the other 18 elements measured.

Seasonal trends of cadmium in char from Resolute Lake were studied in 1997 and 1998 by sampling three times during the open water season (Köck *et al.*, 1999). Results showed that cadmium levels in char increased over the summer corresponding to increased water temperature. Higher cadmium in liver in 1998 compared to 1997 coincides with higher water temperatures in August 1998 (2°C in 1997; 8°C in 1998). The mathematical relationship between lake temperature and metal uptake was similar in high Arctic lakes and previously studied Austrian high mountain lakes, confirming water temperature to be a key factor in metal accumulation in char (Köck *et al.*, 1996).

3.1.2 Levels and spatial trends of organochlorines in freshwater fish

A fairly large data set on OCs in freshwater fish and invertebrates was available for the first CACAR (Muir *et al.*, 1997). Since that report, there have been fewer data on OCs collected especially in comparison to the effort that has been made to measure mercury in fish. This is likely due in part to the low concentrations of OCs generally found in Arctic freshwater systems when compared to temperate and Arctic marine systems, as well as the lower level of concern for OCs compared to mercury. In general, concentrations (lipid corrected) of OCs in freshwater fish of the same species are similar throughout the Arctic (within a factor of two or three) (Annex Table 2), with a few exceptions that can be due to increased exposure or unique biological variation. PCBs and toxaphene continue to be the predominant OCs in freshwater fish.

Great Slave Lake study

A variety of studies have been conducted on OCs in biota of Great Slave Lake, most of which were conducted in the mid-1990s, but monitoring has continued for a number of fish species (Evans *et al.*, 2000; 2001). Great Slave Lake, one of Canada's northern Great Lakes, receives most of its water inputs via the Slave River, formed by the confluence of the Peace and Athabasca rivers. The latter two rivers transport tremendous amounts of sediments from the south, in addition to water into the Great Slave Lake ecosystem. Increased development to the south, therefore, may be resulting in increased contaminant transport to the lake through the Slave River in addition to input via the atmosphere.

OC concentrations were similar between four species of predatory fish (pike, walleye, burbot and inconnu) collected in the same region of Great Slave Lake (Annex Table 2), probably due to these species being predatory and feeding predominantly on fish. OC concentrations [ww and lipid weight (lw)], however, tended to occur in slightly higher concentrations in fish collected in the East Arm of Great Slave Lake compared with the West Basin in 1993 and 1999, although this varied somewhat with the chemical and/or species. The variation in concentrations between basins could be due to differences in the characteristics of the fish (e.g., length and/or age) collected for the two regions and/or to the characteristics of the two basins. OCs concentrations and sedimentation

rates tended to be higher in lake sediments from the West Basin than in the East Arm. Lake water in the East Arm is clear with low concentrations of particulates, and invertebrate biota such as mysids and amphipods are strongly benthic. In contrast, in the West Basin, waters are turbid with high concentrations of suspended silts and clays and invertebrates spending less time in close contact with the sediments than in the East Arm. Biota inhabiting the East Arm of Great Slave Lake may be accumulating higher body burdens of persistent organochlorines than in the West Basin because these contaminants are more bioavailable to them.

Lake trout

Concentrations of OCs in lake trout muscle from a number of lakes in Yukon and the NWT were studied in the late 1990s (Stern *et al.*, 2000; Palmer and Roach, 2001; Evans and Muir, 2001). Previous work on Yukon lakes in 1993 found that OC concentrations were very much higher in Lake Laberge, which was attributed to local point sources and a long food web (Kidd *et al.*, 1995). Since that time, concentrations in Lake Laberge lake trout have dropped up to four-fold over the five-year period from 1993 to 1998 (see Section 4.1.2). Lipid-adjusted OC levels in lake trout from Lake Laberge (1998 and 1999 collections only) are now comparable to results for other Yukon lakes, whereas previous Lake Laberge trout had distinctively higher levels (Figure 3.1.6). However, it should be noted that no effort was made to account for differences in biological characteristics (e.g., size of age) between the lake trout sampled in each lake or in Lake Laberge in different years. In some lakes,

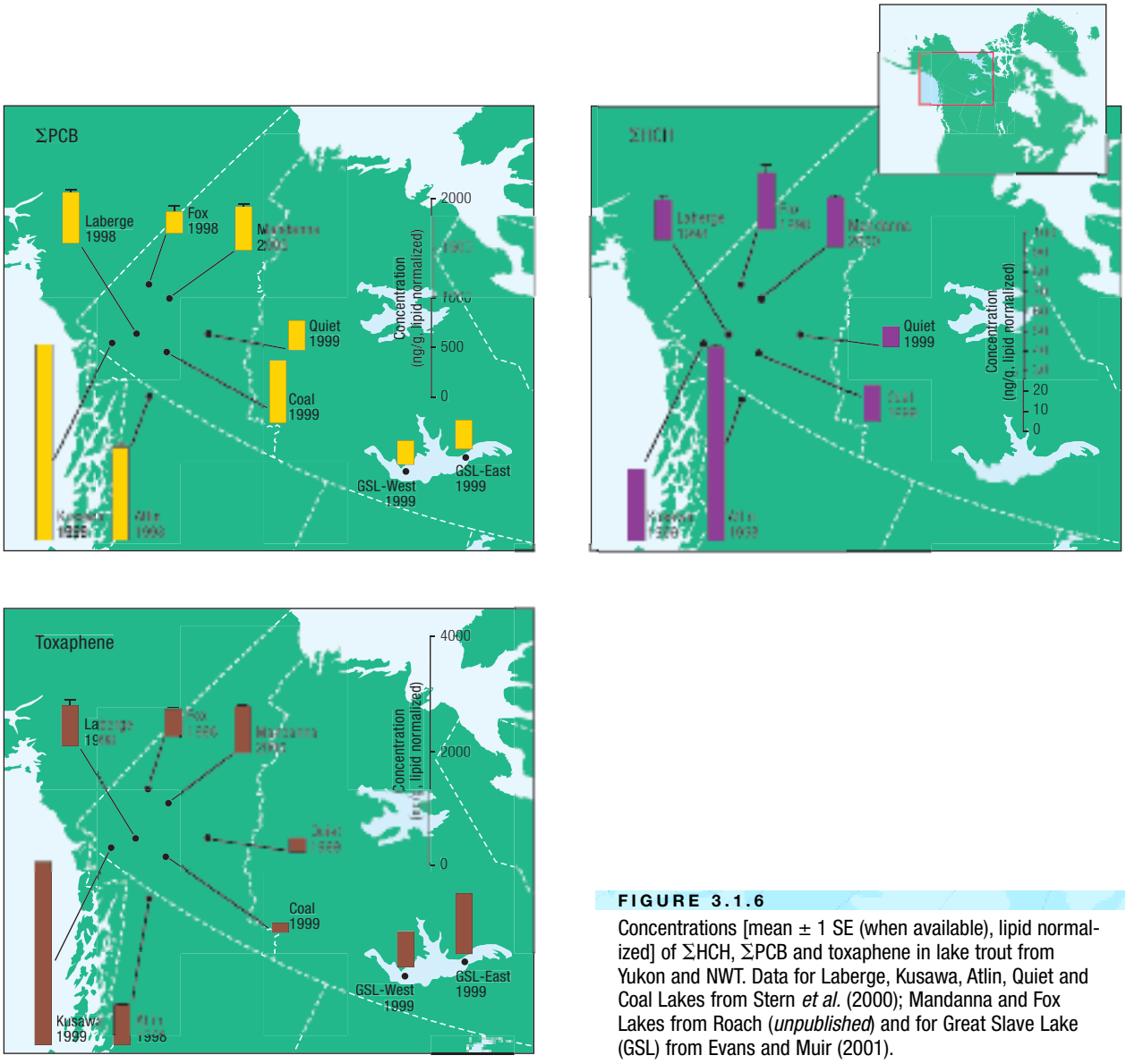

FIGURE 3.1.6

Concentrations [mean ± 1 SE (when available), lipid normalized] of ΣHCH, ΣPCB and toxaphene in lake trout from Yukon and NWT. Data for Laberge, Kusawa, Atlin, Quiet and Coal Lakes from Stern *et al.* (2000); Mandanna and Fox Lakes from Roach (*unpublished*) and for Great Slave Lake (GSL) from Evans and Muir (2001).

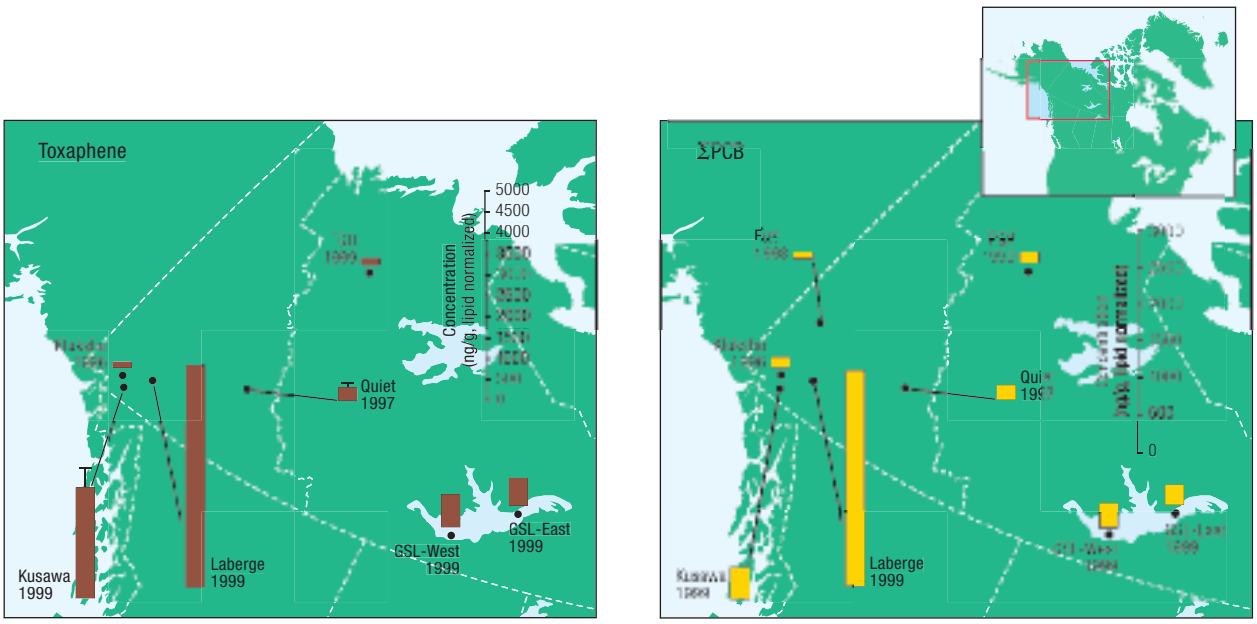

FIGURE 3.1.7

Concentrations [mean ± 1 SE (when available), lipid normalized] of ΣPCB and toxaphene in burbot liver from Yukon and NWT. Data for Fort Good Hope from Stern *et al.* (2001a); for Fox, Klukshu, Laberge, Kusawa and Quiet Lakes from Roach (*unpublished*) and for Great Slave Lake (GSL) from Evans and Muir (2001).

much smaller trout were sampled including Lake Laberge in 1998. It should also be noted that OC concentrations remain elevated in Lake Laberge burbot liver.

Concentrations of the more lipophilic contaminants such as toxaphene and PCBs were much higher in lake trout from Kusawa Lake (1998–1999) while lake trout from Atlin Lake had the highest concentrations of the less lipophilic organochlorines [ΣHCH and ΣCBz (chlorobenzenes)] (Figure 3.1.6). Lakes Kusawa and Atlin receive water from glacial melt, which may in part explain high levels in these lakes. Glacial melt has been implicated in the higher levels in some lakes of Alberta (Blais *et al.*, 2001). The levels in burbot liver from these two lakes, however, are not higher than those observed in other Yukon and NWT lakes (Figure 3.1.7). These data need to be assessed in light of the variable biological characteristics of the fish collected from each lake. For example, lipid was found to be a significant covariate of OC concentrations in Yukon lakes lake trout (Stern *et al.*, 2000). Concentrations (lipid corrected) of most OCs were higher in Yukon Lakes compared to Great Slave Lake (Figure 3.1.6).

Burbot

There is a fairly large data set of OCs in burbot (*Lota lota*) that was reported in the past CACAR assessment (Muir *et al.*, 1997). Levels of OCs in burbot have received attention because indigenous people consume the high lipid livers of this species which has a wide distribution. Burbot are also a predatory, bottom-feeding species. Its sedentary nature and the high lipid content of the liver make burbot very suitable for monitoring lipophilic contaminants (Muir and Lockhart, 1996; de March *et al.*, 1998). There has also been concern in the past about the quality of the liver from burbot in the Mackenzie River at Fort Good Hope.

Elevated PCBs, toxaphene and other persistent OCs continue to distinguish burbot in Lake Laberge from other lakes in the NWT and Yukon (Figure 3.1.7). Atmospheric deposition is the dominant route of OC delivery to these lakes, although this may not be the case for Lake Laberge (Kidd *et al.*, 1995). The high levels of OCs in the Lake Laberge burbot agree with past work on fish from this lake (Kidd *et al.*, 1995; 1998). To thoroughly examine the differences shown in Figure 3.1.7, the recently collected data needs to be statistically evaluated with consideration for the biological characteristics of the fish. The OCs concentrations in these burbot are lower than those previously reported for burbot collected from the Mackenzie River in the Canadian Arctic in the 1980s (Muir *et al.*, 1990a). All of these sites are fairly close to the Mackenzie River so the lower concentrations in the recently collected burbot likely reflect temporal changes as opposed to spatial differences between the sites (see Section 4.1.2).

Land-locked Arctic char of Resolute Lake

Land-locked char have been collected over a number of years from lakes near the community of Qausuittuq (Resolute) in the Canadian High Arctic (Muir *et al.*, 2001a). As detailed in Section 3.1.1.2, the effect of diet and trophic level on mercury and organochlorine levels was investigated using stable isotopes of carbon and nitrogen. A few fish have $\delta^{15}N$ values that were up to 3.8‰ higher than others, which is associated with a change of trophic level because concentrations of the heavier isotope of nitrogen, ^{15}N, are progressively enriched from prey to predator an average of 3 to 5‰ (Peterson and Fry, 1987). ΣPCB, ΣDDT and ΣCHL concentrations (lipid normalized) were significantly correlated with $\delta^{15}N$ in char from Resolute Lake collected in 1997 and 1999, suggesting that biomagnification of OCs is occurring within the char population due to the presence of piscivorous char. These results support the hypothesis that the higher levels seen in char sampled in 1993 in Char Lake (Muir *et al.*, 2000b) are because piscivorous char were analyzed.

Yukon River study

A number of fish species have been sampled and analyzed for OCs from headwater lakes in Yukon River system since 1996 in response to higher concentrations of toxaphene (Palmer, 1999; Palmer and Roach, 2001). Results for lake trout and burbot are covered in previous sections. OC concentrations in whitefish collected in 1998 from Lake Laberge are lower than those observed in lake trout based on wet weight but similar when concentrations are normalized to lipid (Annex Table 1). OC concentrations in Lake Laberge are lower than those observed in Watson Lake (collected in 1997 and 1998), although the Watson Lake whitefish are larger. OC concentrations in inconnu collected from Peel River in 1999 are low compared to most other species of fish from Yukon region but are similar to inconnu collected in Great Slave Lake in 1996 (Annex Table 1). Concentrations of OCs in fish from Yukon region are consistently below levels that result in any health advisories (Palmer, 1999).

3.1.3 Levels of new chemicals in freshwater fish

There have been few recent studies on levels of new chemicals in freshwater fish. The only new chemicals that have been measured are polybrominated diphenyl ethers (PBDEs), a brominated flame retardant, in burbot from Fort Good Hope (Stern *et al.*, 2001a) (Annex Table 2). Liver samples from burbot collected in 1988, 1999 and 2000 at Fort Good Hope (Rampart Rapids), NWT were analyzed for PBDE congeners as part of a project on temporal trends (Stern *et al.*, 2001a) (see Section 4.1.2). Concentrations of ΣPBDE (5 congeners) were 1.6 ± 1.1 ng/g, (ww, mean ± 1 SD). PBDE 47 was the most predominant PBDE congener residue in the burbot liver (0.58 ± 0.52 ng/g, ww, mean ± 1 SD) followed by PBDE 99 (0.37 ± 0.27 ng/g), 100 (0.21 ± 0.16 ng/g), 153 (0.16 ± 0.13 ng/g) and 154 (0.16 ± 0.12 ng/g). A similar profile was observed in Pangnirtung beluga (Stern and Ikonomou, 2000). In comparison, Jansson *et al.* (1993) reported mean PBDE 47 and 99 concentrations of 0.099 and 0.031 ng/g (ww) in whitefish muscle from Lake Storvindeln and 21 and 3.4 ng/g (ww) in Arctic char from Lake Vattern, Sweden. Levels of ΣPBDE in the burbot are orders of magnitude less than ΣPCB, although a much larger number of PCB congeners were measured than PBDEs. Levels of the more common individual PBDEs are approaching levels of some of common PCB congeners.

3.1.4 Overall assessment of contaminants in freshwater biota

A large data set of mercury levels in freshwater fish from the Canadian Arctic has been generated since the first CACAR assessment. These data, along with data collected previously, have increased the understanding of variables influencing contaminant levels in Arctic freshwater fish, but knowledge gaps remain and new questions have been raised. Spatial coverage of mercury levels in a number of freshwater fish species, in particular lake trout, in the Canadian Arctic is extensive and comprehensive. Coverage of OCs is less complete but OC levels appear to be less variable than mercury concentrations and of less concern.

Mercury levels in freshwater fish species in a number of Arctic lakes exceed the maximum recommended concentration of 0.5 μg/g (ww) for commercial sale in Canada. A smaller number of lakes also have levels in fish that exceed the more restrictive recommendation of 0.2 μg/g that applies when people eat large amounts of fish such as in subsistence fishing. These high levels are mostly limited to predatory fish that are exposed via the biomagnification of methylmercury in aquatic food webs. These high mercury levels are geographically extensive but variable. Efforts to explain mercury levels in fish based on larger scale geographic (e.g., lake size) or geological variables have not been successful. Other variables, such as humic content and fish size and age, have been found to influence mercury levels observed in fish. The influence of natural versus anthropogenic sources on levels of mercury observed in fish continues to be an important but unanswered question and may contribute to geographic trends.

Mercury levels in freshwater fish are higher than those observed in sea-run fish. For example, sea-run Arctic char have lower levels of mercury than freshwater char.

Information and data on other metals in freshwater fish is limited. Arsenic and selenium were the most prominent of the five elements measured in sea-run Arctic char samples. Arsenic concentrations are much higher than mercury; however, speciation of arsenic in char has not been examined. In land-locked char, mercury was significantly correlated with thallium, rubidium, and selenium, but not consistently with any of the 18 other elements measured. The significance of these correlations and possible links to long-range transport of, for example, thallium (a highly toxic and bioaccumulating metal that is associated with mining operations), has not been examined.

Levels of OCs in Arctic freshwater fish are lower than levels observed in marine fish and are generally below guideline levels. PCBs and toxaphene are the predominant OCs measured. Geographic trends of OCs in the Canadian Arctic are generally minor but a few systems have elevated concentrations. For example elevated levels of PCBs and toxaphene have been found in predatory fish from Lakes Kusawa and Atlin in Yukon compared to other Arctic lakes. The coverage of OCs in freshwater fish is not as extensive as that seen for mercury, in part because analysis for OCs is more costly and there is less concern about OCs than for mercury. Levels of OCs are well below those observed in fish from temperate regions such as the Great Lakes and there is little or no concern about the effects of OCs on fish health.

There are a number of large data sets on OCs in fish that need to be compiled and statistically analyzed to assess factors that influence OC concentrations, similar to what has been done for mercury. Variables such as lake size, year of collection, and influence of glacial melt could be assessed.


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There is relatively little information on levels of new chemicals in freshwater fish. Data on PBDEs are available for burbot collected at Fort Good Hope. Although levels are much lower than PCBs, there are fewer compounds measured and levels are increasing with time. Information on spatial trends for these compounds is not available and remains a data gap. Information on other new chemicals, such as perfluorinated acids, does not exist.

3.2 Terrestrial biota-levels and spatial trends

Many other studies have been done on OCs, metals and radionuclides and were summarized in the first CACAR (Muir *et al.*, 1997). The spatial coverage of terrestrial mammals and birds was considered "quite complete". In general, pollutant concentrations in these animals were considered low, although relatively high cadmium concentrations were noted in the kidney and liver of caribou from Yukon. Some geographic patterns were noted in caribou and mink and concerns about contamination at DEW line sites were noted. The amount of data generated on terrestrial mammals and birds since the first CACAR was published has been limited, because the levels of contaminants found were considered to be low. Most of the studies since that time have focussed on waterfowl and game birds.

3.2.1 Contaminants in caribou and other big game mammals

Caribou (*Rangifer tarandus*) are a major part of the social and cultural makeup of Aboriginal and northern culture in North America and they comprise a large portion of traditional diets in some areas of the North. There is some continuing concern about the presence of contaminants in caribou, and their potential effect on human and caribou health. Although organic contaminants such as DDE, PCBs, dioxins and furans remain low in large terrestrial mammals (Elkin and Bethke, 1995; Hebert *et al.*, 1996; Thomas *et al.*, 1992), inorganic contaminants like cadmium, lead and mercury, and radionuclides, have been elevated under some conditions. The contribution of caribou to total intake of mercury, cadmium and lead may be relatively small (< 10%) in Inuit communities that also consume large amounts of marine mammals (Chan *et al.*, 1995), but moose and caribou liver can comprise up to 90% of the total cadmium ingested in the Dene/Métis traditional diet in the Northwest Territories (Berti *et al.*, 1998). There is also

concern that the high metal levels in organs may lead directly or indirectly to disease in the caribou.

Data from two large monitoring programs [Government of the Northwest Territories Department of Resources, Wildlife and Economic Development (RWED) in the NWT and Nunavut, and the Yukon Contaminants Committee (YCC)] were analyzed for spatial and temporal trends, and the influence of factors such as gender, age, tissue and season of collection on the levels of a broad range of inorganic elements (Macdonald *et al.*, 2002). This analysis found that OCs remain low in all herds (Braune *et al.*, 1999a; Thomas *et al.*, 1992), often near detection limits. As a result, metals and radionuclides have become the major focus of most human and ecological risk studies involving caribou. Data and trends collected during the 1990s for a large number of herds demonstrated that metal levels can change markedly by region, time of year and age of the animal.

Metal data for kidney and liver were available from a total of 15 herds sampled during the 1990s, including multiple collections for some herds (Macdonald *et al.*, 2002) (Annex Table 3). In the early stages of the surveys, elements were analyzed individually; however, recently there has been an increased use of multi-element analysis, including high resolution ICP-MS. The concentration of As, Ba, Be, B, Co, Mo, Ni, Se, Si, Sr, Te, Th, Sn, Tl, U and V were below detection limits in most herds but were present above detection limits in over half the individuals in at least one herd. Of the major elements [Al, Cd, Cu, Cr, Fe, Pb, Mn, Hg_T (total mercury), Zn], only cadmium, total mercury and zinc increased significantly with age. Age-adjusted least square means for cadmium showed significantly higher concentrations in kidney in herds from Yukon, moderate levels in central areas and low concentration in more northern areas (Figure 3.2.1). Total mercury showed a different trend than cadmium, with the highest concentrations in the central Canadian Arctic Beverly herd and the eastern Arctic South Baffin population (Figure 3.2.1). The pattern for total mercury corresponded with estimates from sediment records suggesting higher rates of anthropogenically-derived total mercury in the central and northeastern regions of northern Canada.

All metals showed significant differences between herds and tissues, but aluminium, cadmium, chromium, total mercury and manganese showed no significant differences in concentration between sexes. Copper was significantly higher in liver, an important metabolic reservoir, than kidney in all herds; however, the kidney:liver ratio ranged from 0.17 to 1. Arsenic was below detection limits in most herds, but the mean levels > 1 µg/g dry weight (dw) in the Porcupine herd suggest that the consumption of kidney and liver may be a significant source of As in some communities. Cadmium and total mercury were significantly higher in the spring than fall, consistent with lower lipid levels and organ weights in spring than fall. Copper concentrations were significantly higher in the fall than in the spring, probably due to an accumulation of the micronutrient over the summer.

Yukon big game and wolves

Gamberg *et al.* (1999) examined the levels of heavy metals in a number of Yukon big game animals. They concluded that metal concentrations in moose (*Alces alces*), caribou, bison (*Bison bison*) and mule deer (*Odocoileus hemionus*) in this study should be considered baseline levels. Yukon moose and some caribou showed higher renal cadmium concentrations than in the same species

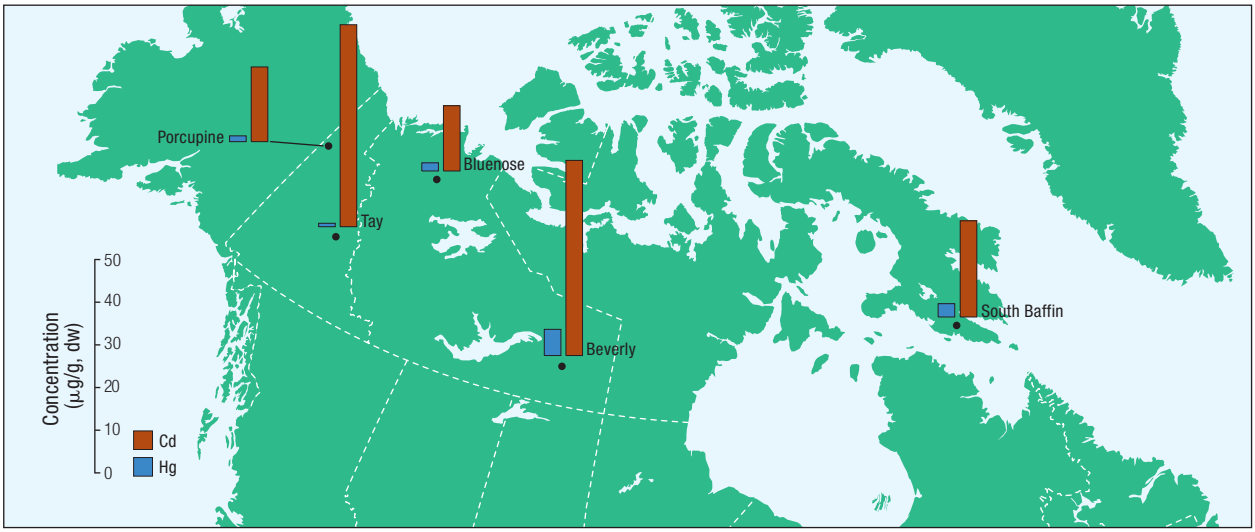

FIGURE 3.2.1
Cadmium and mercury in the liver of caribou from the Canadian Arctic (Macdonald *et al.*, 2002).

from other areas, and caribou had higher renal mercury levels than the other species studied. It is likely that both these contaminants are entering the food chain from natural mineralizations. There are insufficient data to explore geographic trends in Yukon.

Levels of OCs were measured in livers of wolves collected in the Canadian Yukon (Gamberg and Braune, 1999). Concentrations were found to be very low (mean or median < 50 ng/g ww) compared with marine mammals but were slightly higher than recently measured OCs in the livers of Russian wolves (Shore *et al.*, 2001). In many of the samples taken, common OCs, such as *p,p′*-DDE, were not detected. A small number of highly chlorinated PCB congeners dominated the PCB load in both studies reflecting the high biotransformation capacity of terrestrial carnivores. OC were not found to vary between sexes and, with the exception of PCBs, did not vary with age. The study concluded that levels of OCs do not pose a threat to the health of this wolf population.

Wolverines

Wolverines are omnivorous terrestrial mammals that live throughout the alpine and Arctic tundra of Canada. Livers from 12 wolverines were collected at Kugluktuk, Nunavut in the western Canadian Arctic to determine, for the first time, the residue patterns of OCs in this species (Hoekstra *et al.*, 2002a) (Annex Table 4). The rank order of hepatic concentrations for sum (Σ) OC groups in wolverines were ΣPCB > ΣCHL > ΣDDT > ΣHCH > ΣCBz. The most abundant OC analytes detected in wolverine liver (in order from highest to lowest) were CB153, CB180, oxychlordane, CB138, *p,p′*-DDE, CBs170/190, CB99, CB118, and dieldrin. These OCs are among the most recalcitrant and are also the predominant OCs in other terrestrial Arctic mammalian predators, such as wolves (Gamberg and Braune, 1999). Wolverine age and gender did not influence OC concentrations.

Levels of chlordane and HCHs were much lower in the wolverines compared to the Arctic fox collected to the north in Ulukhaqtuuq (Hoekstra *et al.*, 2002a) but are within the range of levels found in other Arctic terrestrial mammals. This is expected because the Arctic fox is known to scavenge marine mammals, resulting in elevated OCs (Klobes *et al.* 1998). Levels of PCBs and DDTs, however, were similar or higher in the wolverine than in the Arctic fox. Concentrations of these OCs in the wolverine are much higher than in other Arctic terrestrial mammals (Annex Table 4). Although these PCB and DDT levels are below threshold effects levels (Hoekstra *et al.*, 2002a), an explanation for these unexpectedly high levels is not evident and merits further research.

3.2.2 Levels of organochlorines and metals in mammals associated with freshwater

Beaver (*Castor canadenis*) and muskrat (*Ondatra zibethicus*) meat is an important part of the traditional diet of the Dene in the Northwest Territories. In 1998 and 2001, beaver and muskrat samples were collected by local trappers, and analysis was completed to determine levels of OCs and heavy metals in muscle and organs at the southern end (Slave River Delta) and northern end (Mackenzie River Delta) of the Mackenzie River watershed, respectively (Annex Tables 3 and 4). For organochlorine analysis, beaver and muskrat muscle (Slave River Delta only) and liver samples were pooled by species, sex and location. Overall, PCBs, DDT and chlordane levels were very low and well below available guideline levels. Other organochlorines were below detection limits. Beaver and muskrat muscle, kidney (Mackenzie River Delta only), and liver were analyzed for heavy metals. Levels of metals in beaver and muskrat muscle (meat) were very low or considered normal for terrestrial wildlife. Levels of cadmium in beaver liver and kidney at the Mackenzie River delta were 10.32 μg/g dw and 55.46 μg/g dw, respectively (Snowshoe, 2002; personal communication). Levels of cadmium in beaver liver at the Slave River Delta were 6.6 μg/g dw (Kennedy, 1999). Beaver livers and kidneys have levels of cadmium that reflect natural background levels and are consistent with other terrestrial wildlife levels.

This study complements both the Fort Resolution and Yukon studies and has provided useful baseline data on beaver and muskrat in the Mackenzie Delta. Overall levels in the Mackenzie River Delta were similar to those in the Slave River Delta for both beaver and muskrat. Since beaver liver and kidney are not consumed by the Gwich'in residents of the Mackenzie River Delta, a health assessment for these organs was deemed unnecessary (Snowshoe, 2002; personal communication).

3.2.3 Spatial trends of contaminants in waterfowl and game birds

King (*Somateria spectabilis*) and common (*Somateria mollissima*) eider ducks are important traditional foods of Aboriginal people in certain Arctic communities (Gilchrist and Dickson, 1999; Fabijan *et al.*, 1997). Populations of eiders have declined precipitously in recent years, raising concerns about the sustainability of the harvest (Gratto-Trevor *et al.*, 1998; Robertson and

Gilchrist, 1998; Suydam *et al.*, 2000). Contaminants are believed to be one of several risk factors that may be contributing to these declines (CWS *et al.*, 1997). Trace elements, in particular cadmium, selenium and, to a lesser degree, mercury have been found at elevated concentrations in eiders (Norheim, 1987; Nielsen and Dietz, 1989; Henny *et al.*, 1995; Dietz *et al.*, 1996; Hollmén *et al.*, 1998; Franson *et al.*, 2000; Trust *et al.*, 2000). Concentrations of selected trace elements (Cd, Hg_T, organic Hg, Se, Cu and Zn) were determined in livers or kidneys of eider ducks, king and common, at three locations in the Canadian Arctic (Wayland, 1999; 2000) (Annex Table 3). Ducks were collected in June 1997 at the East Bay Migratory Bird Sanctuary on Southampton Island, NU, near the village of Holman on Victoria Island, NWT and near Belcher Island in Hudson Bay (Figure 3.2.2). All birds were females with the exception of four king eiders collected at Southampton Island. Cadmium was analyzed in both kidney and liver, total and organic mercury, selenium, copper and zinc were analyzed in livers.

There was a significant interaction between species and location in concentrations of trace elements, indicating that the spatial trend in trace element concentrations differed between king and common eiders. This was primarily because, at Southampton Island, king eiders had higher concentrations of cadmium than common eiders whereas at Victoria Island, concentrations of cadmium in the two species were similar (Figure 3.2.2). The concentrations of cadmium in kidneys of king eiders at Southampton Island (GM: 41; range: 31–57 mg/g ww) were among the highest ever recorded in eider ducks and 4–10 times higher than in concentrations measured in eiders from Europe (Dietz *et al.*, 1996; Nielsen and Dietz, 1989). In Alaska, the mean renal cadmium concentration in spectacled eiders was approximately 23 μg/g ww (Trust *et al.*, 2000). The relatively high cadmium concentrations in king eiders from Southampton Island were consistent with reports of high cadmium levels in marine animals from the eastern Canadian Arctic (Braune *et al.*, 1991; Muir *et al.*, 1997), which has been attributed to elevated levels of natural cadmium in the region's bedrock (Muir *et al.*, 1997). However, the relatively low concentrations of cadmium in common eiders from Southampton Island contrasted with results for marine animals (Braune *et al.*, 1991; Muir *et al.*, 1997). Other factors, unrelated to bedrock geology, may have influenced results. For example, after adulthood is reached, cadmium continues to accumulate with age in some species of seabirds (Furness and Hutton, 1979) but not in others (Stewart and Furness, 1998). If cadmium accumulates with age in adult eiders, then some of the inter-location variation in cadmium concentrations in this study may be attributed to differences in the ages of the birds that were collected.

Total and organic mercury concentrations were higher in eiders from Southampton Island than in those from Victoria Island (Figure 3.2.2). The percentage of total mercury that was in the organic form averaged 82% and ranged from 41–100% and is typical of seabirds exposed to relatively low levels of total mercury (Dietz *et al.*, 1990). This contrasts somewhat with results of Braune *et al.* (1991) and Muir *et al.* (1997), who found that mercury levels were higher in marine mammals from the western Canadian Arctic than in those from the eastern Arctic. Variation in mercury levels in eiders may have been influenced by their exposure to mercury in wintering areas. Eiders from Victoria Island spend much of the year in the northern Bering and southern Chukchi Seas. The low mercury levels in eiders from Victoria Island were similar to those found in spectacled eiders wintering near St. Lawrence Island in the Bering Sea (Trust *et al.*, 2000). Furthermore, mercury levels in livers of polar bears (*Ursus maritimus*) from the western portion of Alaska bordering the Bering and Chukchi Seas were approximately 6–7 times lower than those in bears from northern Alaska bordering the Beaufort Sea (Lentfer and Galster, 1987). This supports the contention that mercury levels in eiders collected at Victoria Island were relatively low because their exposure to mercury was low on their wintering areas in the northern Bering Sea, Bering Strait and the southern Chukchi Sea.

Selenium concentrations were significantly higher in livers of eiders from Victoria Island than in those of eiders from Southampton Island or the Belcher Islands (Figure 3.2.2), consistent with spatial trends in marine mammals (Braune *et al.*, 1991; Muir *et al.*, 1997). It is possible that exposure of eiders to high levels of selenium during their prolonged period of residency in the Bering Sea may have some residual effect on tissue selenium concentrations during spring migration through Victoria Island.

King eiders had significantly higher concentrations of mercury, organic mercury and zinc than common eiders, while, as indicated previously, king eiders from Southampton Island had higher cadmium concentrations than common eiders from that location (Figure 3.2.2). Foraging habitat and dietary segregation between the two species may account, at least partially, for the observed differences in their trace element concentrations. Both species feed heavily on mussels; however, whereas common eiders are mussel specialists, king eiders consume a more varied diet including not only mussels but echinoderms and other benthic invertebrates as well (Bustnes and Erikstad, 1988; Frimer, 1997).

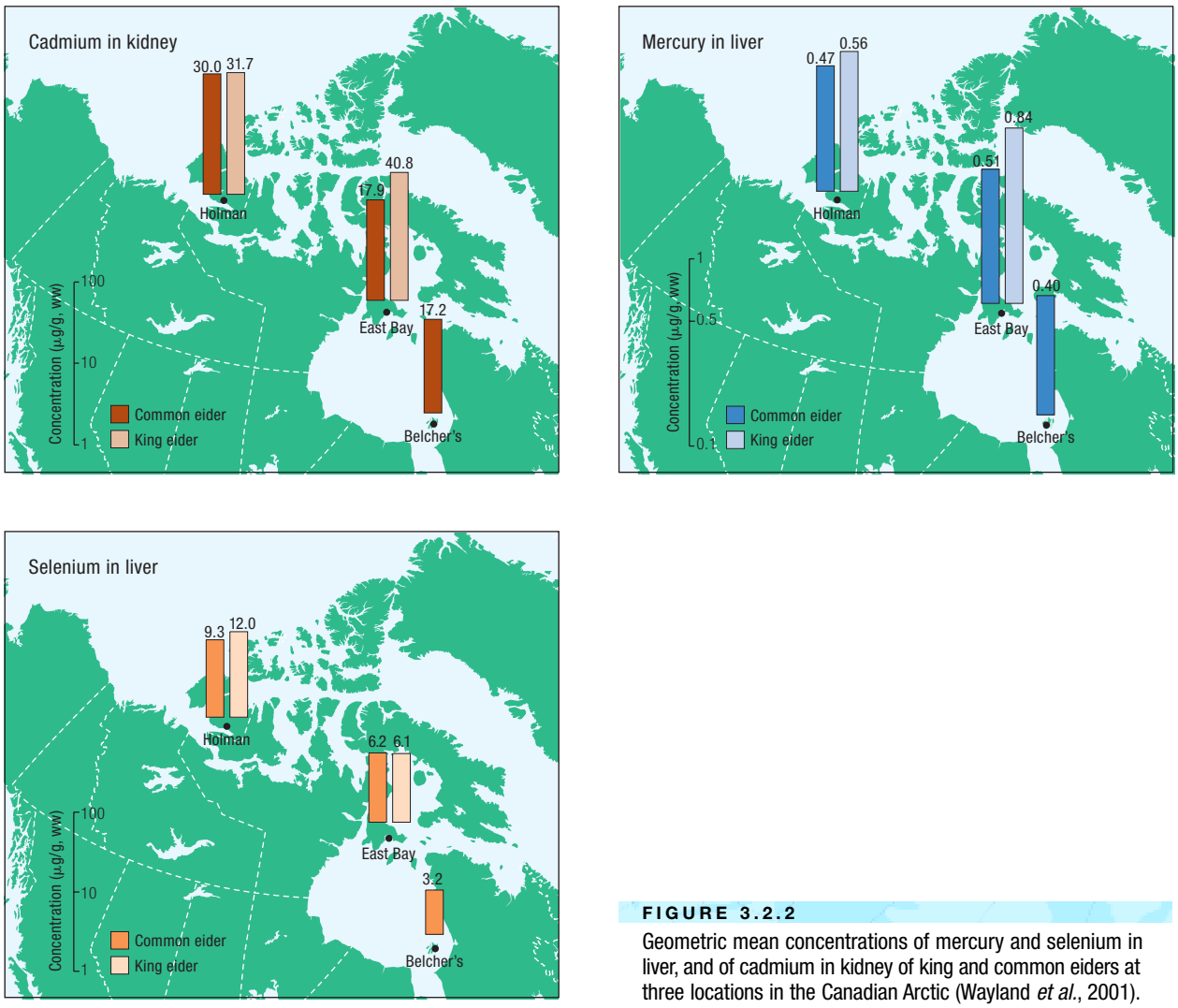

FIGURE 3.2.2

Geometric mean concentrations of mercury and selenium in liver, and of cadmium in kidney of king and common eiders at three locations in the Canadian Arctic (Wayland *et al.*, 2001).

Cadmium was analyzed in liver and kidney of 53 eiders collected in 1997. The regression of liver cadmium concentration on that in kidney was significant. The regression equation [log Cd in kidney = 0.88 + 0.71 (log Cd in liver)] was very similar to one determined in surf scoters (*Melanitta perspicillata*) in California (Ohlendorf *et al.*, 1991). The ratio of cadmium in liver to that in kidney averaged 0.23 and ranged from 0.09–0.61, a range that is indicative of chronic exposure to low levels of cadmium (Scheuhammer, 1987).

3.2.4 Contaminants in plants

Consistent with terrestrial birds and mammals, the levels of OCs and metals in plants in the Canadian Arctic are generally low and subsequently there have been few studies.

Davey *et al.* (1999) examined the levels of arsenic (As) in traditional foods of the Weledeh Yellowknives Dene in response to concerns about local gold mining activity. This study showed statistically higher levels of As in five species of berries (blueberries, cranberries, raspberries, rose hips, gooseberries) from areas around gold mines (and close to these mines) than from the control sites. The results suggest that gold mining has an effect on As levels in these species of berries in the Yellowknives Dene traditional territory. Most berries harvested at the mine sites and some harvested within the city of Yellowknife are above the recommended concentration of 0.1 µg/g for consumption. Proximity to the active mine sites appears to be a significant factor determining the level of As in berries in this region, as berries from the Dettah sites, further away from the mine sites than the town samples, were statistically lower in As than those from the city of Yellowknife. Cloudberries were analyzed in this study but there were insufficient samples to determine if mining has affected this species.

3.2.5 Local sources of contaminants in the terrestrial environment

In addition to PCBs, some former military sites received significant DDT applications. The bioavailability of this localized DDT contamination to the terrestrial Arctic environment was examined in a study at an abandoned Long Range Aid to Navigation (LORAN) station located at Kittigazuit, Northwest Territories (69°16′56″ N, 133°54′32″ W) in the western Canadian Arctic (Nirwal, 2001). The study site received applications of DDT between 1948 and 1950. Despite the passage of time, soil concentrations have remained high (maximum ΣDDT 210,000 ng/g dw), and the composition of ΣDDT compounds in soil still resembled the original pesticide formulation (59% *p,p′*-DDT). In soils, appreciable loss and degradation of DDT was less pronounced than that in temperate and tropical environments. Samples of soil, sediment, willow (*Salix* sp.), grass (*Elymus* sp.), and Arctic ground squirrel (*Spermophilus parryii*) were collected at the LORAN station and at a nearby reindeer herding camp. Concentrations of ΣDDT at the station ranged from 62–210,000 ng/g with highest concentrations found within an area of approximately 4,024 m^2; a smaller area (386 m^2) at the herding camp also had elevated concentrations. Samples of *Elymus* sp. and *Salix* sp. collected from the station had a higher median ΣDDT concentration compared to the area near the herding camp or to background sites. Higher ΣDDT was found in *Salix* sp. (12–10,000 ng/g dw) compared to *Elymus* sp. (3.2–1,500 ng/g dw). The concentration and composition of ΣDDT in Arctic ground squirrels livers were clearly a result of contamination at the study site. Liver concentrations at contaminated areas (maximum ΣDDT 4,300 ng/g lw) declined to background levels (maximum ΣDDT 4.5 ng/g lw) with increasing distance from contaminated areas. Estimated contaminant exposures were below no-observed-effect levels, but a significant relationship between liver size and ΣDDT concentration was found. The concentrations in ground squirrels from background sites in this study were below concentrations reported for Arctic ground squirrels in Alaska (Allen-Gil *et al.*, 1997).

The contribution of atmospheric dispersal and transport at Kittigazuit was negligible because an abrupt transition exists between soil contaminant levels at the sites, and samples collected immediately off site. For example, the median ΣDDT concentrations were 1,300 ng/g dw at the station, and 540 ng/g dw at the camp. In comparison, results for soil samples considered representative of background conditions (3–10 km off site) were below the analytical detection limit of 20 ng/g dw.

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A scenario comparable to that at Kittigazuit was studied near Fort Churchill, Manitoba, on the western coast of Hudson Bay. Three years following aerial spraying for mosquito control, sampling within treated areas showed that the livers of collared lemmings (*Dicrostonyx groenlandicus*) contained 5,400–41,000 ng/g lw of ΣDDT and red squirrels 7,400–17,000 ng/g lw (Brown and Brown, 1970) indicating the persistence and bioavailability of DDT in other terrestrial Arctic environments.

3.2.6 Overall assessment of contaminants in terrestrial biota

Conclusions from the first CACAR biota report revealed that there had been a significant amount of research on contaminant levels in the Canadian Arctic terrestrial environment, and that spatial coverage of OCs and metals was quite complete (Muir *et al.*, 1997). Since that report (1996–2001), there have been few studies on contaminants in the terrestrial environment, which is likely due to the low levels initially found. Radioisotope and heavy metals are of greater concern than OCs in this environment.

A comprehensive examination of contaminants in caribou collected from across the Canadian Arctic concluded that levels of OCs are low and of little concern to the health of the caribou or people who consume them. Mercury, cadmium and lead remain somewhat of a concern for some populations of caribou. In general, metals varied with tissue type, herd and season. Levels of mercury, cadmium and zinc increased with age. Cadmium levels in kidney were higher in Yukon caribou and lowest in the most northern herds, whereas mercury was higher in the central Arctic herds. Copper may be a limiting element in some herds but this may be a

non-anthropogenic effect. In a separate study, metals in big game from Yukon were low and were considered to be baseline.

Concentrations of OCs in Yukon wolves and NWT wolverines were higher than what is observed in terrestrial herbivores, e.g., caribou, but much lower than their marine counterparts. There is little concern about the effects of these levels on the health of wolves and wolverines, and since they are not consumed by humans, there is no concern for human health.

Levels of OCs were low in freshwater mammals, beaver and muskrat, collected in the Mackenzie River valley were low. Levels of metals in these animals are considered to be normal for terrestrial mammals. Neither OCs nor metals are considered a health concern for the animals or consumers.

Metals in Arctic waterfowl varied with the species and location. Cadmium levels in king eider from the eastern Arctic were higher than levels measured in Alaska or Europe but this may be due to local non-anthropogenic sources. Mercury was found to be higher in the central Arctic eiders and this is likely due to accumulation in overwintering habitats.

Data are very limited for plants. A single study found low levels of As in five species of berry. Based on past work in Canada and more recent work in other Arctic regions there is little concern about levels of contaminants in Arctic plants. There may be an issue in cases of local contamination, such as at DEW-line sites.

To date no effort has been made to assess levels of new chemicals in the Arctic terrestrial environment. However, since one of the major criteria for identifying these new chemicals is bioaccumulation and biomagnification potential, it is likely that, compounds with similar properties to the legacy OCs would only achieve low levels in the terrestrial food web.

3.3 Spatial trends of contaminants in marine biota

Presence of OCs in the marine environment has historically received the most attention in the Canadian Arctic environment. The marine environment was the first Arctic system to be examined for the presence of OC contaminants (Holden, 1970). The level of research on OC contamination of the marine environment has continued to outdistance studies of contaminants in the freshwater and terrestrial Arctic. This is clearly due to the higher levels observed in this system and the importance of marine organisms to northern communities.

Since the first CACAR (i.e., post 1996), a large amount of OC data has been produced for the marine environment. A number of studies have been initiated on levels and dynamics of OCs and metals in seals and whales and a number of comprehensive studies on the dynamics and mechanisms of OCs and metals in the marine food web have been completed.

Studies on metals in the marine environment has focussed predominantly on marine mammals. Metals are naturally occurring elements whose levels in the environment have been augmented by human activities. Most Arctic metal contaminant work has focussed on mercury and to a lesser extent cadmium, selenium and lead (Bard, 1999). Mercury and cadmium are not essential elements, can be toxic in small quantities, potentially biomagnify in the food chain, and are present at high levels in some Arctic biota. Mercury is of concern because there are indications that levels have been increasing in the Arctic marine environment over the past few decades (Section 4.3.3).

3.3.1 Contaminants in invertebrates and marine fish

Marine invertebrates provide a link between phytoplankton and fish, seabirds and mammals in Arctic marine food webs. These invertebrates are therefore important in the transfer of carbon and nutrients but also OCs and metals to organisms at upper trophic levels. Marine and anadromous fish occupy a range of trophic levels in Arctic marine ecosystems and hence concentrations of contaminants in their tissues can be quite variable. The Arctic cod (*Boreogadus saida*) is a key link in marine food webs, between invertebrates and seabird and marine mammals, as a major diet item of many important Arctic marine mammals, including the ringed seal and beluga. An understanding of the trends and dynamics of OCs and metals in marine invertebrates and fish are important for understanding the trends of contaminants in Arctic marine ecosystems. A number of fish species are important components of the traditional human diet, e.g., Arctic char, and some have become important commercial species, e.g., Greenland halibut, turbot (*Reinhardtius hippoglossoides*), but few Arctic invertebrates are consumed by humans.

Historically there have been few studies of OCs and metals in marine invertebrates and fish, at least compared to marine mammals and seabirds, which is likely due to the low contaminant levels observed in these organisms and the fact that they are rarely consumed by humans. Recently, however, there have been a number of studies that examined contaminants in these organisms.

3.3.1.1 Metals in invertebrates and fish

Invertebrates

Temporal and spatial data sets for blue mussels (*Mytilus edulis*), ranging from a few years to long-term spans of several millennia, have recently been generated (Outridge *et al.*, 2000). Significant north-south variations in the relative inputs of anthropogenic versus natural mercury and lead in bivalve species at sites across the Canadian Arctic were reported. At two high-latitude sites, Expedition Fiord on Axel Heiberg Island, and Resolute Bay on Cornwallis Island, long-term sequences of bivalve shells collected from Holocene [1,000–9,000 years. before present (BP)] beach ridges contained similar concentrations of mercury, lead and cadmium to modern (1980s–1990s) collections, suggesting an absence of industrial inputs in those areas. At a low-latitude site in southeast Hudson Bay, however, concentrations of mercury and lead were significantly higher in modern (1970s) shells of blue mussels.

Low concentrations of mercury, selenium and lead were found in samples of scallops from Labrador (Muir *et al.*, 1999a; 2000a). Cadmium and arsenic levels were much higher than the other elements, especially in gut samples. Muscle (meat) tissue from the scallops had relatively low levels of all five metals. Organic mercury was determined in a subset of the scallop muscle samples. All mercury in muscle was in the organic form, probably methylmercury.

Arsenic was the most prominent of the five metals determined in blue mussels from Nunavik and Labrador (Muir *et al.*, 1999a; 2000a). Concentrations ranged from 1.5 µg/g ww in samples from Makkovik to 2.3 µg/g ww in samples from Kuujjuaq. Arsenic has been previously found to be present at µg/g levels in mussels from Nunavik (Doidge *et al.*, 1993) but has not been routinely determined in marine or anadromous fish or shellfish from other areas of the Canadian Arctic. Mercury levels were low in mussels ranging from 0.01 to 0.03 µg/g ww and did not vary between locations (Figure 3.3.1). Organic mercury averaged 54% of total mercury. This organic mercury is probably all in the methylmercury form (Wagemann *et al.*, 1997). Cadmium levels ranged from 0.2 to 1.1 µg/g ww. Doidge *et al.* (1993) found similar, low µg/g ww levels of cadmium (0.5 µg/g ww) and mercury (0.03 µg/g ww) in blue mussels from six communities in the Hudson Bay, Hudson Strait and Ungava Bay area. There were no major differences in levels of metals between mussels collected in Nain and Makkovik and those from Nunavik.

As part of a food web study (Section 3.3.4.1), Atwell *et al.* (1998) measured total mercury in 12 invertebrates and particulate organic matter (POM) from Lancaster Sound region. Total mercury was not detected in the POM (< 0.02 µg/g dw) and ranged from 0.03–0.18 µg/g dw in the invertebrates. These levels are similar to those observed in blue mussels from Nunavik and Labrador (Muir *et al.*, 1999a; 2000a). There appeared to be no relationship between mercury concentration and trophic level within the invertebrates as a single group. The authors reported that no evidence of bioaccumulation of mercury with age in the clams (*Mya truncata*) was observed, despite the age range covering 42 years.

Fish

An overview of mercury in sea-run Arctic char is provided in Figure 3.3.2 which shows the ranges of concentrations for about 35 locations. In general, concentrations of mercury were < 0.1 µg/g ww in muscle at all these locations. Muir *et al.* (2000a) found mercury levels in sea-run char from eight communities in Labrador and the Ungava/Hudson Strait region of Nunavik were similar with means ranging from 0.032–0.040 µg/g ww (Annex Table 5). Statistical analysis of the mercury results (ANCOVA, Tukey's test of adjusted means) showed that there were no significant differences in mercury levels among the three Labrador sites; however, one location in Nunavik (Quaqtaq) had higher levels than all other sites (Figure 3.3.2). The Hg levels were slightly lower than reported by Riget *et al.* (2000) in sea-run char from Southwest Greenland. Muir *et al.* (1999a; 2000a) had

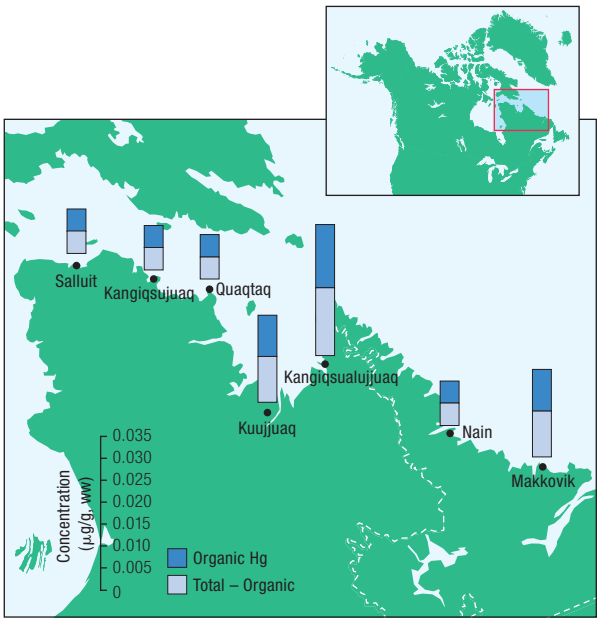

FIGURE 3.3.1

Total and organic mercury concentrations in blue mussels from Nunavik and Labrador (1998–1999) (Muir *et al.*, 2000a). Mercury levels are adjusted for age and length using ANCOVA.

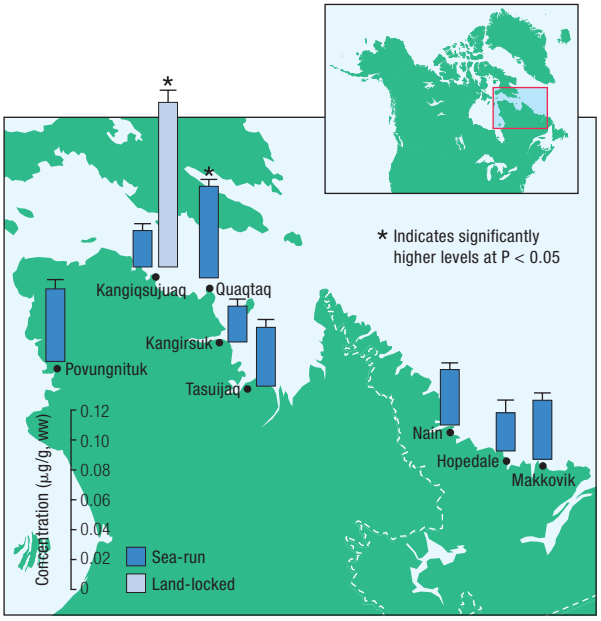

FIGURE 3.3.2

Mercury concentrations in sea-run Arctic char from Nunavik and Labrador (1998–1999) (Muir *et al.*, 2000a). Mercury levels are adjusted for age and length using ANCOVA.

previously reported undetectable levels of Cd and Pb in char muscle (< 0.001–< 0.006 µg/g) and these metals were again near or at detection limits. Mercury levels were influenced by age and size of the fish. About three-fold higher levels of mercury were found in land-locked char from Kangiqsujuaq than sea-run char. However, selenium, which may exert a protective effect for mercury exposure, was also much higher in the land-locked char. Another study also indicated that resident or land-locked populations of Arctic char and Atlantic salmon (*Salmo salar*) from Labrador displayed higher mean mercury values than their oceanic counterparts (Bruce and Spencer, 1979).

Arsenic and selenium were the most prominent of the five metals in sea-run Arctic char samples from three communities in Labrador (Muir *et al.*, 2000a) (Annex Table 5). There were no significant differences in concentrations of selenium or arsenic between the three Labrador locations. Comparing with results from 1998 (Muir *et al.*, 1999a), arsenic levels were significantly higher in sea-run char at two of the four locations in Nunavik compared to the three sites in Labrador. There has been little previous work on arsenic in fish from northern Québec or Labrador. Significantly lower levels of selenium were found in char from Tasuijaq (Nunavik) compared to Labrador while other locations had similar levels. Land-locked char (Kangiqsujuaq) had significantly higher levels of selenium than all sea-run char. Age and size of fish had no significant effect on selenium or arsenic levels in muscle.

As part of a food web study (Section 3.3.4.1), Atwell *et al.* (1998) measured total mercury in muscle tissue of two fish species, Arctic cod (*Boregadus saida*) and twohorn sculpin (*Icelus biocornis*) from the Lancaster Sound region. Concentrations of mercury in the Arctic cod [0.19 ± 0.3 µg/g dw, mean ± 1 SE (standard error)] and twohorn sculpin (0.19 ± 0.3 µg/g dw, mean ± 1 SE) are similar to those observed in Arctic char if concentrations are converted to wet weight.

Recently, data on metals have been produced for livers of Greenland shark (*Somniosus microceplalus*), the only shark species that routinely inhabits Arctic waters (Fisk, 2002a) (Annex Table 5). Little is known about these large fish but evidence suggests that they are long-lived (> 100 year) (see Hanson, 1963), generalist feeders with a potential to feed at a high trophic level in the food web (Fisk *et al.*, 2002a). Greenland shark samples (n = 24) were collected in Cumberland Sound in 1999 and 2000. Arsenic, cadmium, selenium and mercury concentrations varied little between sexes and were not related to length (linear regression, all p-values > 0.20). Arsenic and cadmium were found at the highest concentrations (9.8 ± 0.7 µg/g and 3.9 ± 0.4 µg/g, mean ± 1 SE, respectively) in the sharks followed by selenium (0.52 ± 0.03 µg/g), mercury (0.49 ± 0.06 µg/g) and lead (below the detection limit of 0.002 µg/g). Concentrations of total Hg in Greenland shark liver (0.49 ± 0.06 µg/g) were more than an order of magnitude greater than sea-run Arctic char muscle collected in the same region but orders of magnitude less than levels observed in marine mammal liver.

Butyltins in marine fish

Levels of tributyltin in Arctic char muscle ranged from < 0.01 to 0.85 ng/g ww (Muir *et al.*, 2000a). Tributyltin levels did not vary with age although weight was a significant covariate. Comparison of weight-adjusted means using Tukey's test showed that tributyltin levels in samples from Kangirsuk were significantly higher than other locations, while levels in Arctic char from Nain, Quaqtaq and Tasuijaq did not differ.

3.3.1.2 Organochlorines in invertebrates and fish

Zooplankton

Two studies have been recently carried out to examine OC concentrations in Arctic marine invertebrates and the factors that influence OC concentrations in these

lower trophic level organisms. As well, a large number of samples from single species were analyzed for OCs, particularly for zooplankton. In a number of studies these data were used in food web studies (Section 3.3.4.1). The most common OC compounds in pelagic zooplankton are the more water-soluble compounds, such as the HCH isomers and lower chlorinated PCB congeners. This reflects the smaller size, lower trophic level, and lack of biotransformation capacity generally found in zooplankton as compared to fish, mammals and birds.

Calanoid copepod samples were collected from Holman, NWT, and Barrow and Kaktovik, Alaska in 1999–2000 to examine spatial trends of OCs in the Beaufort Sea region (Hoekstra *et al.*, 2002b) (Annex Table 6). Calanoid copepods are dominant components, in both number and biomass, in high-latitude marine zooplankton communities and play an important role in polar food webs as their high lipid reservoirs and biomass provide higher trophic levels with a high-energy diet (Springer *et al.*, 1996). The ranking of OC group concentrations from highest to lowest were toxaphene ≥ ΣPCB > ΣHCH > ΣDDT > ΣCHL >ΣCBz. The higher abundance of toxaphene congeners relative to other OCs in zooplankton is consistent with OC concentrations in zooplankton collected previously in the Arctic Ocean offshore from Axel Heiberg Island in the Canadian Arctic (Bidleman *et al.*, 1989). The ranking by OC concentration was the same as that observed in samples of *Calanus hyperboreus*, a major component of the Beaufort Sea copepods, collected (n = 20) between April and July 1998 in northern Baffin Bay (Fisk *et al.*, 2001b). The α- and γ-HCH isomers and lower chlorinated PCB congeners were the most common OCs found in the calanoid copepods. The relative abundance of the OC groups and individual chemicals varied throughout the four-month season in northern Baffin Bay *C. hyperboreus*, as did lipid content (Fisk *et al.*, 2001b). Concentrations of ΣHCH, ΣCHL and ΣCBz increased over the sampling period, but no change in ΣPCB or ΣDDT was observed.

No significant differences were found for any ΣOC groups in calanoid copepods among the western Arctic sites (Holman, Kaktovik, and Barrow) when the effect of lipid and water content covariates was removed ($p > 0.05$ for all comparisons) (Hoekstra *et al.*, 2002b). Significant differences were observed, however, between the Alaskan sample sites and northern Baffin Bay/Rankin Inlet sites for toxaphene, ΣPCB, ΣDDT, ΣHCH and ΣCHL (Figure 3.3.3). HCB and HCH isomers in zooplankton and water samples from the Alaskan and western Canadian Arctic were relatively higher than the more easterly sampling locations. The abundance of HCB and HCH in Alaskan and western Canadian water and zooplankton reflects the long-range atmospheric transport of the chemicals and geographic proximity to areas of recent application in Asia (Bailey, 2001; Li, 1999). Total toxaphene levels in zooplankton were lower at Alaskan sites compared to the eastern Canadian Arctic. The concentrations of OC compounds in zooplankton samples, including *C. hyperboreus* collected in the late 1980s were in general agreement with this study (Bidleman *et al.*, 1989). This suggests that OC concentrations in the western Arctic water and zooplankton have remained constant over the past decade.

The relative abundance of hydrophobic chemicals such as ΣDDT and ΣCHL in copepod samples was greater than that observed in water (Hoekstra *et al.*, 2002b; Fisk *et al.*, 2001b), and reflects the greater potential for bioaccumulation from the abiotic environment into marine zooplankton near the base of the food chain than less hydrophobic OCs. Bioaccumulation factor (BAF) — octanol-water partition coefficient (K_{ow}) relationships observed for the Beaufort Sea and northern Baffin Bay calanoid copepods were linear and slightly greater than 1:1 for OCs with a log K_{ow} between 3–6, but curvilinear when hydrophobic OCs (log K_{ow} > 6) were included (Hoekstra *et al.*, 2002b; Fisk *et al.*, 2001b). These results suggest that hydrophobic OC (log K_{ow} 3.8–6.0) concentrations in zooplankton generally reflect water concentrations but dietary accumulation does increase levels above those predicted by simple partitioning from water and K_{ow}.

OC data for a number of other pelagic zooplankton have been recently generated in northern Baffin Bay (Fisk *et al.*, 2001a; Moisey *et al.*, 2001; Fisk *et al.*, 2002b)

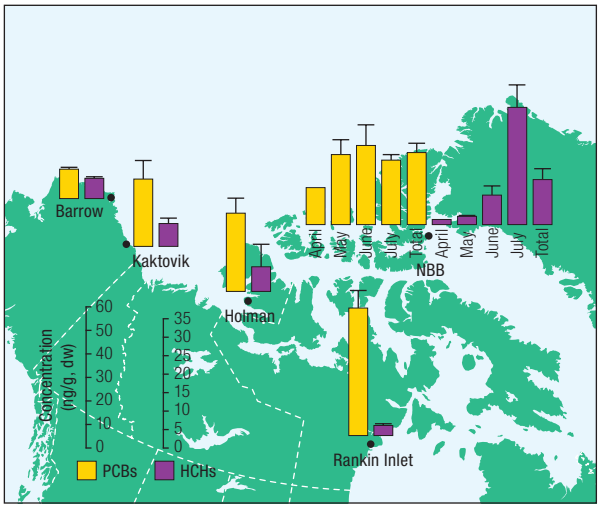

FIGURE 3.3.3

Mean ΣPCB and ΣHCH (± 1 SE) in zooplankton (ng/g dw) in Barrow and Kaktovik, Alaska; Holman, NWT; Rankin Inlet, Nunavut; and North Baffin Bay (NBB). Data from Hoekstra *et al.* (2001b), Fisk *et al.* (2001b) and Muir and Lockhart (1997).

(Figure 3.3.4) (Annex Table 6). Most of these samples were analyzed as part of food web studies. Seven species of zooplankton were collected in northern Baffin Bay and analyzed for OCs, including *Calanus hyperboreus* (herbaceous copepod); *Euchaeta glacialis* (omnivorous copepod); *Metridia longa* (omnivorous copepod); *Mysis occulata* (detritus feeding and predatory mysid); *Themisto libellula* (predatory amphipod); *Sagitta* sp. (predatory arrowworm); and *Pandalus* sp. (predatory shrimp). Lipid content varied between species, ranging from 2.1–7.0%. OC concentrations (lipid corrected) varied between species and appear to be related in part to a combination of trophic position (as determined by $\delta^{15}N$) and body size. The relative ranking of OC groups varies with the species, but in general ΣPCB had the highest concentrations and ΣCBz had the lowest.

Benthic marine invertebrates

Benthic invertebrates have a larger range of sizes, feeding ecology and ecological niches than pelagic zooplankton and therefore have a greater range of OC concentrations. Scavenging benthic invertebrates have the potential to have very high OC concentrations. PCBs are the OC group with the greatest concentrations in benthic invertebrates, with chlordane and DDT having high concentrations in certain species.

The blue mussel is commonly used throughout the world and in the Arctic to monitor OCs. PCBs and other organochlorines were present at very low levels in blue mussels from five locations in Nunavik in the Ungava Bay and Hudson Strait area and two locations in Labrador (Annex Table 6). PCBs were present at much higher concentrations (range of means 3.7–46 ng/g ww) in mussel tissues compared to other organochlorines. HCH isomers were the next most prominent contaminants with means ranging from 1.1–2.9 ng/g ww. The greater sorption of PCBs to sedimenting particles which are filtered by mussels, may account for the much higher amounts of PCBs compared to most other OCs. Doidge *et al.* (1993) found low ng/g ww levels of PCBs and OC pesticides in a survey of blue mussels from six communities in Nunavik. Levels of PCBs in samples from Kuujjuaq are much higher than those found in the same species at most other locations suggesting local contamination sources might be a factor.

A number of Arctic benthic invertebrates have been analyzed for OCs since the last CACAR assessment (Annex Table 6). As with other invertebrates, PCBs have the highest concentration in benthic invertebrates but the concentrations of the other OC groups vary with the feeding strategy of the invertebrates (Figure 3.3.4). Scavenging invertebrates, the amphipod *Anonyx nugax*

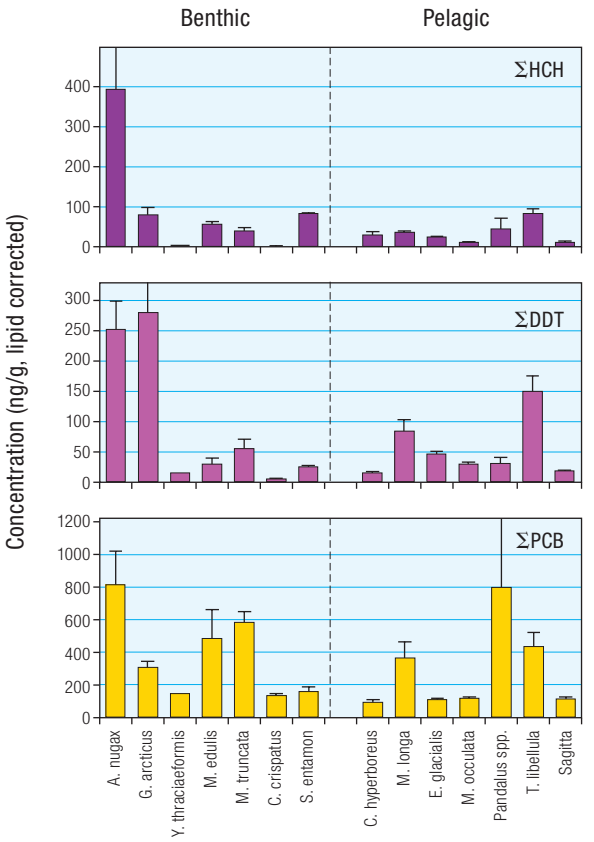

FIGURE 3.3.4

Concentrations (ng/g, lipid normalized) of ΣHCH, ΣDDT and ΣPCB in Arctic benthic and pelagic invertebrates. All samples were collected in 1998 in northern Baffin Bay except *M. truncata* (1993 in Iqaluit harbour), *M. edulis* (1998 at seven locations in northern Québec) and *S. entamon* (1998 in northern Alaska near Barrow). Results of *A. nugax* and *G. arcticus* include samples from Cumberland Sound. All data from Fisk *et al.* (2002b).

and large basketstar *G. arcticus*, have ΣHCH, ΣCHL and ΣDDT concentrations that are within 1/3 to 1/2 the ΣPCB concentrations. In filter-feeding and detritus feeding invertebrates the concentrations of other OC groups are much lower (approximately < 1/5) than PCB concentrations. The greatest concentrations of OCs in invertebrates are found in *A. nugax*, which have concentrations in the range of Arctic cod and the seabird dovekie (*Alle alle*) (Fisk *et al.*, 2001a). OC concentrations in filter- and detritus-feeding invertebrates are among the lowest found in any biota in the world.

Marine and anadromous fish

Low (ng/g ww) levels of organochlorines were detected in Arctic char muscle (+ skin) from two locations in Labrador and three locations in Nunavik (Figure 3.3.4.1) (Annex Table 6). PCBs were the most prominent contaminants averaging 11 ng/g ww in samples from

Makkovik and 31 ng/g ww in samples from Nain. PCB levels in char muscle samples from the three locations in Nunavik were intermediate in concentration, with means ranging from 14–21 ng/g ww. No significant differences in ΣPCB among locations were found. Lipid content, length and age were not significant covariates; however, ΣPCB levels were influenced by the sex of the fish, with males having significantly higher levels than females. This may be due to the timing of sampling near spawning time for the char. Females may have sequestered contaminants in eggs as fat is mobilized; this may lower concentrations observed in muscle.

The next most prominent groups of OCs in char muscle were the HCH and DDT groups which were both present at low ng/g levels. ΣDDT levels were higher at Nain than other locations but this was marginally significant only between Nain and Kangiqsujuaq. The char from Kangiqsujuaq were land-locked. In general, these levels of persistent organochlorines are similar or lower than reported in char muscle from other locations in the Canadian Arctic (Muir *et al.*, 1997).

Arctic or polar cod (*Boreogadus saida*) are a key link in Arctic marine food webs, providing a link between invertebrates and marine mammals and seabirds. ΣPCB was the dominant OC group followed by ΣCHL in Arctic cod collected in northern Baffin Bay in 1998 (Fisk, 2002b) (Annex Table 6). Levels were similar to those observed in sea-run Arctic char.

OCs were measured in 17 Greenland sharks (*Somniosus microcephalus*) and four turbot (*Reinhardtius hippoglossoides*) collected in the Davis Strait and Cumberland Sound region in 1997 and 1999 (Fisk *et al.*, 2002a) (Annex Table 6). Concentrations of OCs in the Greenland shark were in the range of other top Arctic marine predators, such as the polar bear or glaucous gulls. Concentrations (lipid basis) of OCs in the Greenland sharks in this study were not related to fork length, sex, $\delta^{13}C$ or $\delta^{15}N$. Concentrations of OCs (lipid basis) in Greenland sharks were 10–100 and 3–10 times higher than those observed in turbot and ringed seals, respectively, suggesting a higher trophic level for the sharks than implied by $\delta^{15}N$ values. Concentrations of ΣDDT in the study samples of Greenland sharks are among the highest in Canadian Arctic biota, which may be related to the low metabolism and long life span of these sharks. Reported concentrations of ΣPCB and ΣDDT in turbot collected in the Davis Strait in 1992 were 5 and 10 times higher, respectively, but concentrations of ΣCHL and ΣHCH were very similar to those reported for turbot in this study (see Berg *et al.*, 1997). The higher PCB and DDT in turbot reported in Berg *et al.* (1997) could be due to the larger size and the deeper water habits of these turbot.

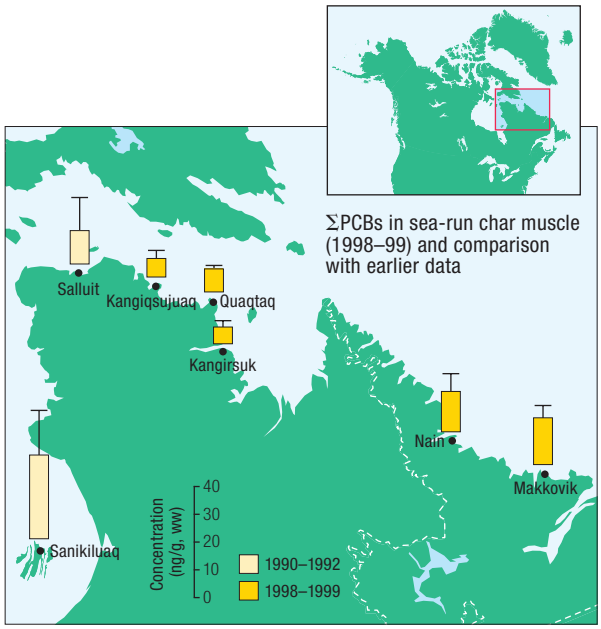

FIGURE 3.3.5

ΣPCB levels in sea-run char of northern Labrador, Nunavik and Sanikiluaq, Nunavut (Muir *et al.*, 2000a).

3.3.2 Spatial trends of organochlorines and metals in seabirds

There are approximately 50 species of Arctic seabirds (de March *et al.*, 1998) that have a variety of feeding and migration habits, and subsequently OC and metal concentrations can vary considerably among species (Fisk *et al.*, 2001c; Braune, 2000; Braune *et al.*, 2001a; 2002). Species, such as the glaucous gull (*Larus hyperboreus*), that migrate to more contaminated regions and/or that scavenge, particularly on dead marine mammals, have the highest OC and mercury concentrations (Buckman *et al.*, 2002).

There has been a considerable amount of work on OCs and metals in Arctic seabirds since the first CACAR assessment. Continued monitoring of OCs in Canadian Arctic seabird eggs, from 1975 to 1998 and beyond, has proved a strong data set for examining temporal trends among different species (see Section 4.2). OC and metal data have also been recently generated for a range of species from northern Baffin Bay, and new data on PCDD/Fs, toxaphene and new OCs have also been generated for Canadian seabirds (Braune, 2001a).

3.3.2.1 Metals in seabirds

Seabirds generally exhibit higher mercury concentrations than terrestrial birds because of the higher mercury burdens encountered in marine ecosystems (Thompson, 1996). Several studies (see Thompson, 1996) have shown that the predominant form of mercury found in seabirds

is inorganic suggesting that biotransformation of ingested methylmercury is an important mechanism by which long-lived and slow-moulting seabirds avoid the toxic effects of accumulating large quantities of methylmercury. As well, the apparent ameliorating effect of selenium on the toxicity of mercury and vice versa has been well documented (see Thompson, 1996, p. 342; Heinz, 1996, p. 448, for references). Seabirds are also known to accumulate high levels of cadmium and appear to be able to tolerate higher levels than other birds without effect probably because of some adaption to the naturally elevated levels of cadmium in seawater (Furness, 1996). Another reason that seabirds may have a higher tolerance to cadmium is that exposure to cadmium may induce production of metallothionein, a low-molecular-weight protein to which cadmium can be bound rendering it less toxic. Lead levels have generally declined in the Arctic due mainly to reduced emissions from industry and bans on the use of leaded gasoline. However, the historical use of lead shot for hunting has left considerable quantities of lead pellets on the ground or at the bottom in coastal, estuarine and wetland areas where birds may ingest them. This is more of a concern for diving species such as sea ducks which feed on the bottom in coastal ponds and wetlands (Franson *et al.*, 1995; Flint *et al.*, 1997; Flint & Herzog, 1999).

Long-tailed duck

Oldsquaw or long-tailed duck (*Clangula hyemalis*) nest in greater numbers in the high Arctic than any other duck (Bellrose, 1980), but North American populations are declining in the west (CWS *et al.*, 1997). Long-tailed ducks nesting in the eastern Canadian Arctic likely winter along the Atlantic coast whereas western Arctic birds winter in the Bering Sea and along the Pacific coast of Alaska and British Columbia (Bellrose, 1980). Considerable numbers also winter on the Great Lakes. It is unknown where the dividing line occurs between the two breeding populations.

Long-tailed ducks are harvested by Aboriginal people and sport hunters alike, but numbers taken are unlikely to be the cause of the decline (CWS *et al.*, 1997). Metal residue data for breast muscle from long-tailed ducks collected from across the Canadian Arctic during 1991–94 suggest that western Arctic populations generally have higher levels of cadmium and selenium than eastern populations (Braune *et al.*, 1999b). Heavy metal contamination from their diet of benthic organisms may possibly be a significant threat to long-tailed ducks. High lead levels, resulting from the ducks ingesting spent shot on the breeding grounds, are suspected of impairing reproductive success of long-tailed ducks nesting in the Alaskan Yukon-Kuskokwim Delta (Flint *et al.*, 1997). It is not known whether or not long-tailed ducks are more exposed to metal contamination on the Arctic breeding grounds or on their overwintering grounds.

Archived liver, kidney and bone samples collected during 1991–94 from 59 long-tailed ducks from the eastern Canadian Arctic (east of 95°W) and 42 long-tailed ducks from the western Canadian Arctic were analyzed for mercury, selenium and copper in liver, cadmium in kidney, and lead in bone (Braune, 2001b). Male long-tailed ducks generally contained higher residue concentrations than females except for lead in bone of the western birds (Figure 3.3.6). Both cadmium in kidney and selenium in liver were higher in western birds than eastern birds with the reverse being true for lead in bone. These data follow the same geographic pattern found by Braune *et al.* (1999b) for cadmium and selenium in breast muscle of long-tailed duck.

Seabirds of northern Baffin Bay

A recent study has measured metals and methylmercury (MeHg, muscle only) in the liver and muscle of eight species of Arctic seabirds to examine inter-species differences in seabirds and seasonal trends in thick-billed-murres (*Uria lomvia*) (Buckman *et al.*, 2002) (Annex Table 7). These samples were used for a larger study examining food web dynamics of metals discussed below in Section 3.3.4.1. All samples were collected in the high Arctic between Ellesmere Island and Greenland in 1998 and 1999, although data have only been generated in late 2001 and early 2002. As of this report all data for this project have not been generated.

Liver concentrations were greater than muscle across all species for all metals except lead. Concentrations did not vary between gender for arsenic, cadmium, lead and selenium. Concentrations of total mercury did vary between gender when liver and muscle samples were combined

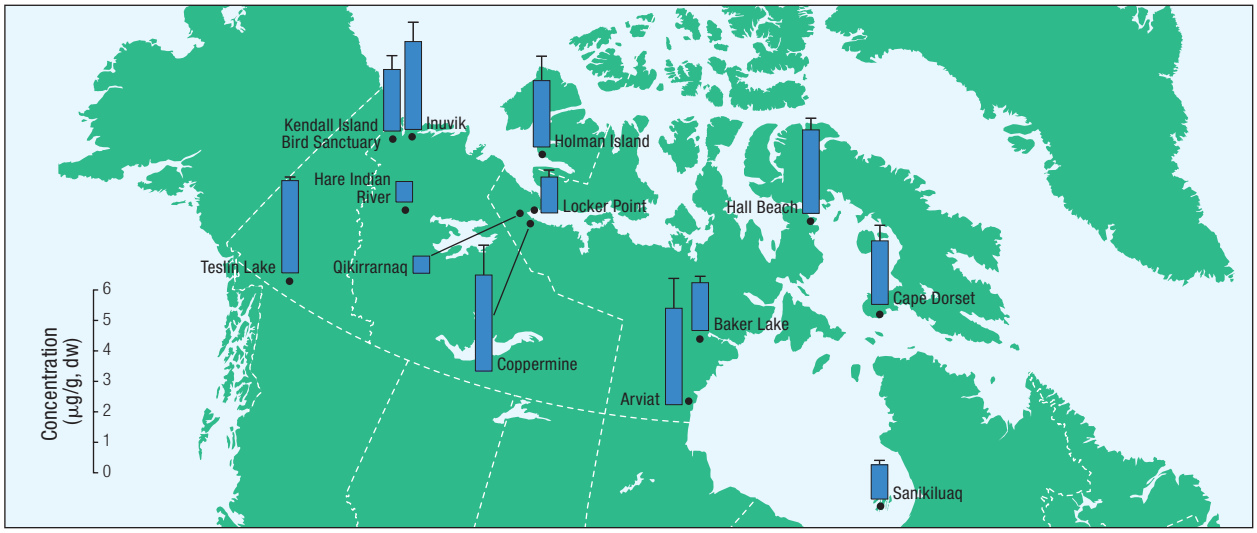

FIGURE 3.3.6

Mercury in the liver of long-tailed ducks collected between 1991 and 1994 (Braune, 2001b).

but were marginally different when only muscle data was compared. Methylmercury concentrations did not vary between sexes for any of the seabird species.

An initial examination of metal concentrations in the seabird species has indicated that age, migration and trophic level are important parameters to consider when interpreting seabird metal concentrations, although this varies with the metal. Metal concentrations in the muscle of the seabirds indicated some differences between species, notably higher levels of cadmium and selenium in northern fulmars (*Fulmaris glacialis*) and higher levels of selenium in black-legged kittiwakes (*Rissa tridactyla*) (Figure 3.3.7). High levels of cadmium in northern fulmars may be an age-related phenomenon. Northern fulmars are a very long-lived bird and Cd-metallothionein binding has been shown to increase with age (Furness, 1996; Hatch and Nettleship, 1998). High selenium concentrations in black-legged kittiwake may be related to accumulation in more southern locations. These birds migrate long distances to contaminated regions in the St. Lawrence estuary and the eastern seaboard of North America. Dovekie had significantly lower As and Hg concentrations than all other seabirds. The low mercury levels are likely due to the low trophic position of the dovekie. Methylmercury concentrations were highest in glaucous gull and lowest in dovekie (Figure 3.3.8), and were highly correlated to trophic position (see Section 3.4.1). The current lack of metal data for glaucous gull and ivory gull livers has precluded species comparison for this tissue.

There was a general trend for metal concentrations to decrease in thick-billed murre from May to September. The strongest decline was observed for As (Figure 3.3.9) but weaker declines were also observed for cadmium, selenium and mercury. Caution should be exercised with this initial data, however, because of the small sample sizes. As well, many of the August and September birds were young of the year and these birds would likely have lower concentrations of most metals. Seasonal changes in metal concentrations could result from changes in diet. Reproduction and migration may also affect the results. Further analysis and study need to be done when all data are available.

Canadian seabird eggs

As part of a continuing temporal study of contaminants in seabirds, eggs of northern fulmars, glaucous gulls, black-legged kittiwakes, thick-billed murres and black guillemots were collected from nests on Prince Leopold Island in Lancaster Sound, Nunavut, Canada in 1998 and analyzed for mercury and selenium (Braune *et al.*, 2001a; Donaldson *et al.*, 2002; in preparation) (Annex Table 7). Selenium concentrations were significantly different among species, with glaucous gull and fulmar eggs being significantly higher than thick-billed murre, kittiwake and guillemot eggs. Concentrations of mercury in glaucous gull eggs were significantly higher than concentrations measured in all other species because of their higher trophic level. Kittiwake eggs contained significantly lower total mercury concentrations compared to all other species. Total mercury was not statistically distinct between thick-billed murres and northern fulmars. Mercury has a tendency to accumulate at higher latitudes (Barrie *et al.*, 1997); therefore, species spending more time at higher latitudes may be expected to exhibit higher concentrations. This was reflected by data on black guillemots, which are most likely to remain resident in the Arctic, and had higher mercury concentrations.

Black-legged kittiwakes migrate south along the eastern seaboard as far south as Florida and they exhibited the lowest mercury concentrations. These results contrast somewhat with results observed in the tissues of adult seabirds from northern Baffin Bay. Levels of metals in eggs may not reflect what is observed in adult tissues.

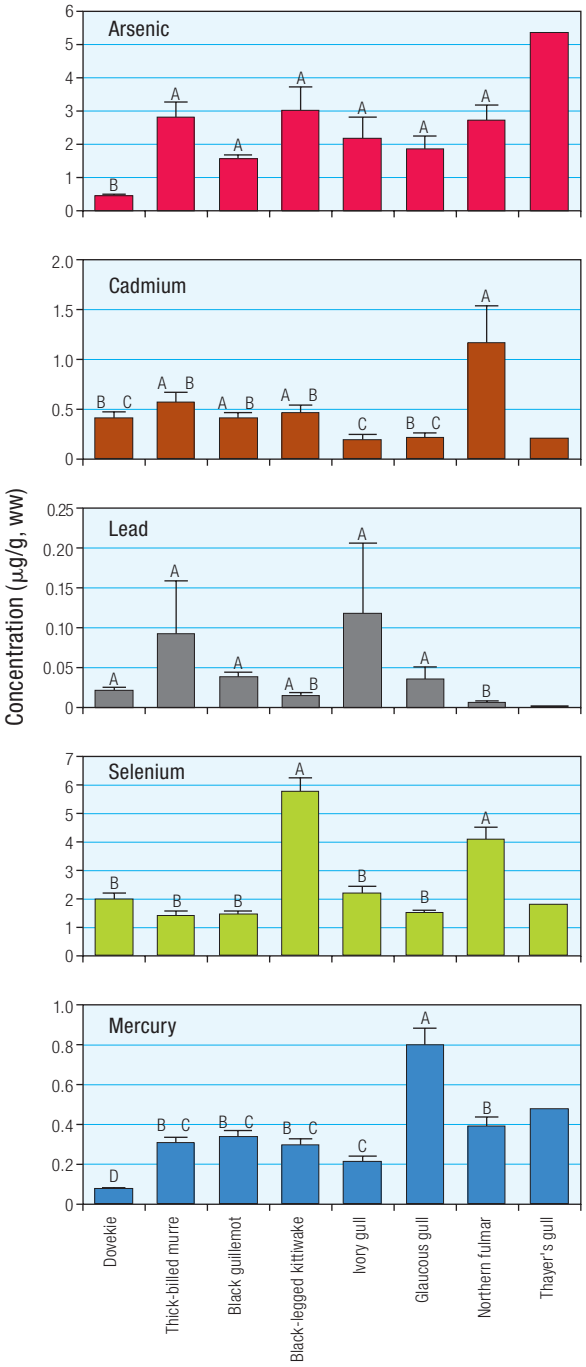

FIGURE 3.3.7

Metal concentrations (bars are mean ± SE, outliers removed) in the muscle of seabirds collected in northern Baffin Bay in May or June 1998 (Fisk, 2002c). Concentrations that were not statistically different ($p > 0.05$) have the same letter and were tested using a general linear model (GLM) and log-transformed data. Statistics did not include Thayer's gull because n = 1 for this seabird.

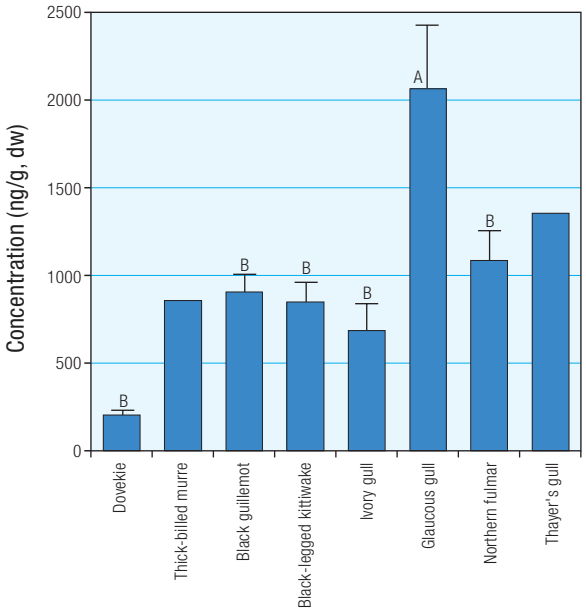

FIGURE 3.3.8

Methylmercury concentrations (bars are mean ± SE, outliers removed) in the muscle of seabirds collected in northern Baffin Bay in May or June 1998 (Fisk, 2002c). Concentrations that were not statistically different ($p > 0.05$) have the same letter and were tested using GLM. Statistics did not include Thayer's gull or thick-billed murre because n = 1 for these seabirds.

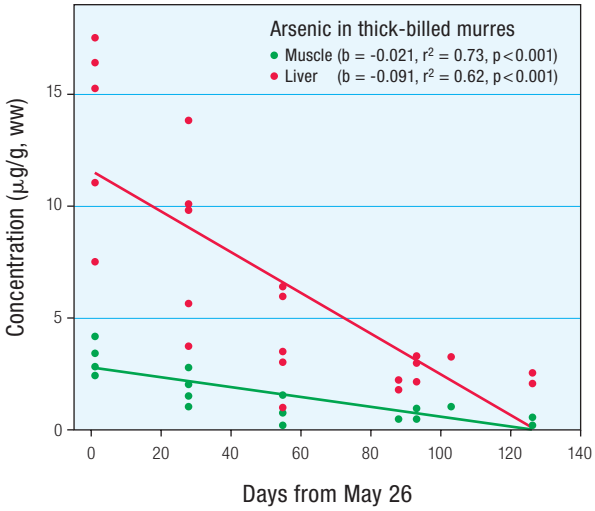

FIGURE 3.3.9

Seasonal trends of arsenic in the muscle (green circles) and liver (red circles) of thick-billed murres collected in northern Baffin Bay (Fisk, 2002c).

Seabirds are able to tolerate exceptionally high levels of mercury, presumably through rapid biotransformation of methylmercury to inorganic mercury. This is thought to be a survival mechanism allowing the birds to cope with the naturally high mercury levels encountered in marine environments (Thompson and Furness, 1989). Seabirds have been shown to tolerate tissue levels much greater than the 30 mg/kg ww which is considered to be lethal to birds of prey (Honda *et al.*, 1990; Muirhead and Furness, 1988). The greatest mercury concentration recorded in this study was much lower than that critical concentration, so clearly mercury is of only limited concern for these seabirds.

3.3.2.2 Organochlorines in seabirds

PCBs are the most common OCs measured in Arctic seabirds, although the relative amount is related to the trophic position of the seabird (Borgå *et al.*, 2001). Higher trophic level seabirds generally have a larger proportion of PCBs as a percentage of total OCs. This is due to greater biomagnification potential of PCBs and the ability of seabirds to metabolize other OCs, such as α-HCH (Moisey *et al.*, 2001) and chlordane-related compounds (Fisk *et al.*, 2001c). Other recalcitrant and biomagnifying OCs also become more prevalent, such as *p,p'*-DDE and oxychlordane (Borgå *et al.*, 2001; Fisk *et al.*, 2001c), although the relative proportions can vary dramatically among species, even within the same family (Fisk *et al.*, 2001c).

Seabirds of northern Baffin Bay

OC concentrations and dynamics were measured in the liver and fat of seven Arctic seabird species collected in northern Baffin Bay in 1998 (Fisk *et al.*, 2001c; Buckman *et al.*, 2002) (Annex Table 8). These samples were used for larger studies examining food web dynamics of OCs discussed below in Section 3.3.4. ΣPCB was the most predominant OC group observed in all seabird species and, along with most of the other OC groups, attained the highest concentrations in the gulls (glaucous and ivory gulls and black-legged kittiwakes) and northern fulmar (Figure 3.3.10). Glaucous gulls, ivory gulls and northern fulmars scavenge, and glaucous gulls prey on other seabirds, which accounts for high OC concentrations. Migration to more contaminated southern locations may also account for levels in the glaucous gull and black-legged kittiwake. Black-legged kittiwakes, a non-scavenging species, had higher concentrations than the alcids (guillemot, thick-billed murre and dovekie), because of their migration to southern habitats. The kittiwakes were found to be feeding at a lower trophic level than black guillemot in northern Baffin Bay (Hobson *et al.*, 2002, in press), and since the black guillemot does not undergo a major migration, the difference between the species highlights the impact of exposure to OCs in non-Arctic habitats by migrating seabirds. ΣDDT and ΣCHL were the next most common OC contaminant

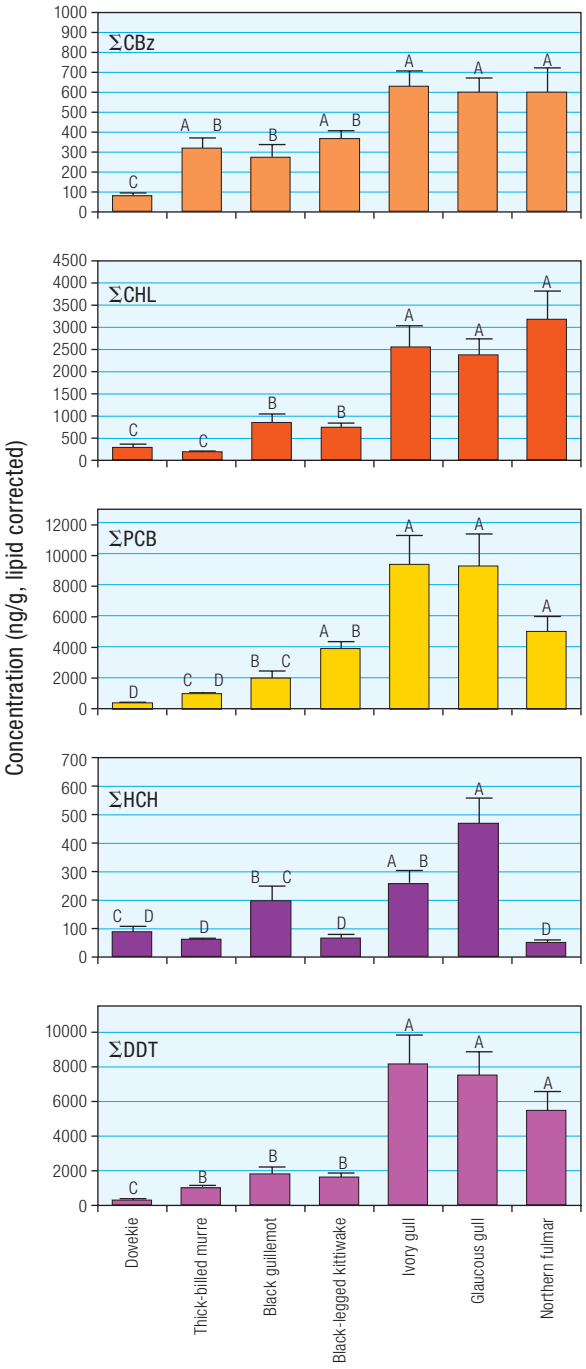

FIGURE 3.3.10

Lipid corrected concentrations (mean ± SE, ng/g) of ΣCBz, ΣHCH, ΣCHL, ΣDDT and ΣPCB in liver tissue of North Water (NOW) Polynya seabirds (Buckman *et al.*, 2002). Male and female did not significantly differ ($p < 0.05$), except for ΣDDT, and were combined. Bars with the same letter do not differ significantly ($p > 0.05$).

groups observed in the seabirds followed by ΣCBz and finally ΣHCH. The exception to this ranking is for thick-billed murres where ΣCHL was found at much lower concentrations than in other seabirds. Thick-billed murres have been shown to have an exceptional ability to biotransform chlordane components (Fisk *et al.*, 2001c). PCB 153 had the highest concentration of any PCB congener in all seabirds.

The relative proportion of ΣPCB compared with the other groups of OCs remained similar between liver and fat although it varied with species. Significant differences in concentrations between fat and liver, however, were found for ΣCBz, ΣHCH, and ΣPCB but not for ΣCHL and ΣDDT. This is difficult to explain because OC groups are generally correlated, especially for biomagnifying compounds such as DDT and PCB. The differences are likely due to a combination of migration, diet shifts and reproduction. Lipid-corrected concentrations of OC components did not significantly vary between sex for any species, except ΣDDT in liver tissue where concentrations in females were found to be lower than in males. This is likely due to recent changes in diet but it is curious that this difference was not observed for other OC groups.

Canadian seabird eggs

As part of a continuing temporal study of contaminants in seabird eggs (see Section 4.2), thick-billed murre, northern fulmar and black-legged kittiwake eggs were collected on Prince Leopold Island in Lancaster Sound in 1998 and analyzed for OCs (Braune *et al.*, 2001a) (Annex Table 8). Concentrations and the relative proportions of OC groups were in general agreement to those observed in tissue of adult birds of the same species collected in northern Baffin Bay (Fisk *et al.*, 2001c; Buckman *et al.*, 2002). Lipid content in the eggs of the three species was similar, allowing comparisons of wet weight concentration data. The predominant OC in all three species was ΣPCB. This was similar in black-legged kittiwakes and northern fulmars (0.28 ± 0.02 μg/g and 0.27 ± 0.02 μg/g ww, respectively) and nearly double that observed in thick-billed murres (0.13 ± 0.01 μg/g ww). This reflects the higher trophic level of northern fulmars and black-legged kittiwakes and the migration to more contaminated habitats by the kittiwakes. ΣDDT was highest in the northern fulmar (0.21 ± 0.02 μg/g ww), followed by the thick-billed murre (0.10 ± 0.01 μg/g ww) and the black-legged kittiwake (0.06 ± 0.01 μg/g ww), and similar trends were observed for ΣCHL.

Recently, compounds, such as dioxins, furans, coplanar PCBs and toxaphene were analyzed in thick-billed murres, northern fulmars and black-legged kittiwakes collected on Prince Leopold Island in Lancaster Sound (Braune, 2001a). These compounds had not been measured previously in Canadian Arctic seabirds. Samples included livers from 1975 and 1993 and egg samples from 1993. This pilot study was initiated to determine if temporal changes have occurred (see Section 4.2). PCDDs, PCDFs and non-*ortho* PCBs were found in all the Arctic seabird samples analyzed in this study. Concentrations of HxCDDs, PnCDFs and HxCDFs, in particular, found in Arctic seabird livers exceeded levels reported for marine mammals in the Canadian Arctic (de March *et al.*, 1998) by several orders of magnitude. Ringed seals, polar bears and walrus in the Canadian Arctic tend to contain relatively high TCDD and low PnCDD and PnCDF levels (de March *et al.*, 1998) whereas Arctic seabirds show the opposite pattern. Concentrations of PCDD/F homologs in the Arctic seabirds are in the range reported for seabirds from temperate North America and Europe (Hebert *et al.*, 1994; Van den Berg *et al.*, 1987). Toxaphene was detected in every seabird sample analyzed except for the pool of kittiwake livers from 1975 and was one to two orders of magnitude lower than levels reported for marine mammals from the Canadian Arctic (Muir *et al.*, 1999b).

3.3.2.3 New chemicals in seabirds

A number of new chemicals were also analyzed as part of the dioxin, furan and toxaphene analysis in Arctic seabirds (Section 3.3.2.2) (Braune, 2001a; 2002). Results of chemical analyses on livers (1975 and 1993) and eggs (1993) from thick-billed murres, northern fulmars and black-legged kittiwakes collected on Prince Leopold Island in Lancaster Sound are discussed in more detail in Section 4.2.

Total polybrominated diphenyl ethers (PBDE) were present at low ng/g levels in most of the samples analyzed (Braune, 2002). The highest estimated PBDE levels (3 ng/g ww; or about 60–70 ng/g lw) were detected in the 1993 egg and liver samples of kittiwakes (1993 murre and fulmar samples also contained ng/g levels). PBDE-47 was the major PBDE in all the samples. Although the data suggest that exposure to PBDEs has increased from 1975 to 1993, the highest level reported is an order of magnitude lower than levels reported for Swedish black guillemots (Sellström *et al.*, 1993), but is still higher than levels reported for Canadian marine mammals (Stern and Ikonomou 2000; 2001).

Halogenated dimethyl bipyrroles (HDBPs), which are naturally occurring, mixed bromine/chlorine, persistent organic compounds, have been shown to bioaccumulate in seabird eggs (Tittlemier *et al.*, 1999). Halogenated

dimethyl bipyrroles were detected in all of the 1993 samples and only one of the liver samples from 1975. The major contaminant found was 1,1′-dimethyl-3,3′, 4,4′-tetrabromo-5,5′-dichloro-2,2′-bipyrrole (DBP-Br_4Cl_2). The highest estimated level (a total of 5 ng/g ww) was found in the kittiwake egg sample from 1993. Trace amounts (< 2 ng/g ww) of chlorinated terphenyls, hexabromo biphenyls and TCP-methane were also detected in most of the Arctic seabird samples. No chlorinated diphenyl ethers, TCP-methanol, or chlorinated naphthalenes were detected in any of these samples (detection limit ~2 ng/g ww).

In a separate study, Tittlemier *et al.* (2002) examined levels of halogenated dimethyl bipyrroles in four seabird species (dovekie, black guillemot, black-legged kittiwakes and glaucous gulls) collected in northern Baffin Bay. This is significant because it represents the first time a naturally produced halogenated compound has been measured in an Arctic species. This was part of a larger study on the food web dynamics of these chemicals in marine food webs (Section 3.3.4.1). HDBPs were detected in all of the samples studied from the North Water (NOW) Polynya. Levels of DBP-Br_4Cl_2 in the seabirds were generally similar to those recorded in seabird samples from Atlantic Canada (Tittlemier *et al.*, 1999a). A comparison cannot be made for the other congeners measured since this study contains the first reported concentration data for these congeners. Dovekies provide the single clear exception were DBP-Br_4Cl_2 concentrations (1.76 ppb) were approximately ten times lower than those of the Atlantic puffin (*Fratercula artica*; 20 ppb), a species that has a similar winter habitat but a more piscivorous diet. The difference in concentrations is likely driven by the dovekies feeding at a lower trophic position just before their collection in early summer (Fisk *et al.*, 2001c). In most of the bird species DBP-Br_4Cl_2 was the predominant congener. The exception occurred with black guillemots, where DBP-Br_6 was the most abundant congener, possibly due to black guillemots feeding on benthic organisms during part of the year (Gaston and Jones, 1998).

Chiral pollutants exist in two forms as optical isomers called enantiomers. Enantiomers have identical physical-chemical properties and abiotic degradation rates, but can have different rates of biotransformation (Buser and Müller, 1992). The chemical manufacturing process results in a mixture containing approximately 50% of each chiral compound, termed racemic. Selective biotransformation of one chiral component over another can occur and result in an enantiomeric enrichment (Buser and Müller, 1992). This selective enrichment originates from one of the enantiomers being more easily biotransformed. This resulting selective accumulation of a single enantiomer can provide information on fate and dynamics of the chemical and may have significant toxicological ramifications. It has been proposed that comparison of enantiomeric ratios (ERs) may provide information on the biotransformation capacity of a species and the trophic transfer of contaminants in a food web (Wiberg *et al.*, 2000).

Enantiomeric fractions (EFs) of chiral chlordane components were examined in the liver and fat of seven Arctic seabird species collected in northern Baffin Bay in 1998 (Fisk *et al.*, 2001c). Enantiomeric fractions (EFs) of chiral components failed to predict concentration or trophic level, but did identify biotransformation differences between species and chlordane components. The relative proportions of chlordane components in seabirds were related to taxonomy and the magnitude of EF values; northern fulmars and gulls (black-legged kittiwake, ivory gull and glaucous gull) had a great percentage and higher EFs of oxychlordane than the alcids (dovekie and black guillemot). The exception was the thick-billed murre, an alcid, where oxychlordane made up a significant percentage of its chlordane. Thick-billed murres appear to have a greater capacity to metabolize and eliminate chlordane, based on high proportions of oxychlordane and the highest EFs for oxychlordane and heptachlor epoxide.

3.3.3 Spatial trends of contaminants in marine mammals

There have been more studies on levels of OCs in marine mammals than any other group of organisms from the Arctic, both historically and recently. The amount of data produced for metals in marine mammals is much less but is still a significant contribution. The focus on marine mammals stems from their importance in the culture and diet of Inuit, but also because levels of persistent OCs, mercury and cadmium, in general, are elevated in marine mammal tissues. As well, many marine mammals are hunted through the Canadian Arctic making the collection of samples cost efficient and relatively easy.

All relevant data on OCs and heavy metals in marine mammals that have become available since the last CACAR (Muir *et al.*, 1997) are reviewed herein. It must be emphasized that the spatial trends of persistent OCs discussed in the following sections are often qualitative because they are based on evaluation of means and ranges of concentrations from different studies. In the case of the sums of groups of compounds, such as PCBs and chlordanes, some laboratories have included more congeners or components than others in ΣPCB and

ΣCHL results. Rigorous comparisons among locations also require information on age, sex, blubber thickness, nutritional status, collection season, and reproductive status of the animals, all of which can have an important influence on contaminant concentrations. This information has been collected for many locations, but has only been used qualitatively in this assessment of spatial trends between studies.

3.3.3.1 Levels and spatial trends of metals in marine mammals

Ringed seals

Ringed seals (*Phoca hispida*) are the most abundant and widely distributed resident Arctic pinniped. Their diet consists of fish, mainly schooling gadids, and crustaceans (amphipods, mysids, and euphausids). Ringed seals have a broad circumpolar distribution and prefer annual, land-fast ice, but are also found near multiyear ice. No clear-cut boundaries are known to separate ringed seal stocks in marine waters. Adults are believed to be relatively sedentary, but sub-adults sometimes disperse over long distances (Reeves, 1998). Ringed seals are a key component of the diet of Inuit.

Studies of mercury and other heavy metals in ringed seals from Labrador, northern Québec (Nunavik) and north Baffin Bay, combined with results from an investigation of temporal trends of these metals in Nunavut (see Section 4.3.3) permit an assessment of spatial trends to be made. Samples of ringed seal liver, kidney and muscle were collected from nine communities in the central and eastern Arctic from 1998–2000 as part of the spring/summer hunt to determine mercury, cadmium, arsenic, selenium and lead concentrations (Annex Table 9). An extremely wide range of concentrations of mercury and cadmium was observed. This reflects the wide range in age of the seals (< 1 to 36 years). For the data set as a whole, mercury and selenium were significantly correlated with age. Cadmium, arsenic and lead concentrations were not correlated with age. Ages of seals from Arviat and Grise Fiord were generally older than those from all other sites making comparison among sites difficult in the case of mercury and selenium. To examine spatial trends, a subset of similar age range was selected. Mercury concentrations were adjusted for age using analysis of covariance with data for samples ranging from 2–15 years of age. This subset represented about 70% of the samples. After adjustment for age (adjusted to a mean age 7.7 years) there were only a few significant differences (SD) among locations for mercury (Figure 3.3.11). Concentrations of mercury were significantly lower in seal livers from Resolute than in those from Ungava Bay and Hudson Strait. For kidney, mercury levels were significantly lower in Hudson Strait and Labrador than at Pond Inlet or Resolute.

There was no significant effect of age or interaction of age with location for the subset of animals ranging from 2–15 years for cadmium concentrations in seal liver; therefore, there was no need to adjust for age. Cadmium concentrations had a different spatial trend compared to mercury with significantly higher levels in samples from Baffin Island (Pond Inlet and Pangnirtung) and lowest levels in samples from Labrador (Figure 3.3.11).

Arsenic levels in seal livers also showed significant spatial trends that were not affected by age. Lowest levels were found in samples from Ungava Bay, Labrador and Arviat

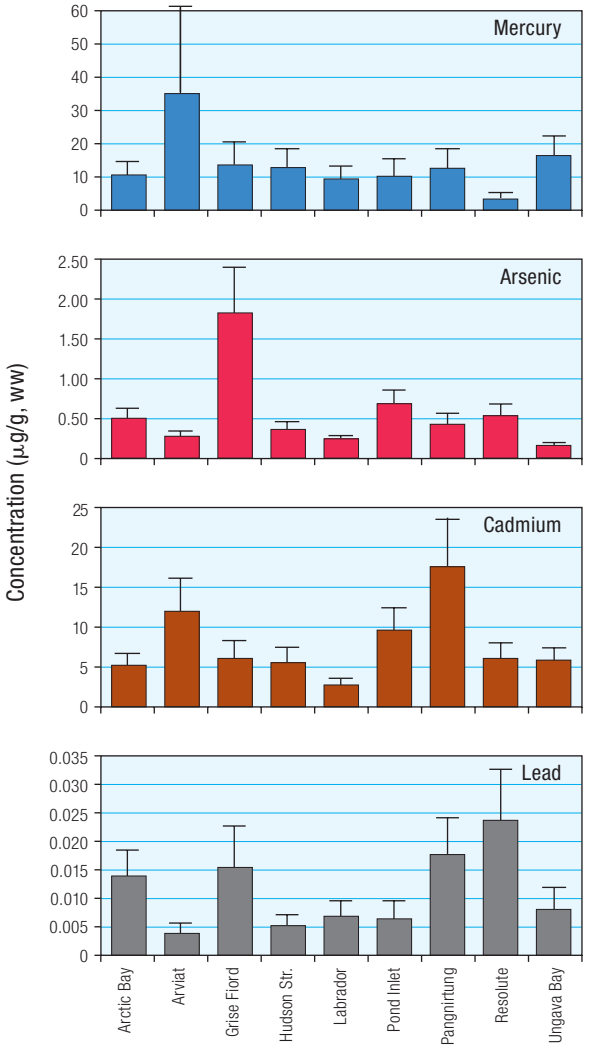

FIGURE 3.3.11

Geometric mean concentrations of mercury, cadmium, arsenic and lead in ringed seal liver from nine locations in the eastern/central Canadian Arctic. Results are based on samples from seals aged 2–15 years. Mercury concentrations are adjusted for age.

compared to samples from high Arctic sites (Grise Fiord, Pond Inlet, Pangnirtung and Resolute) (Figure 3.3.11).

Lead concentrations in seal liver were very low (sub-µg/g ww). Non-detection levels (representing about 10% of samples) were replaced by half the detection limit to compare among locations. Higher lead concentrations were found in samples from Arctic Bay, Grise Fiord, Pangnirtung and Resolute compared to Hudson Bay, Hudson Strait and Labrador (Figure 3.3.11).

Heavy metals were also determined in ringed seal muscle in samples from Nunavik and Labrador (Muir *et al.*, 1999a). All metals were present at much lower concentrations in muscle than in kidney or liver. Concentrations of mercury in muscle were about 20 times lower than those in liver.

Organic mercury levels were determined in muscle, liver and kidney of 20 ringed seals from sites in Nunavik for which total mercury was determined (Muir *et al.*, 1999a). The results show that organic mercury levels increased with age from about 80% in animals from 0–2 years to about 100% in adult animals (Muir *et al.*, 2000a). Percent organic mercury declined with age in seal liver and kidney representing < 5% of total mercury in the oldest animals. Based on results of Wagemann *et al.* (1997) the organic mercury is likely to be entirely in the form of methylmercury.

Metallothionein (MT) concentrations in ringed seals ranged from about 50–600 µg/g dw in liver and from 50–1,200 µg/g in kidney (Muir *et al.*, 2000a). MT levels were strongly correlated with cadmium concentrations in liver and also in kidney (for cadmium concentrations < 150 µg/g). MT is a low relative atomic mass (6000–6500) cytoplasmic protein that occurs naturally at background levels in most organisms. It is characterized by a high content of zinc, copper and cadmium (depending on availability), and an amino acid composition characterized by a high cysteinyl residue content. The induction of MT, primarily in response to copper, zinc and cadmium exposure, is a protective cellular response and MT therefore is a potential biomarker for cadmium toxicity. When this protective capacity is exceeded, particularly for cadmium, toxicity could occur. MT levels in seal liver were within the range found in narwhal liver by Wagemann *et al.* (1993) but lower than the levels found in seabird liver by Sheuhammer and Cherian (1991).

Walrus

Mercury levels in walrus meat (muscle) were relatively low compared with liver and kidney (Muir *et al.*, 2000a) (Annex Table 9). About 75% of mercury in muscle, 5% in liver and 20% in kidney was in the form of organic mercury, probably methylmercury (Wagemann *et al.*,

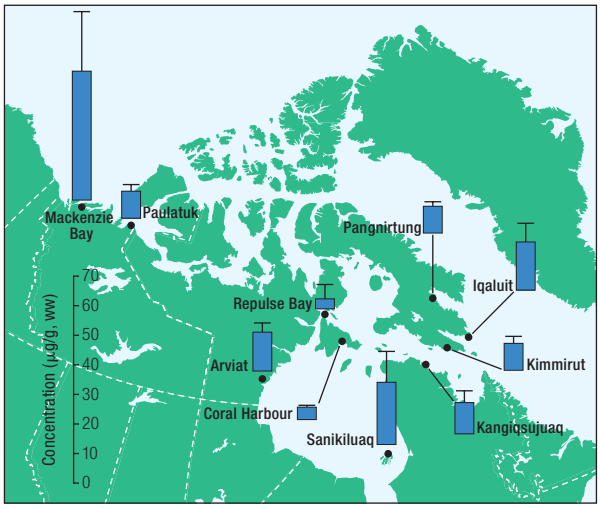

FIGURE 3.3.12

Arithmetic mean concentrations (± 95% confidence limits) of mercury in liver of adult belugas from ten locations in the Canadian Arctic. Sample collections from 1993–2000. Average ages range from 8–15 years.

1997). Selenium levels were high in all three types of walrus tissues; however, cadmium concentrations were elevated only in liver and kidney. The mercury and cadmium levels found in walrus tissues were comparable to those reported previously by Wagemann *et al.* (1996) for the same population at Inukjuak.

Beluga

Beluga liver samples from 10 communities or regional hunts (seven in Nunavut; two in the Inuvialuit Settlement Region (Mackenzie Bay and Paulatuk), and one in the Hudson Strait in Nunavik) collected from 1993–2000, were analyzed for mercury to assess both spatial and temporal trends (Stern *et al.*, 2001b; Sang *et al.*, 2000) (Annex Table 9). Temporal trends of mercury in beluga are discussed in Section 4.3.3. The geographic trend for mercury in adult beluga (range of mean ages from 8–15 years) is shown in Figure 3.3.12. There are major differences in mercury concentrations between the beluga in Mackenzie Bay and beluga at Paulatuk despite all being part of the same southern Beaufort Sea stock. The mean age of the Paulatuk and Mackenzie Bay samples was the same (14 years). The samples from Paulatuk were collected in 1993 and those reported here for Mackenzie Bay are from 1996. While mercury increased in the Mackenzie Bay animals by about 20% from 1993 to 1996 the difference between Paulatuk and Mackenzie Bay was apparent in previous assessments which compared animals collected the same year (Dietz *et al.*, 1998). This regional difference within the stock may be related to a subgroup of animals

returning to their same feeding groups each year and illustrates the large variation that occurs even within a geographically distinct group of beluga. This variation is also apparent in the range of mean concentrations of mercury in the samples from communities in Hudson Bay and Baffin Island (Figure 3.3.12). Mercury concentrations are generally significantly lower than in the Mackenzie Bay group but do not differ from the results for Paulatuk. These differences, even within what are usually classed as the same stocks (e.g., Hudson Bay), were observed in previous surveys (Wagemann *et al.*, 1996; Dietz *et al.*, 1998) and may be due to feeding in the same area each year.

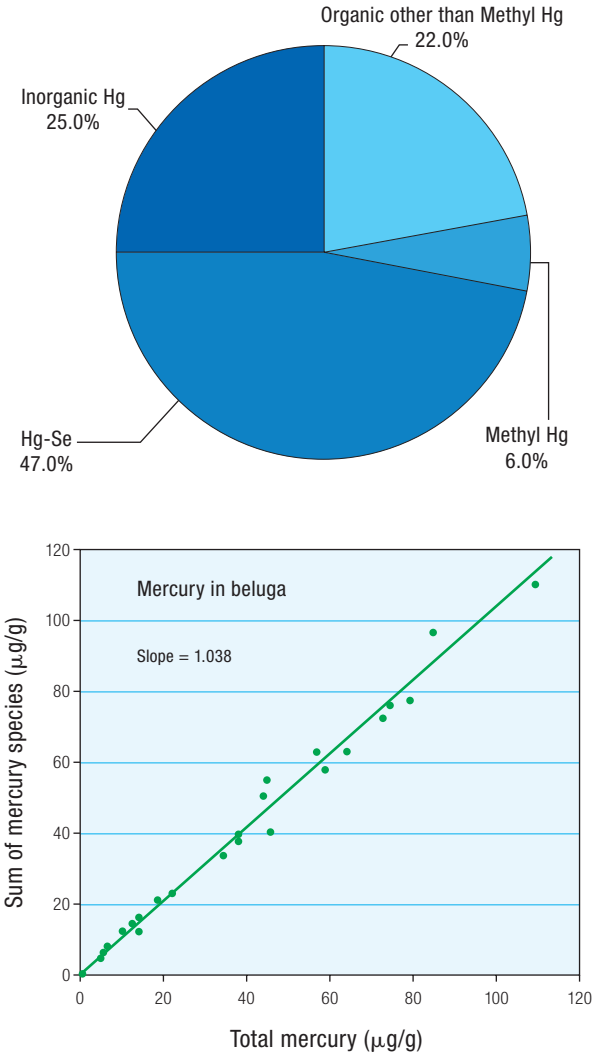

FIGURE 3.3.13

Speciation of mercury in beluga liver is shown in the pie chart. Percentages are arithmetic means. The sum of the individual forms of mercury is highly correlated with total mercury (bottom graph) in beluga liver.

Speciation of mercury in beluga tissues

Results show clearly that there are several forms of mercury present in the beluga tissues with the predominant one in liver showing properties consistent with those of mercuric selenide (Figure 3.3.13). The fact that the four forms identified add up to the same amount as the independent measurement of total mercury argues that these four forms account for all of the mercury. Speciation work in blood of beluga (Lockhart *et al.*, 1999a; 1999b) has shown that the dominant form there is methyl-Hg and so the deep organs must change the form of mercury from that supplied by the blood to that found in the organs. This probably represents a net reduction in toxicity since methyl-Hg is regarded as the most toxic form. This would be consistent with higher proportions of forms other than methyl-Hg at higher levels. Consequently, the conversion of methyl-Hg to other less toxic forms may be an adaptation that allows the whales to survive in spite of their high body burdens of mercury. Microscopic examination of whale samples will offer more detailed insight into the question of the implications of these mercury levels and species for the whales.

3.3.3.2 Organochlorines in marine mammals

Ringed seals

Temporal trend studies have provided new data on OCs in Canadian Arctic ringed seals since the first CACAR (see Section 4.3.1). Previous studies had shown that OC levels were higher in Canadian ringed seals than those from Alaska but lower than what is found in ringed seals from the European Arctic (Muir *et al.*, 2000c). Some geographic variation in OCs in ringed seals was observed within the Canadian Arctic but the magnitude is small and generally within a factor of two (Muir *et al.*, 1999b). OC concentrations in ringed seals fall between lower trophic level organisms, such as the Arctic cod, and the top trophic level organisms, such as the polar bear, reflecting their trophic position.

For most OCs the lowest levels are observed in seals from the western and central Canadian Arctic archipelago as reported for ΣPCB in female ringed seals (Figure 3.3.14) (Annex Table 10). Females were selected because their concentrations of OCs are not strongly affected by age. Ringed seals from Holman, NWT (Hoekstra *et al.*, 2002c) had levels of PCBs and OC pesticides that fell between levels found in ringed seals from the Beaufort/Chukchi Seas on the Alaska north slope (Hoekstra *et al.*, 2002d; Kucklick *et al.*, 2002) and further east (eastern

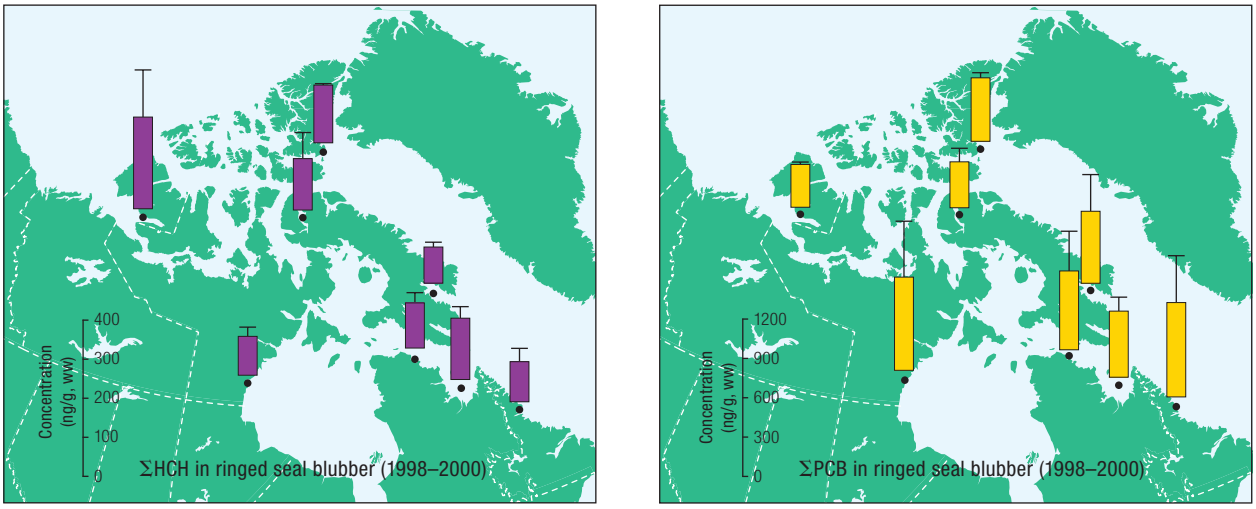

FIGURE 3.3.14

Mean (± 95% confidence interval) concentrations of ΣPCB and ΣHCH in female ringed seal blubber (ng/g ww) from samples collected between 1998–2000.

Canadian Arctic; Fisk *et al.*, 2002c). Similar trends are seen for ΣDDT and chlordane-related compounds. The exception is HCH, which is higher in the western Canadian Arctic and in the Alaska north slope due to prevailing high levels in seawater in the Chukchi/Beaufort Seas (Li *et al.*, 2002).

Smaller differences in mean OC concentrations in ringed seals were seen among seven sites within Nunavik and Labrador in 1998 and 1999 (Muir *et al.*, 2000a) (Annex Table 10). For comparison among regions, three sites within Ungava Bay were combined (Kuujjuaq, Kangirsuk and Kangiqsualujjuaq), as were two sites in the Hudson Strait (Salluit and Quaqtaq). The two sites within Labrador (Nain and Makkovik) were treated separately. Average blubber concentrations of ΣPCB, the major OC contaminants in seals from these four locations, ranged from 572–1042 ng/g ww in males, and from 512–730 ng/g in females (Muir *et al.*, 2000a). The highest levels in males were found in samples from the more westerly locations of Ungava Bay and Hudson Strait, while the highest average levels in females occurred in samples collected at Nain. DDT-related compounds were also prominent contaminants in ringed seal blubber with average concentrations ranging from 198–884 ng/g in males. Analysis of covariance did not reveal any significant differences in concentrations of ΣPCB or ΣDDT levels among the four locations for females. For males, ΣPCB, ΣDDT and ΣHCH concentrations were significantly higher in samples from Ungava Bay, after adjusting for age. There was a significant interaction of age and location (i.e., a differing relationship of age with concentrations of the organochlorines) for males, however, which may have affected the results.

In general, the spatial trends in levels of PCBs, ΣDDT and ΣHCH in ringed seal blubber collected in 1998–2000 from the Canadian Arctic were comparable to those found previously in ringed seal blubber at other eastern Arctic locations (Muir, 1998; Weis and Muir, 1997). Results for the northern Baffin Bay ringed seals were also in the range reported for ringed seals from northeast and central west Greenland (Muir and Johansen, 2001). After removing the influence of age, sex and blubber thickness, OC concentrations did not differ between ringed seals from the east and west side of the northern Baffin Bay, likely due to the relatively small distance between these two sites (Fisk *et al.*, 2002c).

Blubber from ringed seals collected in 1993 near Kimmirut, Nunavut, were analyzed for coplanar PCBs (CoPCBs) (Helm *et al.*, 2002). ΣCoPCB (sum of congeners 77, 81, 105, 126, 118, 114, 156, 169) levels ranged from 14.9–32.7 ng/g ww, with congeners 118, 105 and 156 being the most dominant. Mean (± SD) levels of ΣCoPCB were lower in female (15.4 ± 0.8 ng/g) than male (26.7 ± 8.0 ng/g) ringed seals. Toxic equivalents (TEQs) for CoPCBs in males and females were 624 ± 144 fg/g and 508 ± 88 fg/g, respectively, most of which was accounted for by congener 126 (Helm *et al.*, 2002). No other data were available for comparison.

Toxaphene was measured in the Hudson Strait and Ungava Bay ringed seals (Muir *et al.*, 2000a) (Annex Table 10). As is typical of marine mammals, congeners 26 and 50 were predominant. Octa- and nonachlorobornanes were the major homolog groups (Muir *et al.*, 1999c), and total toxaphene levels in blubber were similar to those reported by Cleemann *et al.* (2000) for seals in Greenland. Toxaphene congeners 39, 40 and 42 were

also identified in the blubber of ringed seals from Arviat, NWT (Loewen *et al.*, 1998), marking the first time that congener 42, the most toxic congener in technical toxaphene, has been found in any significant concentration in a marine mammal.

Walrus

Walrus are long-lived benthic feeders and as such are an important indicator species for bioaccumulating contaminants in benthic marine food webs. Although they have an important role in native traditional hunts, relatively little is known about levels of OCs in walrus compared to seals and beluga. Some individual walrus feed at higher trophic levels than others and as such have much higher contaminant concentrations (de March *et al.*, 1998). These walrus are believed to be including ringed seals in their diet (Muir *et al.*, 1995).

OC levels in walrus from east Hudson Bay (northern Québec) were much lower than those found in earlier studies from the same area (Muir *et al.*, 2000a) (Annex Table 10), likely because the earlier studies sampled walruses that consumed seals (Muir *et al.*, 1995). Nonetheless, concentrations of many OCs were comparable to those in recent reports for walrus from other nearby regions such as Foxe basin (Muir *et al.*, 1995). Levels of ΣPCB were similar to those in walrus from the Bering Sea and NW Greenland, while those of ΣDDT were similar to levels in walrus from NW Greenland (Muir *et al.*, 2000d). ΣHCH and HCB were also detected at concentrations similar to those in walrus from the Bering Sea, E and NW Greenland (Muir *et al.*, 2000d). In the case of ΣCHL, levels in E. Hudson Bay walrus were higher than those in the Bering Sea and NW Greenland, but lower than in east Greenland walrus (Muir *et al.*, 2000d).

Beluga whales

The beluga or white whale (*Delphinapterus leucas*) is a small (up to 4.5 m long) toothed cetacean (odontocete) that has a circumpolar distribution in the Arctic. Beluga whales feed near the top of the marine food web on a variety of fish, as well as invertebrates such as cephalopods and shrimp (Banfield, 1974) and are relatively long-lived (> 35 years). They are present from Alaska across the Canadian Arctic to western Greenland, with large populations in Hudson Bay and among islands in the eastern Canadian Arctic. Beluga movements are extensive, seasonal and are generally predictable. They come into coastal waters and estuaries in mid-summer, and winter offshore in pack ice and polynyas (Brodie, 1989). Like ringed seals, beluga are an important and common diet item of the Inuit.

The previous CACAR assessment included results (to 1996) from belugas from Hudson Bay, Hudson Strait, Cumberland Sound and Mackenzie Bay. Since then, additional samples have been analyzed from Cumberland Sound and Mackenzie Bay as a result of temporal trend studies (Stern, 1999; Stern and Addison, 1999; Stern and Ikonomou, 2001) and as part of studies in Nunavik (Sang *et al.*, 2000). PCBs were quantitatively the most predominant OCs in the blubber of belugas sampled in Mackenzie Bay, east Hudson Bay, Hudson Strait and Cumberland Sound, followed by DDT and chlordane-related compounds (Annex Table 10). OC levels for beluga from Pangnirtung, Baffin Island were similar to, but somewhat higher than levels in the east Hudson Bay/Hudson Strait beluga or Mackenzie Bay (Annex Table 10). Higher concentrations of all major groups of organochlorines, except for ΣHCH, were observed in blubber of males (n = 8) than in females (n = 2). Data were too limited to evaluate effects of age or condition of the animals on contaminant levels. There are no previous studies on contaminants in beluga blubber samples from the Nunavik side of Hudson Strait area (Kangiqsujuaq) for comparison. Levels of major organochlorine groups in male beluga blubber sampled by Sang *et al.* (2000), however, were similar to previous reports for male beluga from eastern Hudson Bay (Nastapoka River) in the mid-1980s (Muir *et al.*, 1990b), and much lower than levels in beluga from the southeast Baffin beluga stock. Muir *et al.* (1990b) concluded that belugas sampled from the Kangiqsujuaq area of the Hudson Strait are from a population that inhabits eastern Hudson Bay and southern Hudson Strait, and not the Southeast Baffin Island area. Innes *et al.* (2002) also concluded that there are distinct differences in the organochlorine signature of southeast Baffin Island and Hudson Bay belugas. Comparisons between eastern Hudson Bay and western Hudson Bay belugas are complicated by missing age estimates and lipid content data for these whales (Hobbs *et al.*, 2002) (Annex Table 10). It is likely, however, that the eastern Hudson Bay whales' blubber would have had higher lipid content, since they were not biopsied (as the western Hudson Bay whales were). With this in mind it seems that the western Hudson Bay beluga may generally have had higher PCB and OC pesticide concentrations than the eastern Hudson Bay whales.

Levels of coplanar PCBs in six belugas from the same region ranged from 14.4–294 ng/g, with congeners 118, 105 and 156 being the most dominant (Helm *et al.*, 2002). ΣCoPCB were lower in females (60.2 ± 40.3 ng/g) than males (228 ± 113 ng/g). TEQs for CoPCBs in males and females were 1,730 ± 627 fg/g and 1,320 ± 909 fg/g, respectively, most of which was accounted for by congener 126 (Helm *et al.*, 2002).

Mean toxaphene levels in beluga from Pangnirtung were about 10,000 ng/g ww (Stern, 2001) (Annex Table 13), which is at the high end of the range reported in beluga from Alaska and Svalbard (Wade *et al.*, 1997) or in previous studies of the Canadian Arctic stocks (Muir *et al.*, 1999b).

Narwhal

Narwhal (*Monodon monoceros*) are deep-water benthic feeders and are an important Arctic species in the diet of Inuit. Despite this, they have received less attention in terms of contaminant studies than beluga. In the Canadian Arctic, levels of PCBs, DDT and chlordanes (not corrected for age) were generally higher in male narwhal from Pont Inlet (NE Baffin Island), than those from Broughton Island (E. Baffin Island), or Grise Fiord (S. Ellesmere Island). Similar trends occurred for toxaphene in narwhal from these sites (Stern, 2001) (Annex Table 10). Levels of PCBs and DDTs in these narwhal were quite similar to levels reported in western Greenland (Riget, 2001), but they were only about half of those measured in narwhal from Svalbard (Wolkers *et al.*, 2002).

Butyltins in cetaceans and pinnipeds

Butyltins (MBT, DBT, TBT) were not detectable in livers from five Hudson Strait belugas sampled in the summer of 1998 (de Mora, 1999). The authors concluded that the limited maritime shipping activities in northern Québec were not sufficient to provide an appreciable input of TBT into the marine environment of northern Québec, and that since these same whales were contaminated with organochlorines, but not butyltins, it is unlikely that aerial input could be a source of organotin compounds to Arctic whale populations. In contrast to this, butyltins were detected in all 21 samples examined from the more southerly St. Lawrence River population of beluga whales located in eastern Canada (de Mora, 1999).

TBT and its metabolites DBT and MBT were non-detectable (sub-ng/g) in ringed seal blubber and liver from Labrador and northern Québec in 1997 (Muir *et al.*, 1999a).

Polar bears

Polar bears (*Ursus maritimus*) are widely distributed throughout the Arctic and sub-Arctic region and range over large areas in search of food. They move south with the ice in the fall and winter and then north as the pack ice melts in the spring and summer. These seasonal movements of the sea ice also influence the distribution and concentration of their primary prey, ringed and bearded seals (Stirling *et al.*, 1982; Kingsley *et al.*, 1985). Polar bears are top Arctic predators, and often eat only the blubber from a seal (Stirling and McEwan, 1975), where the highest concentrations of organochlorines are found. The polar bear therefore has among the highest concentrations of OCs of any Arctic animal. Polar bears also have a superior biotransformation capacity and have the highest levels of OC metabolites, some of which are potent endocrine disruptors (Letcher *et al.*, 2000). Because of this, concerns about the effects of OCs in Arctic wildlife are greatest for this species. Recent research on polar bears in the Canadian and European Arctic have found evidence that the immune system and potentially the endocrine systems of polar bears are compromised by OCs, in particular PCBs (Andersen *et al.*, 2001). Although consumed by the Inuit the polar bear makes up a small percentage of their diet.

A significant amount of research was carried out on polar bears before, and summarized in, the first CACAR. Norstrom *et al.* (1998) assessed the circumpolar trends of OCs in polar bears and included many locations from within the Canadian Arctic. They showed that OC concentrations in Canadian polar bears are slightly greater than in Alaskan polar bears but lower than those observed in eastern Greenland and the European Arctic. Recent work has shown that Russian polar bears possess even greater concentrations of OCs (Lie *et al.*, 2002). Within the Canadian Arctic, minor geographic trends of OC concentrations were observed. Since 1996 (the first CACAR), monitoring of OCs in western Hudson Bay has continued but no other locations have been examined. The work in western Hudson Bay has provided valuable temporal trends (see Section 4.3.2). A significant amount of research has also been carried out on the influence of reproduction on polar bears' OC levels, levels of OC metabolites in polar bears, and on the biological effects of OCs in polar bears.

To study the effect of fasting, gestation and lactation on toxicokinetics in polar bears, OC concentrations were determined in adipose tissue, plasma, and milk samples from seven female polar bears and their cubs near Cape Churchill, Hudson Bay, between 1992–1996 (Polischuk, 1999; Norstrom, 2000). Pregnant females were captured from August 7 to October 7 and the same females with cubs were captured from March 2 to March 17 the following year before they had moved onto the ice to begin hunting seals. All females had therefore been fasting 5–7 months by the time of second capture. Body composition of females was determined from ^{2}H dilution in blood and body weight. The total body mass of females declined by 43 ± 5% and total fat mass declined by 42 ± 3%. The proportion of mass lost as fat ranged between 55–66%.

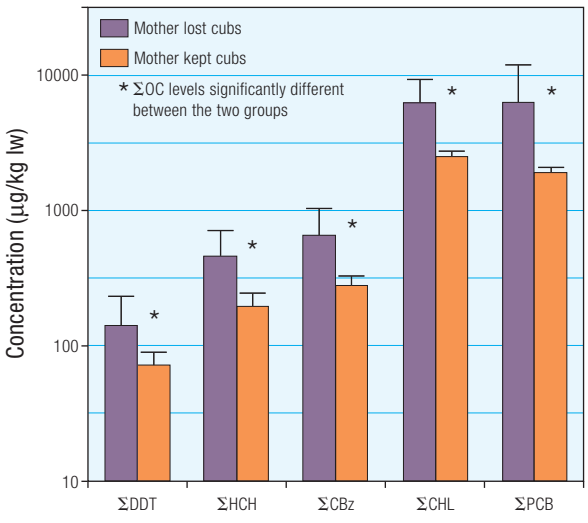

FIGURE 3.3.15

Concentrations (µg/kg lw, mean ± SD) of major POPs in polar bear milk of females with cubs after emerging in March from dens in the Cape Churchill area, Hudson Bay (1992–1996). The data are grouped according to whether the female still had her cubs the following fall, or had lost them.

During gestation and early lactation the mean concentrations of ΣPCB, ΣCHL, and ΣCBz in female adipose tissue increased significantly by 2,096 ± 1,292 µg/kg, 1,600 ± 1,349 µg/kg and 49 ± 23 µg/kg, respectively. Adipose tissue concentrations of ΣDDT declined by 91 ± 82 µg/kg while concentrations ΣHCH remained the same. Despite these increases in concentration, OC body burdens in female polar bears declined during gestation and the early lactation period due to loss of fat mass. In descending order, the mean proportional decrease in body burdens was ΣDDT (75 ± 8%) > ΣHCH (61 ± 8%) > ΣCBz (45 ± 12%) > ΣCHL (29 ± 20%) ≈ ΣPCB (24 ± 16%), while the mean mass of OC lost was ΣCHL (71 ± 57 mg) > ΣPCB (56 ± 34 mg) > ΣDDT (20 ± 8 mg) > ΣHCH (14 ± 7 mg) > ΣCBz (7 ± 2 mg). Biotransformation during gestation and lactation probably accounts for the greater proportional decrease in ΣCBz, ΣHCH and ΣDDT compared to ΣPCB and ΣCHL.

The mean period between first and second capture (mostly in the den) was 188 ± 22 days, and the mean number of days of lactation before sampling was approximately 79 ± 4 days. It is likely that the toxicokinetics of OCs during the first 100 days was similar to that during the subsequent summer fast. The polar bear cub is only about 0.7 kg at birth (Ian Stirling, 1999; personal communication). Two new-born cubs are about 0.4% of the mother's weight; therefore, OC transfer to the fetus is unlikely to be a significant part of the mother's body burden. Most of the 24–29% loss of body burdens of the poorly biotransformed ΣCHL and ΣPCB from the mother during the 188-day fast must have been transferred to the cub in milk during the 79-day lactation prior to capture, at which time the cubs weighed 12.7 ± 0.9 kg. However, on average only about 25–50% of this body burden loss could be accounted for by ΣCHL and ΣPCB burden in the cubs. Furthermore, a crude estimate of lactational transfer based on cub growth rates could only account for half of the body burden loss from the females. These discrepancies are difficult to reconcile and further study is required.

Mothers that were recaptured in the autumn without cubs had high OC concentrations in their milk when they emerged from the den in the previous spring (Figure 3.3.15). By comparison, mothers recaptured in fall still accompanied by cubs had low OC concentrations in their milk the previous spring (Figure 3.3.16). The differences in concentrations were significant for all residue classes. For example, PCBs were about 3 times higher (5,780 ng/g lipid) in females that lost their cubs than in females that kept their cubs (1,830 ng/g lipid). It is not known how much significance can be attached to these findings in terms of reproductive performance, but it is suggestive, at least, that cub survival may depend on the degree of exposure to OCs in milk.

Distribution of OCs in plasma relative to adipose tissue was determined in pregnant females in the summer at the beginning of the on-land fast, in females with cubs emerging from the den, in females with cubs in summer, and in females with cubs after 1–2 months fasting near Cape Churchill, Hudson Bay, between 1992–1996 (Polischuk, 1999; Norstrom, 2000). At all of these stages, the ratio of concentration of ΣCHL and ΣPCB

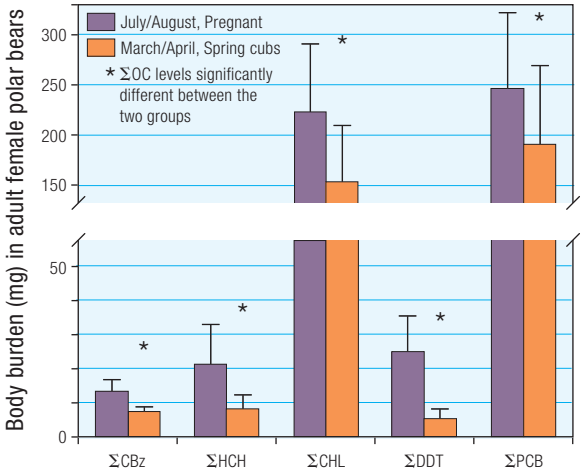

FIGURE 3.3.16

Body burdens (mg, mean ± SD) of major POPs in adipose tissue lipids of polar bear females on land, pregnant in the summer and with cubs-of-the-year (COYs) the following March in the Cape Churchill area, Hudson Bay (1992–1996).

in whole plasma to adipose tissue lipid was the same as the fraction of lipid determined in plasma, 0.01. That is, these OCs were equally distributed in adipose and plasma lipids. Plasma/adipose ratios of ΣCBz in pregnant females in summer and in females the following spring with cubs, and ΣHCH in pregnant females were also distributed according to lipid content. The plasma/adipose ratios of ΣCBz and ΣHCH were 2–3 times higher in the plasma of females with cubs in the summer and fall than predicted by lipid content, suggesting that components of the plasma other than extractable lipid were responsible for some of the carrying capacity of plasma at these times. The plasma/adipose tissue ratio of ΣDDT was close to twice that of the lipid fraction in female plasma at all times. This is probably indicative of binding to proteins in plasma as well as solution in lipids. There was no indication that physical-chemical properties such as water solubility or log K_{ow} were governing the partitioning of OCs between plasma and adipose tissue.

Ratios of OC concentrations in female milk lipid/adipose tissue lipid were determined in spring, summer and fall. The ratio was greatest for ΣHCH and lowest for ΣDDT and ΣPCB. On a lw basis, the ratio ranged from close 1:1 for ΣPCB and ΣCHL to about 2:1 for ΣCBz and ΣHCH, indicating that milk lipids were not at equilibrium with adipose tissue lipids for these compounds. The milk/adipose ratios for ΣCBz were significantly greater than ΣCHL, ΣDDT, and ΣPCB. Similarly, ΣCHL were significantly greater than ΣDDT and ΣPCB.

The rank order of the milk/adipose ratios for the chemical groups was similar to that of the chemical's octanol-water partition coefficients, K_{ow} (Hawker and Connell, 1988; Mackay *et al.*, 1992), unlike plasma. Since it is improbable that concentrations in extractable milk lipids would be higher than those in adipose tissue on a thermodynamic basis, low K_{ow} OCs must partition from adipose tissue to constituents of milk other than extractable lipids, such as lipoproteins.

Concentrations of OCs were determined in adipose tissue from 47 fasting polar bears in the Cape Churchill area of western Hudson Bay in the summer to fall of 1992–95 (Polischuk, 1999; Norstrom, 2000). Body burdens (mg/animal) were also determined based on body composition determined from ^2H dilution in blood and body weight. Adipose tissue, milk and blood samples were taken when the bears came on land in July/August. The same bears were recaptured and sampled in September/October (47–68 days apart). Five categories of bear were defined: cubs-of-the-year, yearlings, females with cubs-of-the-year, females with yearlings, and males. There were no single females sampled.

Concentrations of ΣCBz, ΣCHL and ΣPCB in adipose tissue lipid increased, and concentrations of ΣDDT decreased in all bears during the 47–68-day fast. The changes in concentration of ΣCHL and ΣPCB in adults were mostly significant. In cubs-of-the-year, the concentrations of ΣCHL and ΣPCB increased 30%, while those of ΣHCH and ΣDDT decreased by 20% and 34%, respectively, during the fasting period. Increases in concentration in adipose tissue in adults were entirely due to utilization of lipid by the bears. Most of the increases in concentration in cubs-of-the-year were due to decreased adipose tissue, not uptake from nursing. The body burden of ΣCHL and ΣPCB in females with cubs-of-the-year did not decrease during fasting. Taken together, these data suggest that the cubs-of-the-year were nursing very little during the 47–68-day period.

Biotransformation of 4,4′-DDE and HCHs occurred in all bears during the fast, as did biotransformation of oxychlordane and some other chlordane compounds in adult males. Chlorobenzenes, oxychlordane (except in males) and the major PCB congeners (CB60, CB99, CB138, CB153, CB156, CB170, CB180, and CB194) were not metabolized or cleared by any other mechanism at a measurable rate over 47–68 days of fasting.

The concentration of compounds with slow biotransformation rates during fasting did not increase with age, except for highly chlorinated PCBs (CB180 and CB194) in males. That is, average annual rate of intake and excretion of the OCs are balanced. Clearance of all OCs by a mechanism other than biotransformation is therefore occurring for at least part of the year. For most of the OCs, there was no significant difference in concentrations or trends in males and females. Lactation is therefore not governing the rate of excretion in females, except perhaps for octachloro and nonachloro PCBs. The only clearance mechanism for slowly biotransformed OCs which remains available to both sexes is partitioning to gut contents and excretion in feces during periods when they are feeding.

Toxicokinetics of OCs in polar bears are therefore likely to be variable with season, sex/reproductive status and area. Slowly biotransformed PCBs, oxychlordane (except in adult males) and chlorobenzenes will be taken up and excreted in feces only during periods of active feeding. Oxychlordane in males, and HCHs and 4,4′-DDE in all bears, however, is taken up during feeding periods, but biotransformed throughout the year regardless of whether the bear is feeding or not. Due to lack of ice from which to hunt seals during summer and fall in southwest

Hudson Bay, feeding may be restricted to 7–8 months of the year for males, solitary females and females with cubs or yearlings, and as short as four months for pregnant females. In more northerly areas, polar bears (except pregnant females) may not fast at all if they can remain on ice to hunt ringed seals, or have marine mammal carcasses to scavenge. In this case, uptake and clearance will occur year-round.

Arctic fox

The Arctic fox (*Alopex lagopus*) is one of the few species that constitutes an important component of both the terrestrial and marine ecosystems (Hiruki and Stirling, 1989). On land, Arctic foxes feed mainly on lemmings, birds and their eggs as well as scavenging on caribou remains (Kennedy, 1980; Stickney, 1991; Kapel, 1999) whereas coastal foxes will also eat marine invertebrates (Fay and Stephenson, 1989; Kapel, 1999) and fish (Banfield, 1987) in summer. In winter and spring on the sea ice, Arctic foxes scavenge the remains of ringed seals and bearded seals killed by polar bears (Stirling and Smith, 1977; Andriashek *et al.*, 1985; Fay and Stephenson, 1989; Hiruki and Stirling, 1989; Kapel, 1999) and they will also actively prey on newborn seal pups (Smith, 1976; Andriashek *et al.*, 1985; Lydersen and Gjertz, 1986). Populations feeding at different trophic levels (Peterson and Fry, 1987; Hobson and Welch, 1992), and in marine or terrestrial/freshwater environments (reviewed by Hobson *et al.*, 1997) can be distinguished using stable isotope analyses. The $^{15}N/^{14}N$ isotope ratio in biota increases in a predictable manner between trophic levels (Kelly, 2000) allowing trophic positions and organochlorine concentrations to be interpreted along a continuous variable. Carbon isotopes ($^{13}C/^{12}C$) are not enriched significantly between trophic positions and can be used to evaluate energetic pathways between regions with differing carbon sources. As a result, stable carbon isotope signatures can differentiate an organism's dependence on inshore/benthic and offshore/pelagic regions (Hobson *et al.*, 1995).

Arctic fox muscle and liver tissues were collected at Barrow, Alaska and Holman, NWT while muscle samples were available from Arviat (NU) from 1998 to 2000. Hoekstra *et al.* (2002e) investigated the feeding ecology of this species at these three sites and related these findings to body residue patterns of OCs (Annex Table 11). At all three sites, PCBs and chlordane-related compounds were the predominant organochlorines present in muscle, while in liver chlordanes were the predominant group, followed by PCBs. The most abundant OC analytes extracted from Arctic fox muscle and liver were oxychlordane, the principal metabolite of chlordane-related compounds, CB153, and CB180. While mean

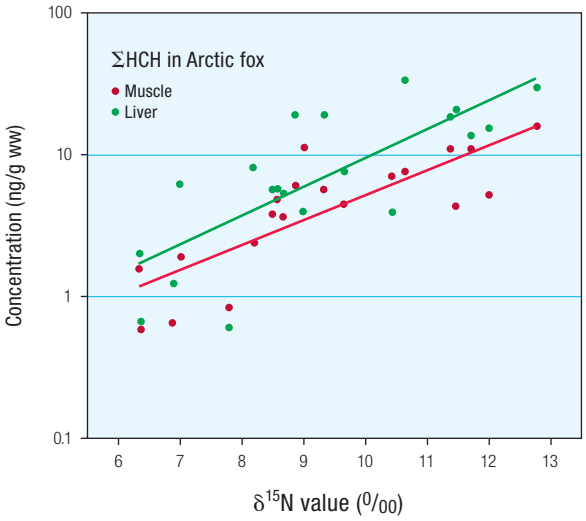

FIGURE 3.3.17

Nitrogen stable isotope ratios ($\delta^{15}N$) versus ΣHCH in Arctic fox by tissue (Hoekstra *et al.*, 2002e). Relationship for muscle (red line; log HCH = 0.175 $\delta^{15}N$ – 1.04; r^2 = 0.67) and liver (green line; log HCH = 0.204 $\delta^{15}N$ – 1.07; r^2 = 0.59) samples.

concentrations of all major organochlorines groups were < 250 ng/g ww, a few individuals had very high levels of total PCBs and oxychlordane. The concentrations of organochlorines were quite elevated on a lipid basis. For example, ΣPCB ranged from 110–14,600 ng/g lw in liver, and from 76–8,047 ng/g and 580–5,020 ng/g lw in muscle in the Holman and Arviat foxes, respectively. There were no significant differences between mean concentrations of major OC groups in Arctic fox muscle from Holman and Arviat. However, the mean lipid adjusted ΣPCB concentrations in the foxes from Holman, NWT and Barrow, Alaska (1,853 ± 730 ng/g and 1,516 ± 370 ng/g, respectively) are lower than ΣPCB in Arctic fox liver from Svalbard, the Norwegian mainland, and Iceland (Klobes *et al.*, 1998; Norheim, 1978; Skaare, 1996; Wang-Andersen *et al.*, 1993), which is consistent with the west-east gradient of ΣPCB concentrations in ringed seals (Muir *et al.*, 2000c). The OC profile in the Arctic fox suggests a similarity in metabolism with the polar bear and that the capacity of this species to biotransform OCs dramatically influences accumulation profiles.

Concentrations of several OC groups in both liver and muscle samples were not significantly correlated with increasing trophic position based on stable nitrogen isotope ($^{15}N/^{14}N$) values from the Holman and Barrow foxes (Hoekstra *et al.*, 2002e). ΣHCH had the strongest positive relationship with trophic position (Figure 3.3.17). The stable isotope data suggest that as the Holman Arctic fox population shifts its feeding from the terrestrial/freshwater systems to the marine environment, its

relative trophic position significantly increases. However, most organochlorine levels for sum-groups (i.e., ΣDDT, ΣPCB, ΣCHL, etc.) were not strongly correlated to trophic position. The comparison of $\delta^{15}N$ with OC concentrations indicated that trophic position does not serve as an accurate predictor for OC bioaccumulation in the Arctic fox, possibly due to different turnover rates of OCs and stable isotopes in metabolically active tissue and (or) that the capacity of the Arctic fox to readily biotransform OCs.

Effects of age and sex on organochlorine levels in marine mammals

Age and sex are important factors that must be taken into account to assure accurate comparisons of contaminant levels in marine mammals, such as variations in geographic and temporal trends. Concentrations of ΣPCB, ΣDDT and ΣCHL compounds were found to increase with age, for both male and female ringed seals in the Nunavut region (Fisk *et al.*, 2002c). Sex was an important variable within the context of seal age, and in seals of similar ages OC concentrations were greater in males than in females (Fisk *et al.*, 2002c), as has been previously observed in ringed seals (Weis and Muir, 1997). These relationships were not observed for ΣCBz and ΣHCH in seals from the Nunavut region, although there was an age-within-site significance for ΣHCHs (Fisk *et al.*, 2002c). These data are consistent with previous findings for ringed seals, where positive relationships between OC concentration and age have been observed for male ringed seals (Muir *et al.*, 2000c; Wolkers *et al.*, 1998) but are less commonly observed in female seals (Kostamo *et al.*, 2000). Reproduction, including birth and lactation, provides a means for female seals to reduce body burdens of OCs, and counters the effect of accumulation of OCs with age. Differences in the slope of age-OC concentration relationships between male and female ringed seals from Nunavut suggest that birth and lactation are important variables in female ringed seal OC concentrations (Fisk *et al.*, 2002c).

The effects of age and sex are also important considerations with respect to PCBs and OC pesticides in polar bears. In 1995, Norstrom (1999) examined concentrations of organochlorines in male and female polar bears from Hudson Bay to test what the effect of sex would have been in a single year. Concentrations of organochlorines in males were similar to those in females; however, there was a (generally non-significant) tendency for most residues to be lower in males, except PCBs and DDTs, which were slightly higher in males. The difference in PCBs between males and females was not as large as observed in more extensive data sets. Norstrom

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et al. (1998) showed that males had 40% higher PCBs than females on average. Younger male bears have PCB concentrations closer to those in females, which may explain the similarity between males and females in 1995. Males had 30% lower levels of chlordanes than females, which is the same as that found by Norstrom *et al.* (1998). Polischuk *et al.* (2002) showed that males are capable of metabolizing chlordanes during a seasonal fast, while females are not. The enhanced metabolic capability of males therefore explains the lower levels of chlordanes, and possibly some of the other compound groups, in males. Lower levels of PCBs and lack of an age effect in females are presumed to be due to the additional losses from lactation.

Organochlorines in other marine mammal tissues

Fewer analyses of brain, liver, kidney, muscle and blood have been made as compared to blubber in pinnipeds and cetaceans. In all tissues, however, OC concentrations are lower than in blubber, because of their lower lipid content. Low ng/g ww levels of PCBs and OC pesticides were found in brain, liver and muscle from beluga whales from Hendrickson Island, in the western Canadian Arctic (Metcalfe *et al.*, 1999) (Annex Table 10). Lipid content is an important factor in controlling accumulation of lipophilic OCs and in beluga whales concentrations of OCs in these tissues, with the exception of brain, are generally more comparable when the values were calculated on a lipid weight rather than on a wet weight basis (Krahn *et al.*, 2001). Beluga brain samples could be distinguished from other tissues by differences in PCB congener patterns and higher concentrations of ΣHCH (primarily α-HCH) (Metcalfe *et al.*, 1999). These variations in patterns of persistent organic pollutants in the different tissues may be influenced by

differences in contaminant metabolism, the content and composition of lipids, or the degree of blood perfusion in the various tissues (Metcalfe *et al.*, 1999).

The need for caution in interpreting OC concentration data from different tissues is further demonstrated by Lydersen, *et al.* (2002) who examined how concentrations of OCs in blood and blubber vary with nutritional condition in captive and wild fasting ringed seals. The study demonstrated that extreme variability occurs in the concentrations of OCs in seal blood in response to change in body condition as a result of fasting, and that the reponse of blubber OC concentrations are also very different than those in blood. The authors concluded by recommending that since the natural variation in condition is extreme during phocid seals' annual cycles, blood should not be used in studies of OCs where the aim of the study is to monitor OC levels of comparative purposes or time-trend analysis.

3.3.3.3 New compounds in marine mammals

The greatest amount of the data produced on new chemicals in the Arctic has been produced for marine mammals and in particular seals and whales (Annex Table 12). These are logically the animals or matrices to check for new chemicals as seals and whales have among the highest levels of OCs in the Arctic.

Synthetic musks

The synthetic polycyclic musk HHCB (1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta[g]-2-benzopyrane; e.g., galaxolide®) was detected in ringed seal blubber samples from Labrador at low ng/g concentrations (Bidleman *et al.*, 2001). HHCB and its metabolite HHCB-lactone musk were near detection limits; nevertheless, strong evidence for their presence was provided by the use of high resolution mass spectrometry and use of blank samples. HHCB is an important artificial fragrance used in a large number of perfumes, laundry detergents, fabric softeners, toiletry products, and other household products. The nitro musks, musk xylene (MX) and musk ketone (MK) and another synthetic polycyclic musk (AHTN) were also included in the study but were not detected.

Perfluorooctane sulfonate (PFOS)

PFOS, a member of the perfluorinated acid group of chemicals, used as stain resisting surfactants and in fire-fighting foams, was detected in the blood of ringed seals from Pangnirtung (Kannan *et al.*, 2001; Geisy and Kannan, 2001). No other samples from Canadian Arctic biota were analyzed. No obvious trends between the Canadian and European Arctic in PFOS in ringed seals was observed, although only a very small data set was available. PFOS was also detected in liver of polar bears from northern Alaska (Kannan *et al.*, 2001; Geisy and Kannan, 2001) and are therefore likely to be present in polar bears in the Canadian Arctic. The levels of PFOS in polar bear liver in the 100s of ng/g ww range suggest that this compound is one of the most prominent individual organohalogen chemicals found in polar bears, when levels of PCBs, chlordane and HCH-related chemicals are considered (Norstrom *et al.*, 1998).

Polychlorinated naphthalenes (PCNs)

Polychlorinated naphthalenes (ΣPCN) concentrations in blubber from six ringed seals sampled in 1993 near Kimmirut, Nunavut ranged from 29–63 pg/g ww (Helm *et al.*, 2002). No differences related to sex were observed for distribution of PCN homolog groups and most of the PCN TEQ was due to two hexaCN congeners, CN66 and CN67. The contribution to TEQ by PCNs was negligible (< 0.10–2.4 fg/g ww) due to lack of the more toxic hexaCN congeners. Blubber from six beluga whales collected in 1994 near Pangnirtung, Nunavut were also analyzed for PCNs (Helm *et al.*, 2002). ΣPCN concentrations in the blubber of 6 beluga from nearby (near Pangnirtung, Nunavut) ranged from 40–384 pg/g ww and pentaCNs dominated, followed by hexa- and tetraCNs. ΣPCN levels were lower in females than in males. Most of the PCN TEQ was due to two hexaCN congeners, CN66 and CN67. Although ΣCoPCB concentrations average about 500 times greater than ΣPCN in beluga blubber, PCNs contributed 11% of the TEQ relative to CoPCBs, about the same as the contribution from mono-*ortho* PCBs CB105 and CB118. Thus, PCNs may be significant contributors to TEQ in beluga.

Polybrominated diphenyl ethers (PBDEs)

There have been two temporal trends studies of PBDEs, in beluga (Stern and Ikonomou, 2001) and ringed seals (Ikonomou *et al.*, 2002), which have shown that these compounds are increasing in Arctic marine mammals (see Section 4.3.3). In general concentrations of PBDEs are orders of magnitude less than legacy OCs such as PCBs and DDTs. PBDEs consist of a much smaller number of individual compounds, however, so the difference between individual PBDE and individual PCB congeners is less. BDE47 is the most common congener measured, followed by BDE99 and 153. Other congeners, such as BDE100 and BDE49 have been measured in Canadian Artic beluga (Stern and Ikonomou, 2001). These congeners are found at lower concentrations in the technical PBDE standards but may also be less prevalent in biota due to biotransformation.

Although it is early to make conclusions about spatial trends of PBDE in Arctic marine mammals, there is sufficient data to suggest that concentrations are higher in European mammals compared with those from the North American Arctic. Concentrations of ΣPBDE were 92.9 ± 56.5 ng/g ww in Svalbard beluga blubber collected in 1998 (van Bavel *et al.*, 2001) compared with concentrations of 15.5 ng/g in beluga from the western Canadian Arctic (Stern and Ikonomou, 2001). As well, ΣPBDE concentrations in the blubber of ringed seals from northeastern Greenland (58 ± 23 ng/g) (Muir and Johansen, 2001) were an order magnitude higher than levels reported from western Greenland (3.6 ± 1.1 ng/g) (Muir and Johansen, 2001) and the western Canadian Arctic (4.6 ng/g ww) (Ikonomou *et al.*, 2002).

Chlorinated paraffins (SCCPs)

Chlorinated paraffins (SCCPs) were detected at ng/g levels in beluga whales and ringed seals in the Canadian Arctic (Annex Table 10) (Tomy *et al.*, 2000; Muir *et al.*, 1999d). Concentrations were similar in beluga from Mackenzie Bay and Kimmirut. Ringed seals from Eureka had higher concentrations (mean 527 ng/g) than seals from Pangnirtung (mean for both sexes was 95 ng/g). The seals from Eureka where much older than those from Pangnirtung and it is possible that SCCP levels increase with age as do other recalcitrant OCs although further measurements are needed to confirm this. Both species had slightly higher mean levels in males than females suggesting lactational transfer of SCCPs. The SCCPs profile consisted of similar proportions of chlorodecanes, undecanes and dodecanes with very low or negligible amounts of tridecanes.

Little other published data exists on SCCPs in marine mammals for comparison and differences in analytical methodology make comparisons problematic. Jansson *et al.* (1993) reported 130 ng/g SCCPs in ringed seal blubber from Svalbard which is quite similar to levels found in this study. St. Lawrence beluga whales had 6–8 times higher levels of SCCPs than Arctic beluga (Tomy *et al.*, 2000) which illustrates the importance of local sources for this southern population of whales. The St. Lawrence animals had a pattern of SCCPs which resembled the pattern found in commercial products while the Arctic beluga had much higher proportions of the more volatile chlorodecanes and undecanes. The pattern of SCCPs in the Arctic beluga is consistent with long-range transport sources, i.e., more volatile components are present in higher proportions. There was no correlation between levels of PCBs or toxaphene and SCCPs in ringed seals and beluga. This was unexpected because levels of most recalcitrant OCs in beluga blubber are correlated. It implies that SCCPs undergo a greater degree of biotransformation or have quite different sources than PCBs.

Chiral contaminants

As discussed in the seabird section (Section 3.3.2.3), chiral pollutants exist in two forms as optical isomers called enantiomers. Enantiomers have identical physical-chemical properties and abiotic degradation rates, but can have different rates of biotransformation providing information on the ability of species to biotranform OCs.

Wiberg *et al.* (2000) examined enantiomer ratios ($ER_{(+/-)}$) of α-HCH and several chlordane compounds in the blubber and liver of ringed seals from Resolute Bay in the Canadian Arctic. The $ERs_{(+/-)}$ in ringed seals were frequently nonracemic [$ER_{(+/-)} \neq 1$], due to enantiomer-specific biotransformation; however, cod from the same region showed near-racemic mixtures [$ER_{(+/-)} = 1$] for most compounds. (+)-α-HCH was more abundant than (-)-α-HCH in ringed seals. There was no uniform trend for the $ER_{(+/-)}$ changes in the various chlordane compounds examined. It was also determined that oxychlordane was formed in ringed seals, and metabolized by polar bears that preyed on them, and the $ER_{(+/-)}$ had an important role in the class separation of male/female seals and fat/liver tissues.

Enantiomer Fractions (EFs) of α-HCH and other chiral contaminants in seal blubber may not reflect the metabolic capability of seals. Wiberg *et al.* (1998; 2000) noted near racemic α-HCH (EFs = ~0.52) in blubber of ringed seals but non-racemic values in liver (EFs = ~0.6). This phenomenon was observed with other chiral pollutants, such as trans-chlordane, but in some cases the EF was greater in blubber (Wiberg *et al.*, 2000). Wiberg *et al.* (1998) attributed this difference to greater metabolic activity in the liver compared to the blubber. This would imply that the proportion of the α-HCH body burden that is transformed is small and consequently the EF in ringed seal blubber is closer to that in the diet than in liver. This is not always the case, since EFs of many chiral OCs in ringed seal blubber have been found that do not match their main prey item, Arctic cod (Wiberg *et al.*, 2000; Moisey *et al.*, 2001). In seven species of seabirds, animals that do not retain such a large reserve of fat as found in ringed seals, there were no differences in EFs of chiral chlordanes between liver and fat (Fisk *et al.*, 2001c). Differences in EFs of chiral pollutants between tissues of seals, and potentially other marine mammals, require further study.

EFs of chiral contaminants and stable isotopes of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) were measured along with OCs in ringed seals collected from the east and west side

of the NOW Polynya in northern Baffin Bay (Fisk *et al.*, 2002c). *Cis*- and *trans*-chlordane, oxychlordane and heptachlor epoxide were all non-racemic in the ringed seal blubber but did not vary with age, sex or collection site. Alpha-HCH appeared racemic (enantiomeric fraction = 0.50 ± 0.01) in the seals, although this EF is different than those previously observed in their prey species, and was found to vary significantly with age. An overall food web assessment of α-HCH in the NOW Polynya, which included the ringed seal data of this study, concluded that ringed seals do not metabolize α-HCH efficiently (Moisey *et al.*, 2001). EF values in the ringed seals varied considerably from other Arctic marine mammals and seabirds providing additional evidence that the type(s) and characteristic(s) of the enzymes involved in biotransformation of chiral OCs vary between these organisms.

Organochlorine metabolites

Knowledge is limited regarding methylsulfone ($MeSO_2$)-PCBs and $MeSO_2$-DDE in cetaceans. PCBs, DDT, 28 $MeSO_2$ metabolites of 14 meta-para chlorine-unsubstituted PCBs, and four $MeSO_2$ metabolites of 4,4′- and 2,4′-DDE were found in the blubber of beluga whales from western Hudson Bay and the St. Lawrence River, which are contrasted by their exposure to different levels of cytochrome P450 enzyme-inducing, chlorinate hydrocarbon contaminants (Letcher *et al.*, 2000). The mean concentrations of Σ$MeSO_2$-PCB in male western Hudson Bay beluga (159 ng/g), and ratios of Σ$MeSO_2$-PCB to ΣPCB (0.03) and Σprecursor-PCB (0.08) were approximately two-fold lower, whereas the Σprecursor-PCB to ΣPCB ratio was approximately two-fold higher, than in beluga males from the St. Lawerence. Both populations had a low formation capacity for $MeSO_2$-PCBs with ≥ six chlorines (< 4% of Σ$MeSO_2$-PCBs). The congener patterns were dominated by trichloro- and tetrachloro-$MeSO_2$-PCBs in the western Hudson Bay animals, whereas they were dominated by tetra- and pentachloro$MeSO_2$-PCBs in the St. Lawrence beluga. In addition to 2- and 3-$MeSO_2$-4,4′-DDE, two unknown $MeSO_2$-2,4′-DDEs were detected. The mean 3-$MeSO_2$-DDE concentrations in western Hudson Bay (< 0.01ng/g) animals was much lower than in the St. Lawrence animals (1.2 ng/g). The authors demonstrated that sulfone formation and clearance is related to metabolic capacity, and thus PCB, DDE and $MeSO_2$-PCB and -DDE toxicokinetics differ for St. Lawrence and western Hudson Bay beluga.

A potential metabolite of octachlorostyrene (OCS), an industrial by-product, was recently discovered at relatively high concentrations (similar to CB153) in polar bear plasma (from the Barrow Strait) (Sandau *et al.*, 2000). The metabolite was identified in the phenolic compound fraction and was one of the most abundant compounds in that fraction. Further investigation revealed that 4-OH-HpCS was also present in the blood of other Arctic species including the beluga whale, walrus and ringed seal (unpublished results). Concentrations of CB153, 4-HO-CB187 (the main hydroxylated PCB metabolite in polar bears), OCS and 4-OH-HpCS in Arctic marine mammals are given in Table 3.3.1. Polar bears have demonstrated superior metabolic capability and this is demonstrated with the concentration of 4-HO-HpCS found to be 20–30 times more abundant than OCS. The other Arctic species have much lower concentrations of the metabolite in their blood, which may indicate less metabolic capability.

Concentrations of OCs were determined in the plasma of Resolute Bay polar bears (Sandau *et al.*, 2000) (Annex Table 11). This work also included plasma samples from Svalbard polar bears. ΣCHL and ΣPCB were the dominant OC groups found in the plasma of polar bears from both regions. OC concentrations were 2 times higher in subadults than adults, except for DDTs that were similar, in line with previous findings in polar bear adipose tissue. The exceptions were chlordanes, which were 30–60% lower in males, but concentrations were similar comparing the same sex in both areas. It has been shown that males metabolize chlordanes more readily than females, which accounts for the differences between

TABLE 3.3.1 Concentrations (mean ± SD, wet weight) of contaminants in Arctic marine mammal plasma

Species	4-HO-CB187 (pg/g ww)	4-HO-HpCS (pg/g ww)	OCS (pg/g ww)	CB153 (pg/g ww)	4-HO-HpCS:OCS
Walrus (n = 9)	66.9 ± 113	15.7 ± 14.1	216 ± 144	662 ± 898	0.08 ± 0.08
Ringed seal (n = 5)	17.1 ± 12.8	45.1 ± 18.4	293 ± 95.3	568 ± 409	0.17 ± 0.09
Beluga whale (n = 6)	8.60 ± 11.7	1.47 ± 0.67	31.6 ± 13.9	2910 ± 1530	0.05 ± 0.02
	(ng/g ww)	**(ng/g ww)**	**(ng/g ww)**	**(ng/g ww)**	
Polar bear (Svalbard, n = 32)	54.2 ± 67.2	7.34 ± 4.18	0.493 ± 0.314	31.3 ± 19.9	20.1 ± 16.3
Polar bear (Barrow Str, n = 24)	25.5 ± 14.9	7.82 ± 3.60	0.342 ± 0.195	14.7 ± 8.71	31.0 ± 27.9

genders (Polischuk *et al.*, 2002). ΣPCB concentrations were similar in males and females from both areas, and 2–3 times higher in the Svalbard bears, in line with previous analyses of adipose tissue from these areas.

There have been a number of comprehensive studies on OC metabolite formation, mainly $MeSO_2$- and OH-PCBs, in polar bears (Letcher *et al.*, 1996; Norstrom, 1997; Sandau *et al.*, 2000). Polar bears have a superior ability to biotransform OCs. For example, the PCB burden in polar bears is dominated by a small number of congeners, much less than what is observed in their major prey item the ringed seal (Muir *et al.*, 1988).

Phenolic compounds in blood plasma were identified and determined along with neutral OCs in polar bear plasma from the Resolute Bay area in the Canadian Arctic and Svalbard (Sandau *et al.*, 2000). A total of 35 compounds in the phenolic fraction were identified as hydroxy-PCBs (OH-PCBs) in the two populations. In addition to OH-PCBs, a previously unidentified phenolic metabolite of octachlorostyrene, 4-hydroxyheptachlorostyrene (4-OH-HpCS) was identified in polar bear plasma (Sandau *et al.*, 2000). Traces of pentachlorophenol at about 0.2 ng/g were also found. Mean concentrations of ΣOH-PCB in polar bear plasma ranged from 57 ng/g in males from Resolute to 218 ng/g in females from Svalbard. Females had significantly higher concentrations of ΣOH-PCB than males. The ratio of ΣOH-PCB/ΣPCB in plasma was also significantly higher in females (mean 1.49) than males (mean 4.08). It appears that females either have a higher binding capacity for OH-PCBs in plasma, probably due to higher TTR (transthyretin) concentrations, than males, or a higher capacity to form OH-PCBs. The concentration of ΣOH-PCB was 2–3 times higher than any other residue class in female plasma, and in the other age groups and males was equal to, or higher than, the concentration of next highest residue class, ΣPCB. The concentration of ΣOH-PCB in subadults was the same as that in females. Thus, it appears that there is no selective transfer of OH-PCBs in polar bear milk, unlike $MeSO_2$-PCBs.

To put the importance of OH-PCBs and other phenolic compounds in polar bear plasma into better context, the rank order concentrations of the first 50 individual OCs chemicals, both neutral and phenolic, was determined in the combined Resolute Bay/Svalbard data set. Out of the 10 highest OCs in plasma, six are phenolic compounds, three are PCBs, and one is oxychlordane. These six phenolic compounds, including 4-OH-HpCS, constituted 42% by weight of all OCs in polar bear plasma. Of the remaining 40 compounds, nine are chlordane-related compounds, 17 are PCBs, five are OH-PCBs, and the rest are chlorobenzenes, DDTs, HCHs and PCP.

Patterns of accumulation of these metabolites and their precursors were studied in ringed seal, and compared to those in polar bear to determine the potential for bioaccumulation of hydroxy metabolites of PCBs and octachlorostyrene (OCS). Concentrations of ΣOH-PCB were 1,000 times lower in ringed seal than in polar bear plasma, whereas ΣPCB were only 2 times lower. Considering that most of the hydroxy metabolites in ringed seal are probably in blood, and at low concentrations, it would seem that the potential for the polar bear to bioaccumulate OH-PCBs and 4-OH-HpCS is very low.

$MeSO_2$-PCB and -4,4′-DDE metabolites were examined in the tissues of polar bears shot in 1993 in Resolute Bay (Norstrom, 1997; Letcher *et al.*, 1996). Concentrations of the Σ$MeSO_2$-PCB were highest in liver (3049 ± 1290 ng/g lw) and were 1/9 the concentrations of ΣPCB. In fat, testes, lung and brain Σ$MeSO_2$-PCB had 2, 9, 13 and 60 times lower levels, respectively, than liver on a lipid weight basis. The major congeners in all tissues were 3- and 4-$MeSO_2$-CB87 and 3- and 4-$MeSO_2$-CB101. $MeSO_2$-PCB uptake from seal appears to be the most important source of $MeSO_2$-PCBs in bears. 3-$MeSO_2$-4,4′-DDE concentrations (303 ± 85 ng/g, lw) in liver were nearly half those of DDE and were 126 to 337 times higher than found in testes, adipose and lung. The highly asymmetric tissue distribution of Σ$MeSO_2$-4,4′-DDE may be due to several factors, such as the liver being the site of formation or because of highly selective binding in liver cells. It is not possible to sort out the relative importance of $MeSO_2$-DDE bioaccumulation versus formation in the bear based on adipose tissue concentrations.

$MeSO_2$-PCBs are efficiently transferred from polar bear females to cubs via milk, resulting in concentrations that are about 3 times higher in the cubs than their mothers, compared to about 2 times higher for PCBs. The 3-$MeSO_2$-4,4,′-DDE was not preferentially transferred to cubs. Cub survival is emerging as one of the major issues in ecotoxicology of polar bears; it is usually assumed that PCBs are the cause. However, Σ$MeSO_2$-PCB are present at 8–10% of ΣPCB in cubs, and their involvement should be considered in any future studies.

3.3.4 Arctic marine food web studies

A number of studies have been carried out on the food web transfer of OCs and metals in marine ecosystems since the first CACAR. These studies incorporated a larger number of species and trophic levels than was previously available for a single Arctic marine food web and incorporated stable isotopes of nitrogen to discern trophic position. These recent studies provide an advantage over food web relationships for OCs developed for

the original CACAR assessment in that all samples were collected at the same time and in the same region, and the analytical methods were consistent for all samples.

3.3.4.1 Trophic transfer of metals in marine food webs

Mercury in the Resolute Bay food web

Atwell *et al.* (1998) examined the concentration of total mercury in tissues from 27 species from the Arctic marine food web of Lancaster Sound, Northwest Territories. Samples ranged from particulate organic matter through polar bears. $\delta^{15}N$ values showed that total mercury in muscle biomagnified in this food web. Polar bears were a notable exception, having a lower mean Hg concentration than their main prey, ringed seals. This is likely due to the fact that the bears tend to eat only the blubber of seals, which is lower in mercury concentrations than muscle or liver (Atwell *et al.*, 1998). Most vertebrates showed greater variance than invertebrates, and there was a trend in seabirds toward increased variability with trophic position. No evidence of bioaccumulation of mercury with age was found in clams (*Mya truncata*) or ringed seals within species.

Metals in the food web of northern Baffin Bay

Metals and methyl-mercury (MeHg) were recently measured in mixed zooplankton, *Calanus hyperboreus* (a herbaceous copepod), eight species of seabirds, and ringed seals from the North Water Polynya to examine the food web dynamics of a range of metals in an Arctic marine food web (Fisk, 2002c). The North Water Polynya is located in northern Baffin Bay and is the largest and most productive polynya in the Canadian Arctic. Polynyas are areas of open water, often surrounded by sea ice, which persist throughout the winter in polar seas. All samples were collected in 1998 and 1999, although samples were only analyzed in late 2001 and not all data are available yet.

Concentrations of Hg and MeHg were positively correlated with trophic level ($r^2 > 0.85$), based on $\delta^{15}N$, demonstrating that these metals biomagnify (Figure 3.3.18). Atwell *et al.* (1998) reported a slope of 0.2 for the log mercury concentration-$\delta^{15}N$ relationship for the Resolute Bay food web that is very close to the slope of 0.21 found in this work. This would suggest that the rate of trophic transfer of mercury is similar throughout marine food webs of the Arctic. The log methylmercury concentration-$\delta^{15}N$ relationship slope of 0.28 found in this project suggests that methylmercury accounts for a

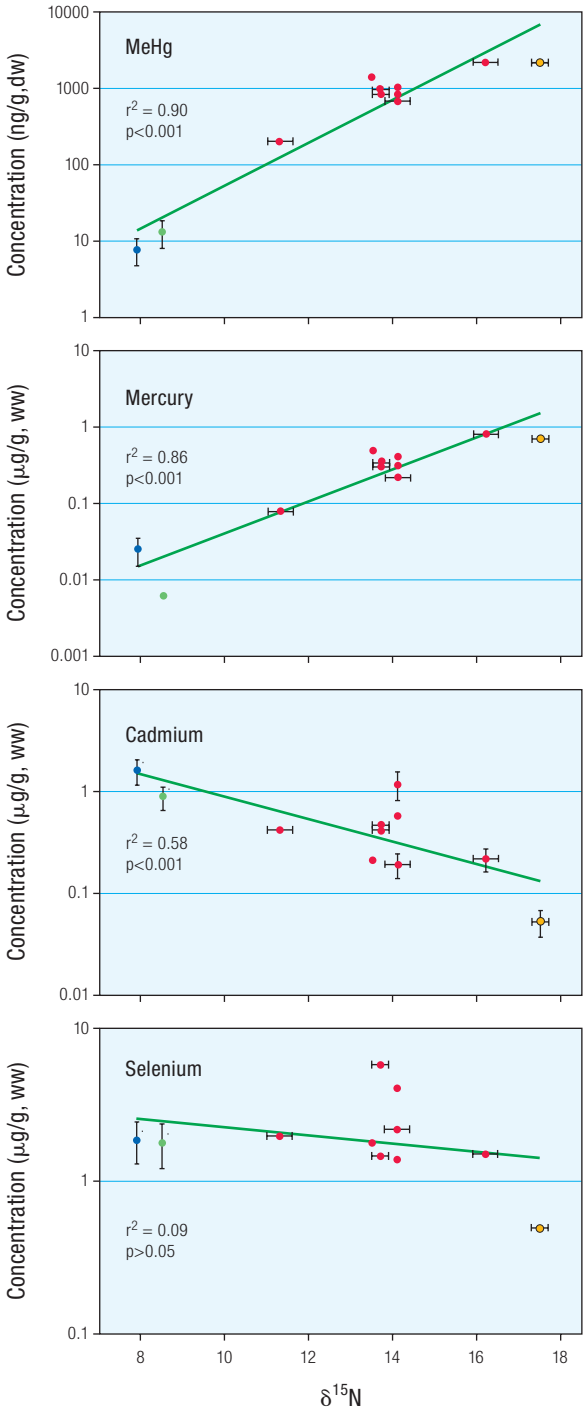

FIGURE 3.3.18

Relationships between metal and methylmercury concentrations and $\delta^{15}N$ in the marine food web of northern Baffin Bay (Fisk, 2002c). Metals and $\delta^{15}N$ determined in muscle tissue of seabirds and ringed seals and whole body for zooplankton. Each point is the mean ± 1 SE. Linear regressions are green lines, blue circle is *Calanus hyperboreus*, green circle is mixed zooplankton, red circles are seabirds, and yellow circle is ringed seal.

large percentage of the biomagnification of mercury in marine food webs.

Arsenic, cadmium, lead and selenium were not positively related to trophic level, in fact, selenium and cadmium decreased with trophic level suggesting a "food web dilution" (Figure 3.3.18). Arctic zooplankton have been shown to accumulate high levels of cadmium.

3.3.4.2 Trophic transfer of organochlorines in marine food webs

Organochlorines in the food web of northern Baffin Bay

OCs and stable isotopes of nitrogen ($\delta^{15}N$) were measured in zooplankton (six species), a benthic invertebrate (*Anonyx nugax*), Arctic cod (*Boreogadus saida*), seabirds (six species) and ringed seals (*Phoca hispida*) collected in 1998 in the North Water Polynya, located in northern Baffin Bay, to examine the effects of biological and chemical factors on trophic transfer of OCs in an Arctic marine food web (Fisk *et al.*, 2001a). These samples are the same or similar to those used in the analysis of metals described in Section 3.3.4.1 but were generated previously, include more species and samples, and have been more comprehensively studied. The trophic relationships derived from stable isotope analysis for the North Water Polynya (NOW) food web fell into the range expected based on stable isotope results for another Arctic polynya food web (Hobson *et al.*, 1995), with seabirds and ringed seal at the top and zooplankton species occupying lower trophic levels.

Strong positive relationships were found between recalcitrant OCs concentrations (lipid corrected) and trophic level based on stable isotopes of nitrogen, providing clear evidence of OC biomagnification in Arctic marine food webs (Figure 3.3.19 and Table 3.3.2). Food web magnification factors (FWMF) determined from the slope of POP-trophic level relationships in this work are in good agreement with values obtained for food webs from temperate and Arctic ecosystems involving marine birds and mammals (Jarman *et al.*, 1996; Norstrom, 1994).

Biomagnification factor (BMFs = lipid-adjusted concentration in predator/lipid-adjusted concentration in prey) values for the NOW food web are summarized in Table 3.3.2. BMFs were corrected for trophic level differences based on stable isotopes because many of these species have varied diets, and for many of these comparisons the predator was not a full trophic level above the prey based on $\delta^{15}N$ values (Fisk *et al.*, 2001a). Generally, PCBs and *p,p'*-DDE had highest BMFs, followed by chlordanes and HCBz. Compared to other compound classes, the BMFs for HCHs were low, reflecting the low octanol-water partitioning coefficient and greater susceptibility to metabolization. BMFs determined for amphipods that consume copepods were very high. Fisk *et al.* (2001a), however, suggested that the NOW food web BMFs were not realistic because concentrations of OCs in zooplankton may be controlled by OC concentrations in water and not prey and that the large size of the amphipods may drive the higher concentrations in the amphipods. BMFs for Arctic cod/amphipod were slightly below and above one but are in the range of

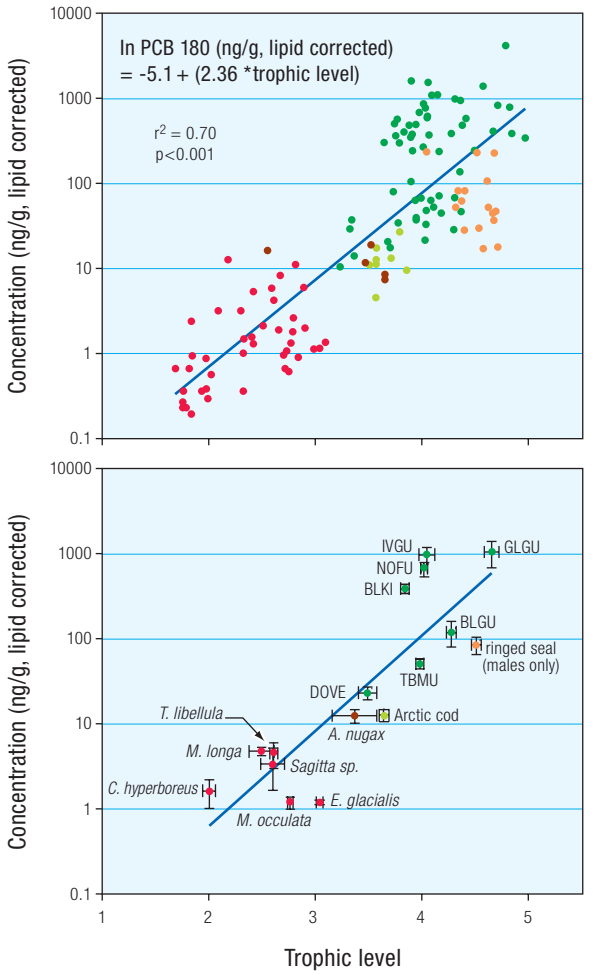

FIGURE 3.3.19

PCB 180 concentration (ng/g, lipid corrected) — trophic level relationships for the North Water (NOW) Polynya marine food web (Fisk *et al.*, 2001a). The top graph contains all data points and the bottom graph contains mean (± SE) values for each species. Lines are linear regression. Trophic level based on $\delta^{15}N$. Symbols: Red circle — pelagic zooplankton; brown circle — benthic amphipods; light green circle — Arctic cod; orange circle — ringed seals; dark green circle — seabirds.

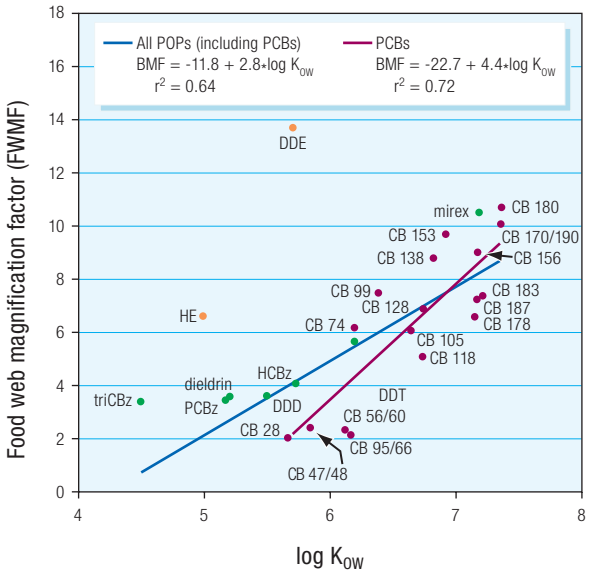

FIGURE 3.3.20

Relationship between food web magnification factors (FWMF) and log K_{ow} of recalcitrant POPs in the North Water (NOW) Polynya marine food web (from Fisk *et al.*, 2001a). FWMFs were determined from the relationship between concentration (lipid corrected) and trophic level.

BMFs reported for similar sized fish in laboratory experiments (Fisk *et al.*, 1998) and field observations (Rasmussen *et al.*, 1990). BMFs calculated for NOW ringed seals (Table 3.3.2) were slightly lower but within the range of those reported for male ringed seals from the east central Canadian Arctic (Muir *et al.*, 1988). The BMFs of the seabirds appear to be related to scavenging of marine mammals and/or migration to more contaminated regions. The highest BMFs were found in scavenging seabird species (glaucous gulls, ivory gulls and northern fulmars) and those that migrate to more contaminated southern regions (black-legged kittiwakes and glaucous gulls).

One of the most striking differences in BMFs was between poikilotherms (fish) and homeotherms (seabirds and mammals) (Table 3.3.2). Large differences in BMFs between poikilotherms and homeotherms were first demonstrated in herring gulls and salmon for Lake Ontario (Braune and Norstrom, 1989). Greater BMFs, and hence exposure to OCs in homeotherms have been attributed to their greater energy requirements and feeding rates (Braune and Norstrom, 1989; Fisk *et al.*, 2001a).

A strong relationship between food web magnification factors (FWMF), determined from concentration-trophic level relationships, and log K_{ow} were found for recalcitrant OCs (Figure 3.3.20). It is clear that increasing log K_{ow} results in greater trophic transfer of recalcitrant POPs in Arctic marine food webs. This relationship provides insight into the behaviour of a number of POPs. For example, DDE and heptachlor epoxide have values of FWMFs that are much greater than predicted based on the FWMF-log K_{ow} relationships. DDE has been well established as a metabolite of DDT formed in animals. These results suggest that a large percentage of the high concentrations of DDE in upper trophic level Arctic organisms are due to metabolic formation. Heptachlor epoxide, which is not in technical mixtures, is formed from heptachlor by photooxidation and in rat liver homogenate (Buser and Müller, 1993). The results from Fisk *et al.* (2001a) suggest that heptachlor epoxide is formed in upper trophic-level Arctic organisms, and may account for a large percentage of their concentrations.

BMFs of ΣOC groups vary considerably from those calculated for individual components of these groups (Fisk *et al.*, 2001a). This is due to a combination of differential susceptibility to biotransformation and variation in kinetics which results from different physical-chemical properties. For example, ΣHCH was observed to biomagnify in seabirds and throughout the entire food web, although neither α- nor γ-HCH were found to biomagnify; the biomagnification was mainly with β-HCH (Moisey *et al.*, 2001). These results suggest that caution should be exercised when using ΣOC groups across food webs.

TABLE 3.3.2 Biomagnification factors (BMFs) and food web magnification factors (FWMF) determined for key fauna in the marine food web of northern Baffin Bay (Fisk *et al.*, 2001a). BMFs based on lipid corrected concentrations, corrected to one full trophic level based on trophic levels derived from stable nitrogen isotopes.
TBMU – thick-billed murre, BLGU – black guillemot, BLKI – black-legged kittiwake, GLGU – glaucous gull

Predator/Prey site	HCB	ΣHCH	ΣCHL	*p′,p′*-DDE	ΣPCB
Amphipod/Copepod	3.8	4.5	26.5	16	4.6
Arctic cod/Amphipod	6.1	1.1	1.6	3.1	0.9
Seabirds					
TBMU/Arctic Cod	10.9	2.1	1.8	19	8.2
BLGU/Arctic Cod	5.0	3.5	4.0	18.5	8.9
BLKI/Arctic Cod	21.6	4.2	11.6	56	60.5
GLGU/Arctic Cod	6.7	5.2	80	49	28
Marine mammals					
Ringed Seal/Arctic Cod	0.5	2.0	2.4	7.0	5.5
FWMF	4.1	2.7	7.0	14	4.6

ITK/Scot Nickels

3.3.4.3 Trophic transfer of new and chiral chemicals in marine food webs

Concentrations of four possibly naturally produced organohalogens (collectively termed HDBPs) — 1,1′-dimethyl-3,3′,4-tribromo-4,5,5′-trichloro-2,2′-bipyrrole (DBP-Br_3Cl_3); 1,1′-dimethyl-3,3′,4,4′-tetrabromo-5,5′-dichloro-2,2′-bipyrrole (DBP-Br_4Cl_2); 1,1′-dimethyl-3,3′,4,4′,5-pentabromo-5′-chloro-2,2′-bipyrrole (DBP-Br_5Cl); and 1,1′-dimethyl-3,3′,4,4′,5,5′-hexabromo-2,2′-bipyrrole (DBP-Br_6) — were quantitated for the Arctic marine food web of northern Baffin Bay (Tittlemier *et al.*, 2002). The extracts used for this analysis were also used for the OC analysis of the food web in Section 3.3.4.2, but with fewer species. All HDBP congeners were found to significantly biomagnify, or increase in concentration with trophic level in the invertebrate–fish–seabird food web. None of the four HDBP congeners in ringed seals followed the general trend of increasing concentration with trophic level, which was likely due to the ability of the seals to metabolize HDBPs.

Chiral contaminants have the potential to identify differences in species biotransformation ability and provide insights into the fate of OCs in food webs (see Section 3.3.4.2). Concentrations of hexachlorocyclohexane (HCH) isomers (α, β and γ) and enantiomer fractions (EFs) of α-HCH were determined in the northern Baffin Bay marine food web (Moisey *et al.*, 2001), the same food web that was examined in Section 3.3.4.1. For invertebrates and fish, the biomagnification factors (BMFs) of the three isomers were > 1 and the proportion of each isomer and the EFs of α-HCH were similar to water, suggesting minimal biotransformation. Seabirds appear to readily metabolize γ- and α-HCH based on low BMFs for these isomers, high proportions of β-HCH (62–96%), and high EFs (0.65 to 0.97) for α-HCH. The α- and β-HCH isomers appear to be recalcitrant in ringed seals based on BMFs > 1 and near racemic EFs for α-HCH. The β isomer appears to be recalcitrant in all species examined and had an overall food web magnification factor of 3.9. EFs of α-HCH provided conclusive evidence that biotransformation was accounting for much of the HCH isomer patterns observed in the northern Baffin Bay food web.

3.3.4.4 Trophic transfer of organochlorine metabolites in marine food webs

Concentrations of $MeSO_2$-PCBs and -4,4′-DDEs and their precursor PCBs and 4,4′-DDE were compared in the Arctic cod–ringed seal–polar bear food chain from Resolute Bay to determine the relative importance of bioaccumulation from the food chain and formation by metabolism of precursors in the bear (Letcher *et al.*, 1996; Norstrom, 1997).

Overall, there was little difference in the PCB pattern in cod relative to the Aroclor standard (1242:1254:1260, 1:1:1); however, the PCB patterns changed noticeably in seal and bear, especially PCB congeners possessing hydrogens at the *meta-para* (3,4) position on one or more ring. *Meta-para* PCBs were present in seal blubber but notably absent in polar bear (< 0.05 ng/g lw). These PCBs included CB31, CB49, CB64, CB70, CB91, CB101, CB110, CB141, CB132, and CB174, all of which were also present in the form of their 3- and 4-$MeSO_2$-PCB metabolites in seal and bear. Metabolites of CB52 and CB95 were not found in the polar bear. These PCB congeners possess hydrogens at both *meta-para* positions, and may be metabolized to bis-$MeSO_2$-PCBs or to OH-PCBs.

3.3.5 Local sources of contaminants in the marine environment

From 1996–2001, additional studies and remediation of old military sites contaminated with PCBs were conducted in the Canadian Arctic. Detailed assessments of PCB contamination were conducted at two sites in the Eastern Canadian Arctic, Saglek Bay (58° 29′N, 62° 40′W) in northern Labrador and Resolution Island at the southeastern tip of Baffin Island.

At Saglek Bay high levels of ΣPCB (more than 50 μg/g dw in soils) were found at a former military radar site (ESG, 2002). The cleanup from 1997 to 1999 effectively removed the terrestrial sources of contamination to the surrounding land and to the marine environment of Saglek Bay.

ΣPCB concentrations in sediment cores from lakes near the former radar facility declined with increasing distance from the site (Betts-Piper, 2001). Four of eight lakes

within 5 km of the site had elevated levels (greater than100 ng/g dw) compared to background sites 30 km away (0.3–17 ng/g dw) or to other remote lakes in the Canadian Arctic (Muir *et al.*, 1996). The PCB congener profile in both the near-site and remote lakes had a clear Aroclor 1260 profile similar to the product used at the radar facility. Bright *et al.* (1995) and Dushenko *et al.* (1996) reported similar results from a study of 24 military radar sites in the Canadian Arctic. They concluded that the radar sites were acting as sources of contamination to nearby lakes via aerial redistribution. This was confirmed at Saglek Bay by the detailed study of Betts-Piper (2001).

Marine investigations were conducted to study the PCB contamination in Saglek Bay over three seasons from 1997 to 1999. These studies delineated PCB-contaminated sediments and quantified PCB uptake in the marine food chain, including benthic invertebrates, bottom-feeding fish, pelagic fish, marine mammals and seabirds. These studies also indicated that the contaminated sediments represented a source of PCBs to local wildlife. Inputs of PCBs to the marine environment occurred from a contaminated beach area associated with the original station. Sediment ΣPCB concentrations near this beach were 500–130,000 ng/g dw in the intertidal sediment, and 600–6,200 ng/g in Saglek Anchorage. Concentrations declined exponentially with distance, approaching background concentrations (< 2 ng/g) within 6–10 km (ESG, 1999; 2002). PCBs were also present in deepwater sediments of the bay, particularly immediately offshore and west of the contaminated beach (Figure 3.3.21).

The samples of invertebrates collected from the immediate vicinity of the contaminated beach contained PCBs at high concentrations, consistent with the concentrations measured in the beach sediment (ESG, 1998). Snails and scuds from the intertidal area had ΣPCB concentrations of 8,000 and 49,000 ng/g ww, respectively. The concentration in sea urchins from this area was slightly lower (5,200 ng/g ww). Invertebrate tissue contaminant concentrations generally return to near-background

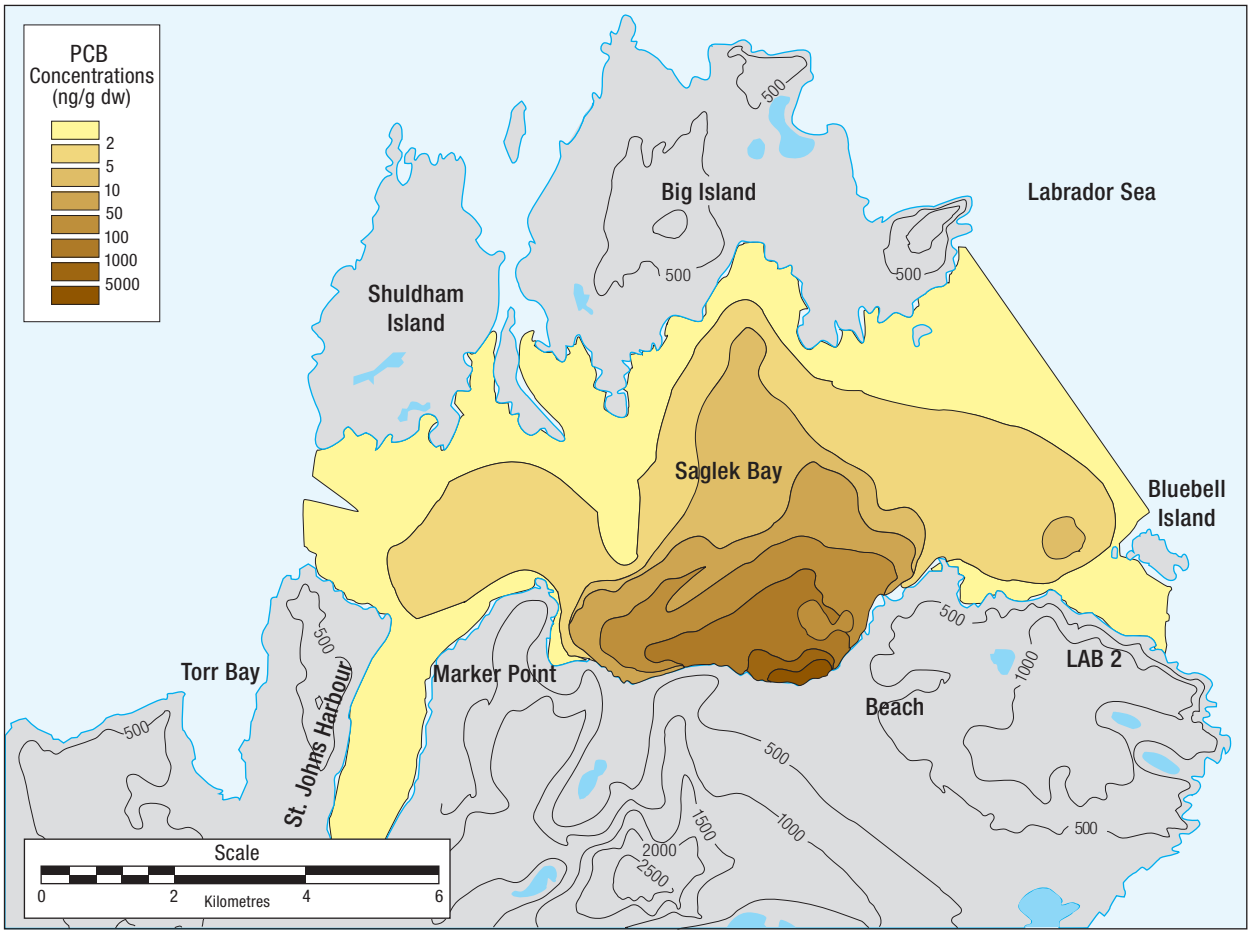

FIGURE 3.3.21

PCB contamination in marine sediments of Saglek Bay in northern Labrador based on 243 surface sediment sample results from 1997, 1998 and 1999 indicating the dispersion from a contaminated beach and anchorage area.

GNWT/RWED/Brett Elkin

concentrations within 7–10 km of the formerly contaminated beach.

ΣPCB concentrations were relatively high in sculpin (*Myoxocephalus scorpius*) sampled in the nearshore area close to the contaminated beach, exceeding 1,000 ng/g ww (whole body minus liver, Aroclor equivalent) in two-thirds of the samples and exceeding 10,000 ng/g in a few samples. Concentrations also exceeded 1,000 ng/g ww in some sculpin samples collected at sites up to 1.5 km east and 3 km west. PCB concentrations in muscle and liver of Arctic char (*Salvelinus alpinus*) did not vary with the proximity of the fish to the contaminated beach (ESG, 1998). PCB concentrations ranged from a low of 13 ng/g ww to a high of 66 ng/g ww (Aroclor equivalent), with both of these extremes measured in char collected at the Okak reference site (ESG, 1998). These were similar to results for PCBs in anadromous char from the Labrador coast (Muir *et al.*, 1999a; 2000a) and are consistent with the largely pelagic feeding of char.

ΣPCB concentrations in seals from the Saglek Bay area varied over a wide range, from a low of 500 ng/g ww in the adipose tissue of one 11-year-old seal, to a high of 9,400 ng/g ww in a 10-year-old seal. The result for this seal is exceptionally high, exceeding the results for the other seals by a factor of 4–6. It is also much higher than results reported for ringed seals from the community of Nain, further south on the Labrador coast, and elsewhere in the Arctic (see Section 4.3.3.2).

Black guillemots (*Cepphus grylle*) in Saglek Bay, Labrador also had elevated ΣPCB concentrations due to marine sediment contamination around the former military site. ΣPCB concentrations in liver of nestlings ranged from 15–46 ng/g ww in a reference group, 24–150 ng/g ww in a moderately exposed Islands group and 170–6,200 ng/g ww in the highly exposed Beach group. Biomarker responses were dose-dependent and in some cases sex-dependent (see Section 5.2.2).

Similar contamination by PCBs was investigated at Resolution Island (61°35′ N, 60°40′ W) at the south-eastern tip of Baffin Island in the eastern Canadian Arctic (ASU, 1997; 1998). Initial assessment of the site in the early 1990s showed it to be highly contaminated with PCBs (ESG, 1994). The site contained at least twenty buildings requiring demolition, along with a very large amount of visible debris, fuel tanks and many barrels. Buildings in the communications complex contained electrical equipment suspected of containing PCBs, and other buildings contained asbestos. The total volume of soil which was contaminated with ΣPCB at concentrations above 50,000 ng/g (exceeding Canadian Environmental Protection Act regulations) was estimated to be 5,000 m^3 with about 20,000 m^3 contaminated at the 1,000–50,000 ng/g level. Site remediation initially involved building barriers composed of various oil-absorbent booms across PCB leachate pathways to prevent further PCB migration. PCB-containing components and liquids from transformers were removed from the main station to the registered PCB storage facility on the site. An environmental assessment of contaminant migration to the marine environment assumed PCB movement was controlled by the barriers and aerial transport of PCBs was assumed to be insignificant. The assessment concluded that the site does not present unacceptable contaminant exposure conditions to humans or terrestrial and marine wildlife (Golder, 1997). Remediation work conducted from 1997–2001 involved excavation and removal of highly contaminated soils and debris from dump sites on the island (ASU, 2001). This material was stored in sealed containers on site for future disposal.

3.3.6 Overall assessment of contaminants in marine biota

A significant amount of data on contaminants in marine biota has been generated since the first CACAR assessment, particularly on levels in invertebrates and transfer within the marine food web. This has lead to a better understanding of levels and spatial trends.

The new results on persistent OCs in marine zooplankton and benthic invertebrates addresses an important data gap identified in the first assessment. Less hydrophobic OCs, in particular HCHs, are more prevalent compared to the other OCs in zooplankton and benthic invertebrates when contrasted to seabirds and marine mammals. This is due in part to the lower trophic level and limited biotransformation capacity of these invertebrates. Concentrations of OCs in zooplankton are determined to a large extent by water concentrations but dietary accumulation does play a role, particularly for more hydrophobic OCs (i.e., PCBs, DDT). Variability in OC concentrations between zooplankton species is limited, but larger variation is seen between benthic invertebrates. Scavenging benthic invertebrates, such as *A. nugax*, can achieve OC concentrations that are in the range of fish and lower trophic level seabirds.

Concentrations of HCH are higher in zooplankton from the western Arctic and Alaska Beaufort/Chukchi Sea area but most other OCs are slightly higher in zooplankton from the eastern Canadian Arctic. This is consistent with trends observed in higher trophic level organisms. Levels are generally lower than concentrations observed in the European Arctic, consistent with trends in higher trophic level organisms. There are no data on new chemicals in Arctic zooplankton or benthic invertebrates.

There are more limited data on metals in marine zooplankton and benthic invertebrates. Levels of mercury are low in these organisms compared to higher trophic level organisms, reflecting the ability of mercury to biomagnify in food webs. Levels of other metals, notably cadmium, can often be found at higher concentrations than what is observed in marine mammals and seabirds. There is limited data with which to assess spatial trends but results suggest little geographic variation of metals in marine zooplankton or benthic invertebrates. There is a trend for mercury and lead to decrease from south to north in mussels.

The limited amount of work on OCs and metals in marine fish show a wide range of concentrations that appear to be related to trophic position, age and size. Levels of OCs in sea-run Arctic char, Arctic cod and Greenland halibut are orders of magnitude lower than those observed in the large, long-lived and upper trophic level Greenland shark. Similar trends are seen for mercury. In general, levels of OCs are higher but metals are lower in marine fish compared with freshwater fish. OCs and mercury are influenced by the size and age of the fish. Cadmium and lead are near detection limits in most marine fish but As and Se are prevalent. Levels of OCs, in particular DDT, in Greenland sharks are among the highest seen for any Arctic organism. There are limited data for assessing spatial trends of OCs or metals in marine fish. Sea-run Arctic char from Labrador and Nunavik (Ungava Bay area) had very similar concentrations of all major OC groups.

A significant amount of data on OCs and metals in Arctic seabirds has been generated since the first CACAR was published. This work has provided information on spatial trends, interspecies differences, and factors that influence concentrations. Trophic level is an important variable influencing OC and Hg concentrations. Higher trophic level seabirds, such as the glaucous gull, have the highest concentrations. Scavenging, particularly of marine mammals, can also lead to high concentrations of mercury and OCs. Migration to more contaminated regions, generally to temperate zones, can also increase concentrations of OCs and metals.

A wide range of metal and OC concentrations were observed in Canadian Arctic seabirds. Seabirds such as the glaucous gull and northern fulmar can achieve concentrations that are among the highest of any Arctic animals. Therefore there is some potential for these chemicals to cause biological effects in these species. Biological effects related to PCBs have been reported for glaucous gulls from Svalbard, although levels in these birds are higher than those observed in the Canadian seabirds.

Examination of metals in long-tailed ducks provided the only information on spatial trends of metals. Cadmium was found to be higher in western ducks but lead was higher in eastern ducks. No significant trends were observed for the other metals. There is a lack of data on geographic trends of metals in seabirds. Previous work has found that spatial trends of metals in seabird eggs are minimal, although mercury tended to be higher in high Arctic seabird eggs. A recent study found that metals

ITK/Eric Loring

varied seasonally in thick-billed murres, although this is a preliminary assessment.

As with metals, there was no further work to assess geographic trends of OCs in Canadian Arctic seabirds. Results reported in the first CACAR showed that PCBs and HCBz were higher in seabirds from more southerly locations and concentrations increased from west to east, with the exception of HCH. High concentrations of PCDFs were found in the northern fulmar, at levels that are higher than found in most marine mammals. This warrants further study.

A fairly significant amount of information has been generated on new chemicals in seabirds. Toxaphene, PBDEs, chlorinated terphenyls, hexabromo biphenyls, halogenated dimethyl bipyrroles and TCP-methane were all found in seabird eggs or tissues. No chlorinated diphenyl ethers, TCP-methanol, or chlorinated naphthalenes were detected in seabird samples (detection limit ~2 ng/g ww).

A significant amount of OC data has been generated for marine mammals since the first CACAR assessment. This has resulted in a greater understanding of levels, geographic trends, and variables influencing concentrations.

GNWT/RWED/R. Schweinsburg

Data for OCs in ringed seals from a number of locations spanning the Canadian Arctic are available and show geographic trends in concentrations, although concentrations are generally within a factor of 2. Only qualitative comparisons of results were possible for this assessment; however, it is clear that, for female seals, levels of ΣPCB were slightly higher in the eastern Canadian Arctic (Hudson Bay, Hudson Strait, Labrador) compared to the western and central Canadian Arctic archipelago. ΣHCH concentrations are the only OC group to show the opposite trend. Age and sex have been shown to be important variables, OC concentrations increase with age in male seals but not in female seals. Data sets that include very old seals (> 20 years), however, have shown that OCs can increase with age in female seals and this could affect the interpretation. PCBs are the predominant OC found in ringed seals and toxaphene has also been established as a significant contaminant. Levels of OCs in Canadian ringed seals are intermediate between levels observed in Alaska and in eastern Greenland, Svalbard and northern Russia (deWit *et al.*, 2002; Muir *et al.*, 2000c).

Only a limited amount of new information was available on spatial trends of OCs in beluga and narwhal since the last assessment. Recent results from the Western Arctic beluga continue to show lower OC concentrations than those from East Hudson Bay/Hudson Strait or Pangnirtung. This is in general agreement with results for ringed seals and zooplankton. Combined results from the early 1990s (reported in Muir *et al.*, 1997) and recent analyses (Stern, 2001) suggests higher levels of most OCs in narwhal sampled at Pond Inlet (NE Baffin Island), than those from Broughton Island (eastern Baffin Island), or Grise Fiord (S. Ellesmere Island). The reason for this is unclear.

A number of new chemicals have been found in ringed seals and beluga. PBDEs have been measured in both of these species. Although levels are low compared to legacy OCs, PBDEs include a small number of individual chemicals and levels are increasing in the Arctic (see Section 4.3.1).

Since the first CACAR assessment, research on OCs in Canadian polar bears has focussed on the western Hudson Bay population. This work has examined temporal trends and biological effects. Past work on OCs in polar bears from the late 1980s and early 1990s showed some geographic trends in OCs in the Canadian Arctic but no spatial study has occurred since that time. Polar bears continue to be among the most contaminated Arctic animals and concerns about the effects of OCs remain. Recent work has found that OC metabolites, $MeSO_2$ and OH metabolites, are high in polar bears and

are of concern due to their endocrine disrupting potential. Recent results have also highlighted the importance of reproduction, sex and age in OC levels in polar bears.

There is limited work on new chemicals in polar bears. Recent work on Alaskan polar bears suggests that PFOS is a significant contaminant and it is likely that these chemicals are present in Canadian bears.

Recent work on Arctic fox has found relatively low levels of OCs in fat and liver with a few individuals having relatively high concentrations. For example, ΣPCB ranged from 110 to 14,600 ng/g lw in liver of Arctic foxes at Holman. There were no significant differences in the concentrations of major OC groups in the muscle of Arctic fox at Holman and Arviat.

Metal data for marine mammals is more limited than OC data but there is still a significant amount. A wide range of metal concentrations has been found in ringed seals. Mercury and selenium were related to age of the ringed seals, after normalizing for age minor geographic variation was found for mercury. Concentrations of mercury were slightly higher in ringed seals from Hudson Bay (Arviat) and Ungava Bay compared to more northerly populations. Cadmium has the opposite geographic trend. Major differences in mercury concentrations were seen in beluga. Mercury concentrations were three-fold higher in Mackenzie Bay animals than in beluga from the eastern Baffin and western Hudson Bay. There were major differences in mercury concentrations between the beluga in Mackenzie Bay and beluga at Paulatuk despite all being in the same Southern Beaufort Sea stock. The reason for this is not known, but it illustrates the large regional variation in mercury in beluga liver.

There have been a number of studies on the trophic transfer of OCs and metals through marine food webs. Mercury, methylmercury and most OCs were found to be highly correlated to trophic level, increasing with each step in the food web. Arsenic, cadmium, lead and selenium were not related to trophic level and cadmium was found to decrease with trophic level. This research has shown that warm-blooded animals bioaccumulate OCs to a greater extent than cold-blooded animals and that the chemical physical properties of OCs influence the magnitude of trophic transfer.

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GNWT/RWED/R. Popko

Temporal Trends of Contaminants in Biota

One of the major data gaps identified in the first CACAR assessment was the lack of temporal trend data sets for both metals and OCs (Muir *et al.*, 1997). One exception was the data set for OCs in Holman Island ringed seals that spanned from 1972–1991 (Addison and Smith, 1998). The number of sampling years was limited, however, and comparison of the data from the 1970s was problematic because of changes in analytical methodology. Bans on the use of many of the legacy OCs, e.g., PCBs and DDT, have been in place in North America and western Europe since the 1970s but only recently have agreements been signed that will completely eliminate the use of these chemicals throughout the world. Release of heavy metals, especially mercury, through coal combustion and other industrial and mining activities continues to be a major concern. The continued emission of legacy chemicals from use areas (e.g., toxaphene and DDT in agricultural soils; PCBs from buildings, waste disposal sites) in temperate regions, their transport in air and seawater, and the slower rate of transformation of organic chemicals in the Arctic make temporal trends and predictions about future levels of OCs and metals in biota an important issue. Wildlife, and in particular higher trophic level organisms that tend to have high levels of contaminants, are a significant component of the Inuit diet making temporal trend data sets particularly important information at the community level. As well, the discovery of new chemicals, some of which have been increasing in the Arctic in recent years (e.g., PBDEs), demonstrates the value of temporal trend data sets and the archiving of samples for future research.

One of the major accomplishments of the Northern Contaminants Program over the past six years has been the support of temporal trend studies. A number of studies were initiated during this time and because of the short time span have only limited ability to provide strong statistical trends. Other studies have used archived samples to generate new data sets that have a greater range of years. As well, a number of studies initiated before and summarized in the first CACAR have continued.

4.1 Temporal trends of contaminants in freshwater fish

Much of the early temporal trend data on OCs and heavy metals in fish in the Arctic were too limited to be scientifically credible because they are based on two or at most three sampling times. In addition, much of this is confounded by changes in analytical methodology (use of packed column gas chromatography in the studies from the 1970s) as well as variability due to age/size, or dietary and population shifts (Muir *et al.*, 1997; de March *et al.*, 1998). In comparison, temporal trend data for contaminants in Lake Ontario lake trout (Borgmann and Whittle, 1991) and in pike muscle from Storvindeln Sweden (de March *et al.*, 1998) are available over a 15- to 30-year period.

Freshwater organisms are exposed to atmospheric contaminants deposited on snow or directly onto lake surfaces or lake watersheds. Fish accumulate contaminants, metals and organometallics as they pass water over their gills and from their diet. It has been shown that rising water temperatures lead to increased metabolic rates and thus pumping of higher volumes of water across the gills, which in turn results in increased uptake of metals from the water. Unlike mammals, fish have limited capacity to degrade most OCs and metals and excrete many of these chemicals very slowly. Fish are therefore good indicators of freshwater contamination from OCs and metals.

Small lakes in the high Arctic are replenished annually with snowmelt runoff and direct precipitation, which represent significant fractions of their water budgets. Declining concentrations of OCs, or increasing levels of previously unstudied OCs, in air and precipitation should be reflected relatively quickly in changes in levels in food webs and top predator fish, compared to the vast marine environment. This is illustrated by the sedimentary record of OCs and mercury in small Arctic lakes (Muir *et al.*, 1996; Lockhart *et al.*, 1998).

4.1.1 Metals in freshwater fish

Fort Good Hope and Lake Laberge burbot

Burbot were collected from Fort Good Hope on the Mackenize River to examine temporal trends of metals during various years between 1985 and 1999 (Stern *et al.*, 2001a). Mean mercury concentrations in muscle from male burbot have increased by 36% over the 15-year time period from 1985 to 2000 (Figure 4.1.1). No significant correlation between length and Hg concentration was observed in muscle or liver for either sex. Arsenic levels in the 1988 male burbot liver samples were significantly higher than those from 1993, 1999 and 2000. Selenium concentrations in burbot from Fort Good Hope (Rampart Rapids) decreased 1.4- and 1.8-fold between 1985 and 1999, in the muscle and liver, respectively. However, levels of selenium were much higher in 2000. The length-adjusted mean selenium concentrations in liver were 1.625, 1.156, 0.917 and 1.520 µg/g for the 1985, 1988, 1999 and 2000 time points, respectively. Similarly, no change in mercury concentrations was observed in the liver of burbot from Lake Laberge between 1996 and 1999.

Resolute land-locked Arctic char

Land-locked char have been collected over a number of years from lakes near the community of Qausuittuq (Resolute) in the Canadian High Arctic to examine temporal trends of metals (Muir *et al.*, 2000b; 2001a). Lakes near Qausuittuq are ideal sites for study of temporal trends of POPs and metals in fish. Many of the lakes, e.g., Char, Resolute, and Meretta, are well studied limnologically and the food chain in Char Lake has been well documented (Hobson and Welch, 1995). Char from Char Lake collected by H. Welch in 1993 have been previously analyzed for organochlorine contaminants and metals (Muir and Lockhart, 1996; Muir *et al.*, 1997). Statistical analysis showed that concentrations of mercury and most other elements in char muscle from Resolute and North lakes were normally distributed.

There has been no significant change in Hg concentrations in Arctic char from Resolute Lake during a nine-year period from 1992–2001, although mean concentrations in char from 2000/2001 were 20% lower than those from 1997 and 1993 (Figure 4.1.2). If a slow decline is occurring, however, it will take additional annual sampling to confirm this. In nearby Char Lake, higher levels of mercury were found in samples from 2000 than from 1999, but the fish collected in 2000 were longer and heavier (37 ± 3 cm; 329 ± 80 g) compared to those from 1999 (28 ± 19 cm; 201 ± 40 g), and may have been feeding at a higher trophic level. The limited number of char available from this lake makes comparison between years problematic.

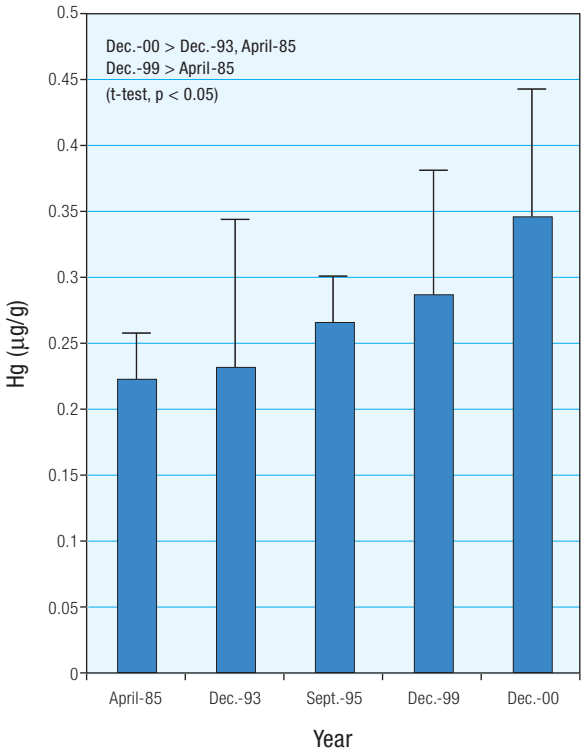

FIGURE 4.1.1

Mercury concentrations in Fort Good Hope burbot liver (Stern *et al.*, 2001a).

Mercury in lake trout in Yukon, NWT and Nunavut lakes

Lockhart (2002) investigated the temporal trends of mercury in lake trout from 18 lakes from Yukon, NWT and Nunavut where data were available from the 1970s, 1980s, and in some cases as recently as 2000. Analysis of covariance was used to adjust for fish length. There was no significant relationship of fish length and sampling year for any of the 17 lakes, thus permitting a simple comparison among sampling years using length adjusted data. The results are summarized in Annex Table 1. In general no increases in mercury were seen in lake trout from these lakes. An exception was Lake Belot where a significant increase was found between 1993 and 1999. In nearby Colville, on the other hand, mercury concentrations declined over the same period. The overall weight of evidence is that mercury concentrations have not increased in the past 10–25 years in NWT lake trout.

A significant decline in mercury in lake trout from Lake Laberge was observed (Annex Table 1), however, no significant change in Hg levels occurred in muscle of lake trout from another Yukon lake, Quiet Lake, between 1992 and 1999 (Stern *et al.*, 2000). These mercury levels are a concern, as 97% of all the trout muscle samples had Hg concentrations that exceeded the guideline level

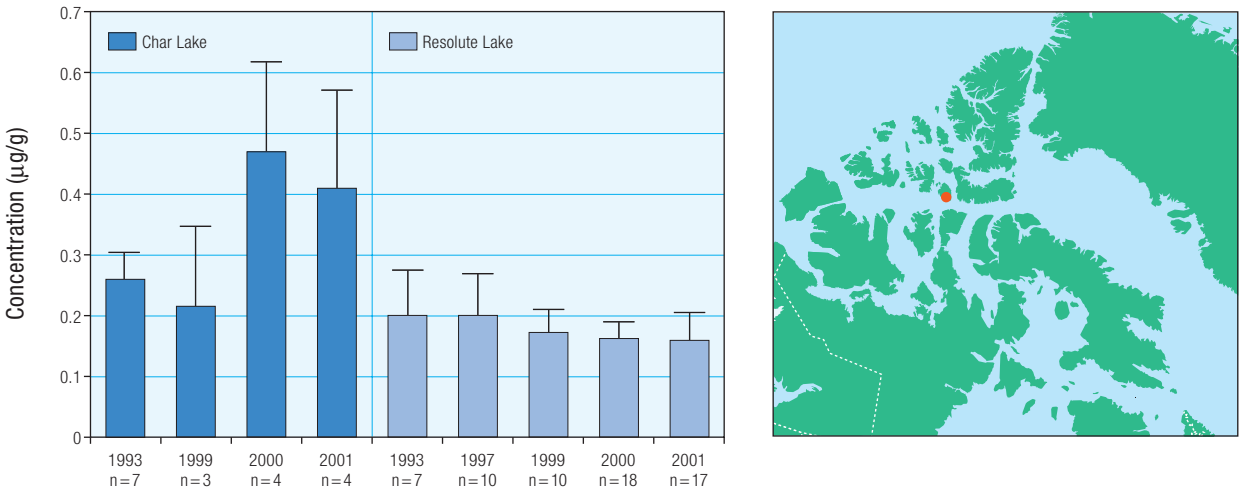

FIGURE 4.1.2

Mercury concentrations (µg/g, mean ± SE) in land-locked Arctic char from Char and Resolute Lakes (Muir *et al.*, 2000b; 2001a). Concentrations are corrected for length.

of 0.20 mg/g recommended for fish used for subsistence, 71% of the lake trout ≥ 500 mm in length had concentrations at or above the recommended guideline level of 0.50 mg/g.

Great Slave Lake

Great Slave Lake was selected as one of the sites for the long-term biomonitoring of contaminant trends in freshwater fish for a number of reasons.

1. In the mid-1990s, the NCP supported various studies of POPs in Great Slave Lake, including food web biomagnification studies.
2. Great Slave Lake is strongly influenced by the Slave River, formed by the confluence of the Peace and Athabasca Rivers. Increased development in the Peace-Athabasca drainage basins (oil sands, pulp and paper mills, agriculture, urban areas, etc.) may potentially affect an increase in contaminant loading to Great Slave Lake.
3. Studies conducted in the early 1990s as part of the Northern River Basins Study (Pastershank and Muir, 1996) showed elevated concentrations of PCBs in burbot liver along several reaches of the Peace and Athabasca rivers. Levels approached or exceeded those observed in the Slave River and Great Slave Lake. These higher concentrations may have been related to trophic feeding and/or to local PCB inputs.
4. Great Slave Lake is a major population centre in the NWT. Yellowknife is the territorial capital while Hay River is an important transportation hub with road and rail links to the south, and is a port serving the Mackenzie River, the outflow from Great Slave Lake. Fort Resolution and Lutsel K'e are two major communities located on the Great Slave Lake shoreline that make extensive use of its fish populations.
5. In contrast to most aquatic systems in the NWT, Great Slave Lake is well studied and the major features of its limnology and fisheries ecology are well understood.
6. The Great Slave Lake commercial fishery has been in existence since the early 1950s. It is a highly regulated fishery with fishing quotas established for some areas of the lake while other areas are closed to commercial fishing. While long-term records have been maintained on the Great Slave Lake fisheries, few studies have investigated contaminant concentrations in the fish.

Temporal trends of mercury in five species (burbot, lake trout, lake whitefish, walleye, and northern pike) from the 1970s to 1990s are shown in Figure 4.1.3. These data were assembled from a Fisheries and Oceans (Fish Inspection Service) database encompassing samples from the 1970s and late 1980s along with results from studies in the 1990s (Lockhart, 2002; Evans and Muir, 2001). Mean concentrations of mercury in walleye declined significantly ($P < 0.1$) from 1987–2001 with a half-life of 11 ± 3 years (Table 4.1.1). Mercury concentrations in northern pike also declined, if results for late 1980s to 2001 were used, with a half-life of 24 ± 6 years. No significant trends in mercury were found for lake whitefish or burbot. Mercury in lake trout muscle had the opposite trend from the other species, actually increasing slowly from 1978–2001 with a doubling time of 86 ± 18 years if all data were used or 33 ± 10 years if the results for 1988–2001 were used (Table 4.1.1). The variation of temporal trends of mercury among five species suggests

TABLE 4.1.1 Half-lives or doubling times for mercury in four species of fish from Great Slave Lake

Species	Time period	N	r^2	Half-life ± SE (yrs)	Doubling time ± SE (yrs)	Prob > F
Walleye	1987–1998	79	0.18	11 ± 3	–	< 0.001
Lake trout	1979–2001	176	0.11	–	87 ± 18	< 0.001
Lake trout	1988–2001	152	0.06	–	33 ± 10	0.0021
Burbot[1]	1975–2001	84	0.02	54 ± 45	–	0.24
N. Pike	1988–2001	143	0.12	24 ± 6	–	< 0.001

[1]Trend for burbot is not statistically significant.

that ecological changes in Great Slave Lake either from climate or more directly from commercial fishing may be influencing the observed levels through changes in fish diets.

4.1.2 Organochlorines in freshwater fish

Lake Laberge

Monitoring activities in 1991 revealed that lake trout muscle and burbot livers from Lake Laberge contained elevated concentrations of toxaphene, which prompted a closure of the fishery. Kidd *et al.* (1993; 1995) attributed the elevated concentrations to heavy fishing pressure that altered the food web structure of the lake. DeGraff and Mychasiw (1994) suggested that rehabilitation of the lake trout stock in Lake Laberge, by reducing fishing pressure, should restore the predator/prey balance to a level comparable with other regional lakes, and promote a decline in contaminant levels.

A study was initiated to evaluate potential changes in OC levels in Lake Laberge burbot and lake trout (Stern *et al.*, 2000). Combining data from this study with data from previous studies (Kidd *et al.*, 1998; Muir *et al.*, 1990a) allows an examination of temporal trends spanning almost 10 years. Levels of OCs in the lake trout have declined nearly four-fold between 1996 and 1999, although no effort was made to control for size and growth rate of the fish (Figure 4.1.4). This decline contrasts with the burbot collected in Lake Laberge, which showed no decline in OC concentrations between 1990 and 1999 (Figure 4.1.4). It would appear very unlikely that the rapid decline in the OC levels in the lake trout is due to a reduction in loadings to the lake, although Lake Laberge is a unique system with past OC use in the lake and a long food chain (Kidd *et al.*, 1995). Such drastic changes in OC levels may be due to changes in human fishing pressure and a subsequent change in the carbon and contaminant dynamics within the lake. Studies continue on the Lake Laberge system (Stern *et al.*, 2000).

Fort Good Hope burbot

Concentrations of OCs were determined for burbot liver collected from Fort Good Hope in 1988 and 1999 to examine temporal trends (Stern *et al.*, 2001a). An examination of only the 1988 and 1999 data suggest significant declines for most OCs (Stern *et al.*, 2001a). Significant declines, 2.0 and 3.1-fold, were observed for

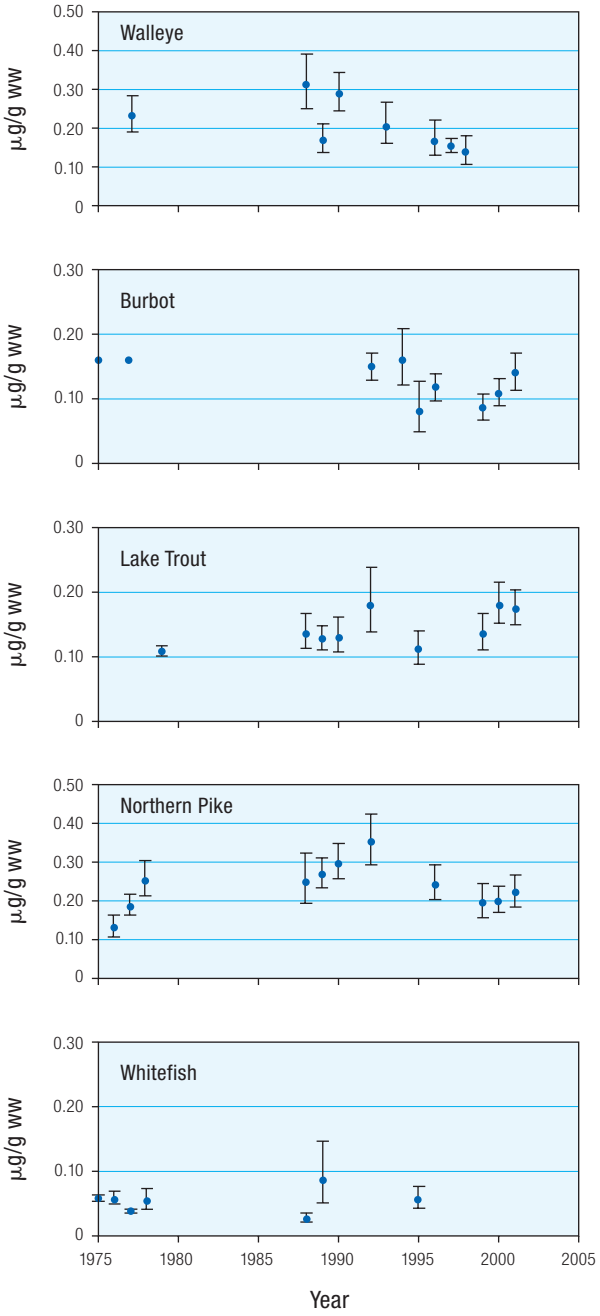

FIGURE 4.1.3
Temporal trends in mercury concentrations (µg/g, geometric mean ± 95% confidence limits) in muscle of five fish species from Great Slave Lake (1970s–1990s).

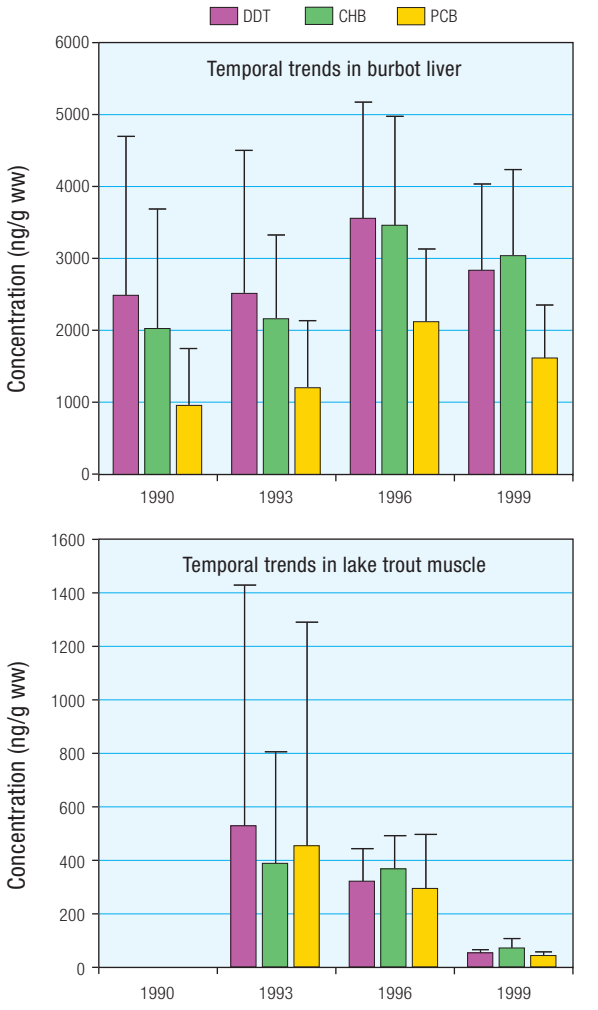

FIGURE 4.1.4

Concentrations, mean ± 1 SD, of DDT, PCBs and toxaphene (CHB) in liver of burbot and muscle of lake trout from Lake Laberge, Yukon (1990–1999).

both α- and γ-HCH over this 11-year time period. β-HCH concentrations were below the detection limit in all samples. Interestingly, the α/γ-HCH ratio has increased from 4.5 to 6.9 which is opposite to what one might have expected based on the decreased use of the technical mixture and the corresponding increase in the use of lindane since the early 1990s (Li *et al.*, 1998). ΣDDT concentrations did not change over this 11-year time interval; however, a 1.8-fold decline and a 2.2-fold increase in the concentration of *p,p′*-DDT and its metabolite *p,p′*-DDE (not age adjusted), respectively, was observed. These changes translated into a significant increase in the *p,p′*-DDE/ΣDDT ratio from 0.39 to 0.60 and suggests "old" rather than recent DDT inputs. Overall, a 1.7-fold decline in the lipid adjusted mean concentrations of ΣCHL was observed. Oxychlordane, the principal metabolite of *cis*- and *trans*-chlordane, and second only to *trans*-nonachlor as the most abundant chlordane-related residue in the Fort Good Hope burbot liver, did not change significantly over this 11-year period. The decreasing *t/c*-CHL ratio suggests "old" rather than recent chlordane inputs. For *t*-nonachlor, lipid-year interactions (lipid*year) were significant ($p > 0.05$) and as a result, ANCOVA could not be used to correct for the observed effects from lipid (homogeneity of the slope between percent lipid and *t*-nonachlor concentration). While heptachlor concentrations in the burbot liver have decreased 2.3-fold over this 11-year time period, the level of its oxidation product, heptachlor epoxide, increased by 1.9-fold. ΣTri-PCB concentrations have increased 2.9-fold, while all other PCB homologue groups have either declined in concentration or did not change significantly. ΣPCB levels have declined 1.3-fold. No significant differences were observed for *n*PCB 77, 81 or 169. CB126 decreased 1.8-fold. Toxaphene and dieldrin concentrations have decreased 1.7- and 1.5-fold, respectively.

Combining the OC data generated for burbot by Stern *et al.* (2001a) with previously published data for burbot at Fort Good Hope (Muir *et al.*, 1990a; 1997), which includes the years 1986, 1988 and 1994, provides a longer-term data set. The results of 1988 reported by Stern *et al.* (2000) are much lower than those reported previously (Muir *et al.*, 1990a) and have been excluded. Slow declines in all of the major OC groups and toxaphene are observed in the Fort Good Hope burbot, although the rate of change varies with the chemical and the year (Figure 4.1.5).

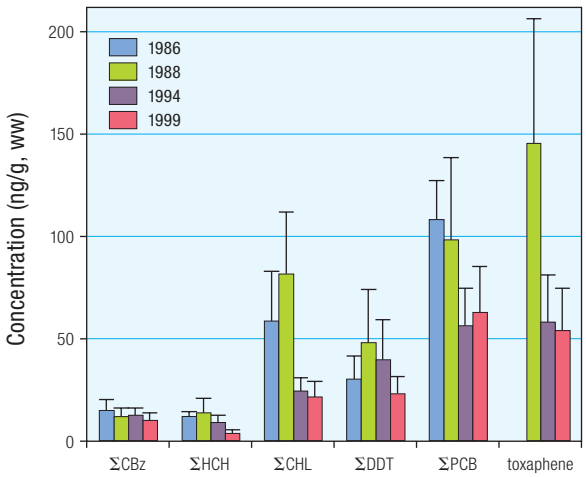

FIGURE 4.1.5

Concentrations, mean ± 1 SD, of major organochlorine groups in liver of burbot at Fort Good Hope, NWT (1986–1999). Data are combined from Muir *et al.* (1990a) and Muir *et al.* (1997) for 1986, 1998 and 1994, and from Stern *et al.* (2001a) for 1999.

Organochlorines in Resolute land-locked Arctic char

Long-term studies of contaminant levels in land-locked Arctic char from lakes in the area of Qausuittuq (Resolute) and on Devon and Somerset Islands are underway. As discussed in Section 4.1.1, char from lakes near or accessible by short air flights from Qausuittuq, including Resolute Lake, Char Lake, Amituk Lake and North Lake have been previously analyzed for mercury and/or OCs (Muir and Lockhart, 1996; Muir *et al.*, 1997). Levels of PCBs and total DDT in char muscle (plus skin) from Char Lake in 1993 were relatively high compared to other land-locked or sea-run char (Muir *et al.*, 1997).

Results for persistent organochlorines measured in char from Resolute, Char and Amituk Lakes collected between 1997 and 2001 showed that PCBs were the major organochlorines present (Muir *et al.*, 2000b; 2001a). Results for PCBs, ΣDDT and ΣCHL for Resolute Lake char were skewed indicating that a few individual fish tended to have higher levels. Log transformation reduced skewness and the transformed, lipid weight results were used to compare between years (Figure 4.1.6). No significant difference in mean PCB concentrations were found from 1997–2001 for Resolute Lake char. The small numbers of char from Char Lake and Amituk Lake precluded any detailed statistical analysis. ΣPCB levels in char from Char Lake were lower in the four fish from 2000 compared to those collected in 1999 and much lower compared with results from char collected in 1993. The samples sizes are very small, however, and piscivorous char may have been selected for analysis in 1993. In Amituk Lake, ΣPCB

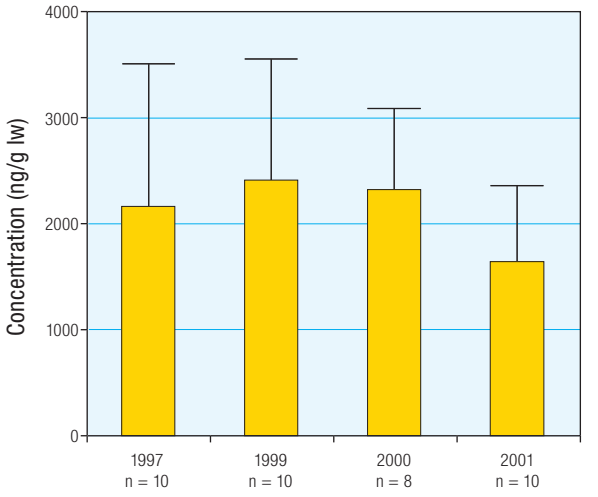

FIGURE 4.1.6

Concentrations, mean ± 95% confidence limits, of total PCBs in lipid of land-locked Arctic char from Resolute Lake near Qausuittuq, Nunavut (1997–2001).

levels in char from 1993 (73 ± 40 ng/g ww) were similar to results for char collected in 2001 (108 ± 49 ng/g ww). ΣHCH concentrations, which are less likely to be affected by trophic level of the char, declined by about three-fold in Amituk Lake and about five-fold in Char Lake over a 7–8 year period (Annex Table 2).

Great Slave Lake monitoring

OC data spanning eight years (1993–2001) in burbot and lake trout are also available for Great Slave Lake (Evans *et al.*, 2000; 2001). No clear trend emerges for ΣPCB in either species using lipid-normalized data, but ΣCHL, ΣDDT and toxaphene do appear to be declining slowly (Annex Table 2). These conclusions should be used with caution because no effort has been made to correct for biological variation such as size or growth rate.

4.1.3 Overall assessment of temporal trends of contaminants in freshwater fish

From the data sets that are available for Arctic freshwater fish in Canada it is apparent that legacy OCs are declining. The picture for metals is less clear but there is evidence that mercury is increasing in some freshwater fish populations.

Mercury levels have increased significantly in Mackenzie River burbot and similar increases have been seen in beluga whales at the mouth of the Mackenize River. It has been suggested that increases in temperatures in this region associated with climate change have liberated mercury and made it more bioavailable. This would suggest that there is no increase of mercury loading to this region but that other anthropogenic effects are influencing metal levels. Levels of As and Se, however, have not increased in these burbot. There have also been no increases in metal levels in fish from Yukon Lakes or Great Slave Lake or in Arctic char from high Arctic lakes. Monitoring and research on temporal trends of metals, in particular mercury, in freshwater fish is warranted.

All freshwater fish temporal trends studies have reported decreases in legacy OC concentrations. This is consistent with many temporal trends observed in marine organisms from the Canadian Arctic and in freshwater fish studies in Sweden. These decreasing trends and low concern about present levels of OCs would suggest that there is limited need for continued monitoring of OCs in the freshwater environment of Arctic Canada. There is solid evidence, however, that some new chemicals, many of which are still heavily used such as PBDEs, are increasing in freshwater fish. Without monitoring programs these chemicals would not likely have been measured and found.

4.2 Temporal trends of contaminants in seabirds

Eggs of seabirds have been used to monitor contamination of the Canadian Arctic marine environment since 1975 (Noble and Elliott, 1986; Nettleship and Peakall, 1987; Noble, 1990; Muir et al., 1999b). Retrospective analyses of OCs, mercury and selenium in archived Arctic seabird eggs from Prince Leopold Island in the Canadian High Arctic (between 1975 and 1998) have provided one of the most comprehensive contaminant temporal trend data set available for the Canadian Arctic (Braune et al., 2001a; Braune, 2001a; Braune, 2002). Eggs from three species of seabirds, thick-billed murres, northern fulmars and black-legged kittiwakes have been used for this work. Dioxins, furans and non-*ortho* PCBs and new chemicals were also examined in the archived liver samples from the same species.

Temporal trends of organochlorines in seabird eggs

At the time of egg formation, organochlorine compounds are transferred along with fat to the eggs (Mineau *et al.*, 1984). Contaminant burdens in the egg reflect residues assimilated over a long time period by the female and, particularly in migratory species, may integrate exposure from a number of different locations (Hebert, 1998; Monteiro *et al.*, 1999).

With the exception of ΣHCH, OCs in Arctic seabird eggs have shown declines or, in some cases, no significant change in levels between 1975 and 1998 (Braune *et al.*, 2001a). Levels of ΣPCB and ΣDDT (Figure 4.2.1) as well as ΣCBz decreased significantly in eggs of all three species while ΣCHL, dieldrin and mirex levels decreased in kittiwake eggs only. Kittiwakes, whose migration pattern would have historically brought them into closer contact with industrial sources of contaminants such as PCBs in the more temperate latitudes, showed the most dramatic declines through to 1998. The significant declines in concentrations of ΣPCB and ΣDDT in this study have also been observed in seabirds from other areas including the Baltic Sea (Olsson and Reutergårdh, 1986; Andersson *et al.*, 1988; Bignert *et al.*, 1995), the Barents Sea (Barrett *et al.*, 1996), and the Great Lakes (Hebert *et al.*, 1997). The only organochlorine compound in this study for which a significant increase in concentrations was seen for ΣHCH, particularly for β-HCH in murres and fulmars.

Stable-nitrogen isotope analyses (δ15N) indicate that the temporal trends observed in organochlorine and trace metal concentrations in seabird eggs were not the result of shifts in trophic level over time (Braune *et al.*, 2002). More likely, the trends reflect changes in contaminant

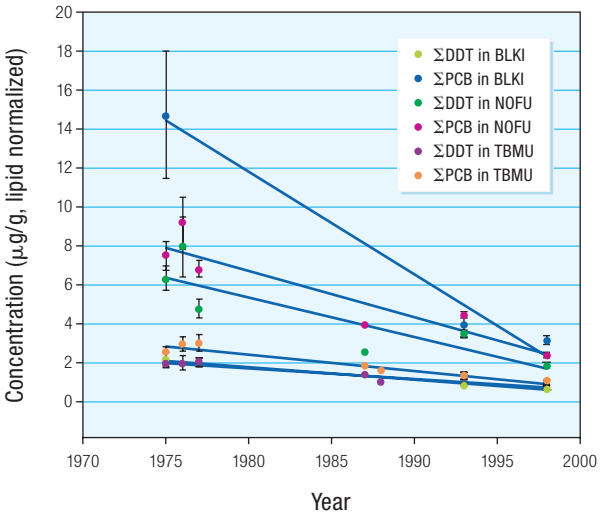

FIGURE 4.2.1

Concentrations of ΣDDT and ΣPCB in seabird eggs collected between 1975–1998 on Prince Leopold Island (Braune 2000; 2001a). BLKI = black-legged kittiwake, NOFU = northern fulmar and TBMU = thick-billed murre.

deposition into the various marine environments that these birds occupy throughout the year as well as the toxicokinetics of each contaminant as it is transported through the food chain.

Temporal trends of dioxins, furans, non-ortho PCBs and toxaphene in seabird livers

Dioxins, furans and toxaphene have also been measured in Canadian Arctic seabirds for almost 20 years (Braune, 2000; 2001a; 2002) providing a temporal trends data set. Liver samples of thick-billed murres, northern fulmars and black-legged kittiwakes collected in 1975 and 1993 from Prince Leopold Island in Lancaster Sound, were analyzed for polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), non-*ortho* PCBs (noPCBs) and toxaphene (Annex Table 13).

Concentrations of total PCDDs, PCDFs and non-*ortho* PCBs decreased in kittiwakes from 1975–1993. Concentrations of total PCDDs and PCDFs in fulmars decreased and non-*ortho* PCBs increased from 1975–1993, and in murre liver samples, concentrations of total PCDDs and PCDFs increased and non-*ortho* PCBs decreased from 1975–1993. PCB 126 was the predominant non-*ortho* PCB congener in all years and all Arctic seabird samples analyzed. The non-*ortho* PCB fraction of the TEQ decreased and the PCDF fraction increased from 1975–1993 in kittiwake and murre livers. In fulmars, the PCDFs constituted the dominant fraction in both 1975 and 1993 livers. Calculated TEQ values were highest in northern fulmars both in 1975 and 1993 (Figure 4.2.2).

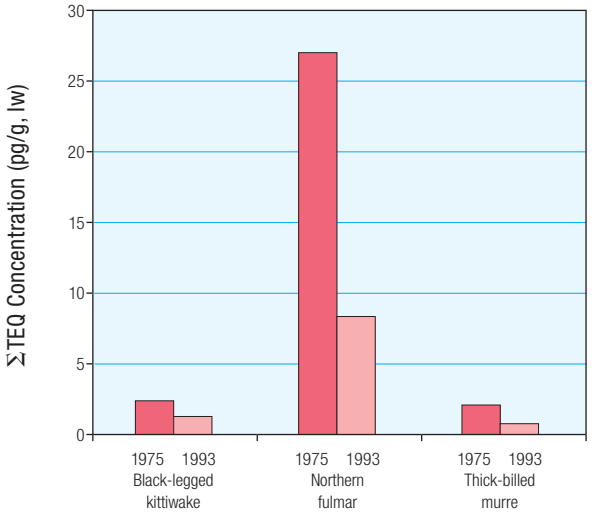

FIGURE 4.2.2

Toxic equivalent (TEQ) values in seabird livers collected in 1975 and 1993 on Prince Leopold Island (Braune 2002).

Toxaphene was detected in every seabird sample analyzed except for the pool of kittiwake livers from 1975. The highest estimated level of toxaphene was found in the recently collected samples suggesting that exposure to toxaphene has increased in seabirds between 1975 and 1993.

Temporal trends of mercury and selenium in seabird eggs

Total mercury concentrations have almost doubled between 1975 and 1998 in eggs of thick-billed murres, and a 50% increase in levels was observed in northern fulmars (Figure 4.2.3). Data for marine mammals (Wagemann *et al.*,, 1996), seabird feathers (Thompson *et al.*, 1992), and lake sediments (Lockhart *et al.*, 1995; Mannio *et al.*, 1995) also indicate that mercury levels have increased in the North over the last few decades. Concentrations of total mercury in black-legged kittiwake eggs did not change significantly through time. This may be partially because of kittiwakes overwintering at lower latitudes where mercury concentrations are now lower than they once were because of a reduction of mercury released from point sources. The murres and fulmars overwinter in northern waters which have not experienced the decline in mercury. Selenium levels decreased significantly in northern fulmar eggs but not in eggs of thick-billed murres or black-legged kittiwakes.

4.2.1 Conclusions on temporal trends of contaminants in seabirds

The continued monitoring of OCs and metals in seabird eggs at Prince Leopold Island represents one of the most comprehensive temporal trend data sets available for the Canadian Arctic. The consistency of sampling and analytical methods along with in-depth consideration of the biological characteristics of the seabirds further strengthens the conclusions from this data set and make this a benchmark study for comparison to other studies.

It is clear that levels of legacy OCs in Arctic seabirds are declining with the exception of β-HCH, which is increasing slightly. This is consistent with other temporal trend data sets from Arctic Canada and Europe. Levels of PCDD/Fs and coplanar PCBs are, in general, decreasing in seabirds, although PCDFs are increasing in northern fulmars. PBDEs are increasing in seabirds, consistent with other Arctic studies. Although still at low levels, monitoring of PBDEs and other new chemicals should continue. Mercury is increasing at a significant rate, and is clearly an area of concern for seabirds.

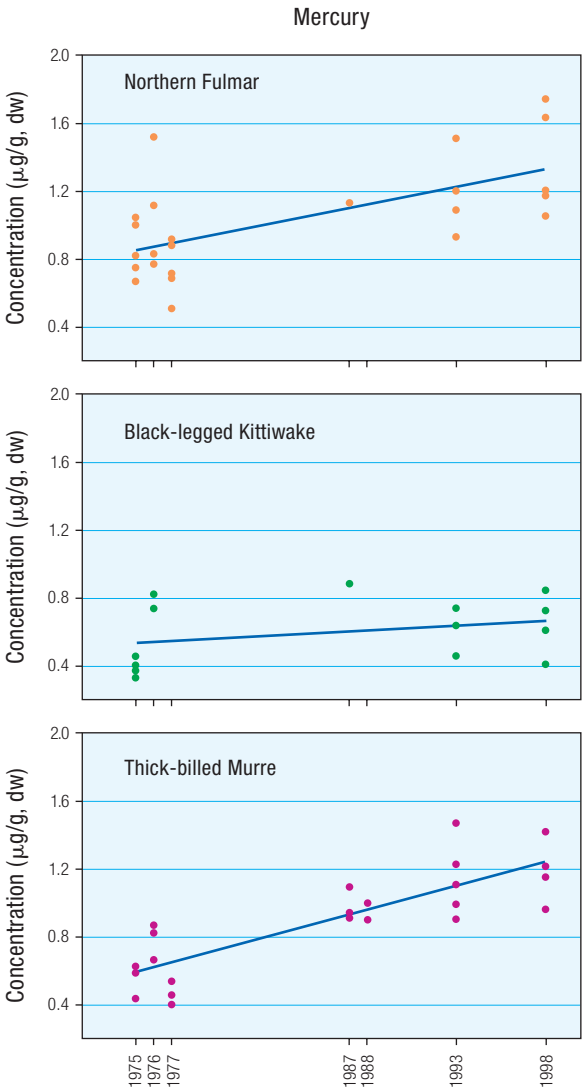

FIGURE 4.2.3

Concentrations of mercury in seabird eggs collected between 1975–1998 on Prince Leopold Island (Braune 2000; Braune *et al.*, 2001a).

4.3 Temporal trends of contaminants in marine mammals

4.3.1 Organochlorines in marine mammals

Ringed seals

Temporal trends of PCBs, DDT, β-HCH and γ-HCH in ringed seals in the Canadian Arctic have been studied at the communities of Ausuittuq (Grise Fiord), Ikpiarjuk (Arctic Bay), and Holman (Muir *et al.*, 2001b; Addison and Smith, 1998). The results for Holman are part of a long-term study (Addison and Smith, 1998; Addison *et al.*, 2000). Elsewhere sampling locations were selected based on whether previous results were available (Muir *et al.*, 1988; Weis and Muir, 1997). At all three locations results are available from the early 1970s to the late 1990s and 2000. At all locations samples from female seals only were selected for analysis to minimize age-related effects on organochlorine levels (Addison and Smith, 1974). Mean concentrations (± 95% confidence limits; ng/g lw) in ringed seals for ΣDDT, ΣPCB, ΣHCH, ΣCHL, along with ratios of recalcitrant members of each class for each location are shown in Figures 4.3.1, 4.3.2 and 4.3.3. ΣPCB10 in seals from Ikpiarjuk declined significantly (2.4 times) from 1975–2000 (Figure 4.3.1) and 1.5 times at Ausuittuq based on comparison of arithmetic means. ΣPCB10 (sum of congeners 28, 31, 52, 101, 105, 118,138, 153, 156, 180) was used for comparison with earlier data rather than all congeners. All previous results, including the samples from 1972–75, were based on capillary GC-ECD with quantitation using authentic standards (Muir *et al.*, 1988; Weis and Muir, 1997; Muir *et al.*, 1999a).

At Holman, ΣPCB (based on conversion of Aroclor 1254 to a ΣPCB value consisting of 20 major congeners), was significantly higher in 1972 than in 1981 and declined significantly over the 10-year period from 1991 (510 ± 133 ng/g lw) to 2001 (335 ± 18 ng/g lw). The overall decline for PCBs was 5.5 times. Ratios of CB153/ΣPCB increased over 25 years at Ikpiarjuk and Ausuittuq and between 1991 and 2001 at Holman.

ΣDDT declined significantly in female ringed seals from all three locations between the early/mid-1970s to late-1990s/2000. ΣDDT had the largest decline of any "legacy" organochlorine — 2.5 times at Ausuittuq, 3.3 times at Ikpiarjuk and 3.3 times at Holman over 25–30 years. Significant increases in *p,p'*-DDE/ΣDDT

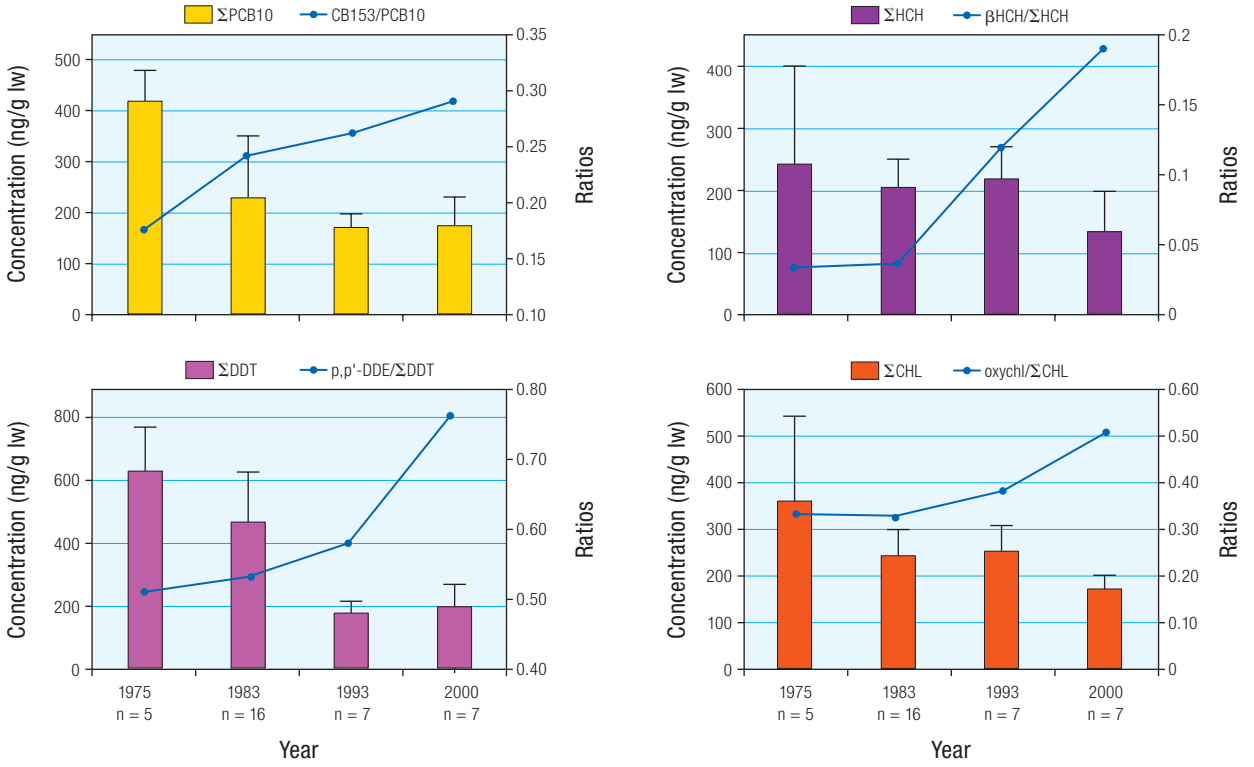

FIGURE 4.3.1

Trends in concentrations and proportions of major components in blubber of female ringed seals from Ikpiarjuk (Arctic Bay). Bars are arithmetic means and vertical lines are 95% confidence intervals. Significant differences between 1975 and 1993 were found for ΣPCB10 and ΣDDT.

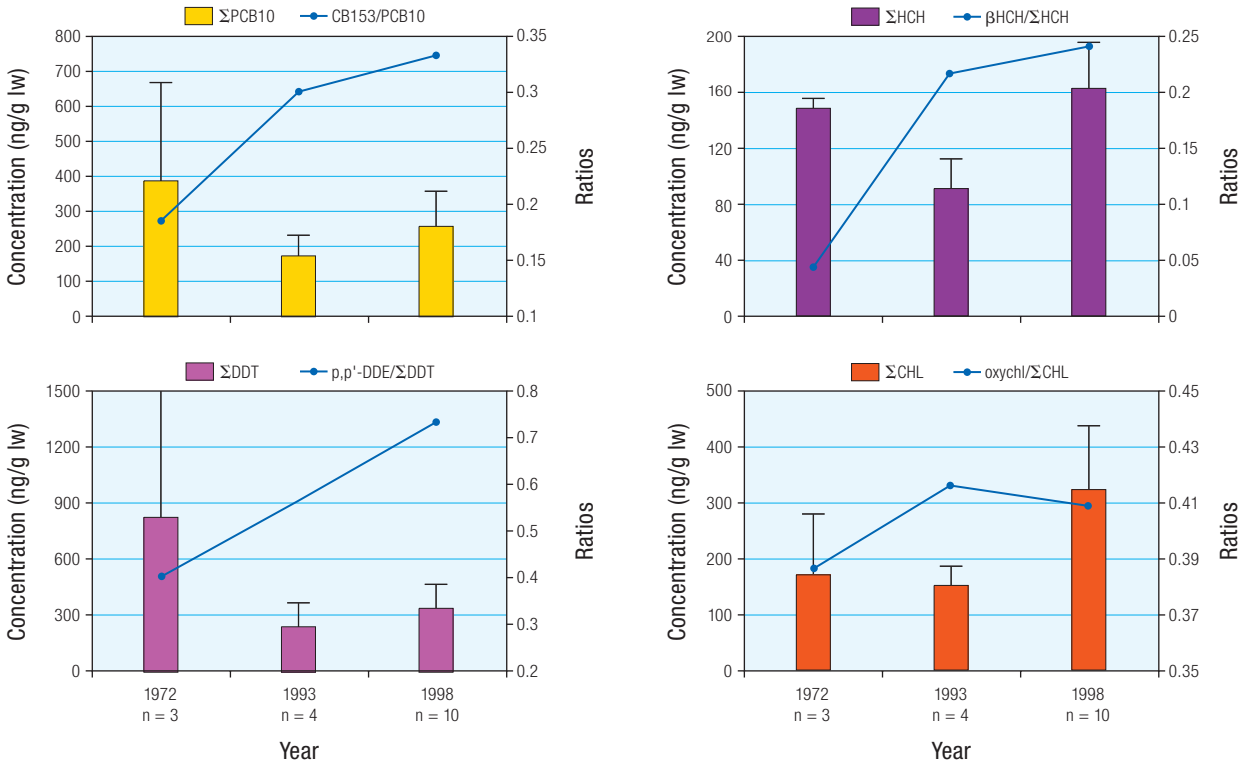

FIGURE 4.3.2

Trends in concentration and proportions of major components in blubber of female ringed seals from Ausuittuq (Grise Fiord). Because of the small sample sizes from 1972 and 1993 no significant differences over time could be found. Nevertheless, the results suggest similar trends to those observed at Ikpiarjuk for concentrations and proportions of major components.

were found at all three locations reflecting the shift from fresh DDT to "weathered" or degraded older sources.

ΣHCH concentrations showed no significant changes in concentrations from the 1970s to 2001 (1981 in the case of Holman). However, β-HCH/ΣHCH increased (3 times at Ikpiarjuk). This shift in the composition of HCH with higher proportions of β-HCH has also been reported in seawater in the western Canadian Arctic during the 1980s and 90s (Li *et al.*, 2002).

ΣCHL showed quite a different trend from ΣDDT, with increasing concentrations at Holman and Ausuittuq, and a slow decline (2.1 times in 25 years) at Ikpiarjuk. Proportions of oxychlordane, a recalcitrant metabolite of chlordane in mammals, increased at all three locations.

Ikonomou *et al.* (2002) examined temporal trends of mono-*ortho* and non-*ortho* PCBs (mo+no PCBs) and PCDD/Fs in male ringed seals from Holman (Figure 4.3.4). Concentrations of mo+no PCBs (149–174 ng/g) and PCDD/Fs (8.6–14.6 pg/g) in ringed seals aged 0–15 years remained approximately constant from 1981–2000 (Ikonomou *et al.*, 2002; Figure 4.3.4). Total PCB concentrations did not decline significantly in males or females from 1981–1991 (Addison and Smith, 1998). Older male seals (16–35 years) from the 2000 sampling group had higher levels of mo+no PCBs than their younger counterparts (0–15 years; 302 versus 150 ng/g). In female seals from 1996 and 2000, mo+no PCB levels were much lower in the 16–35 years age group from 2000 (43 ng/g) than the 0–15 years age group from 1996 (105 ng/g).

Beluga

Stern (1999) examined temporal trends of "legacy" OCs in blubber from beluga in Cumberland Sound (SE Baffin Island, Canada) between 1982 and 1997 (Annex Table 10). They reported a significant decline in α-HCH concentrations over the 15-year interval from 1982 to 1997, while no significant differences were observed for the β- and γ-HCH isomers (Figure 4.3.5). In this regard, belugas differ from seabirds, ringed seals and polar bears, which all show increasing proportions of β-HCH. ΣDDT concentrations did not change over this 15-year interval, however, a 2.2-fold decline and a 1.3-fold increase in the (age-adjusted) concentration of *p,p'*-DDT and its metabolite *p,p'*-DDE, respectively, was observed (Figure 4.3.5). These changes translated into a significant increase in the *p,p'*-DDE/ΣDDT ratio from 0.37 to 0.48 and suggests "old" rather than recent DDT sources. Two of the most abundant congeners in

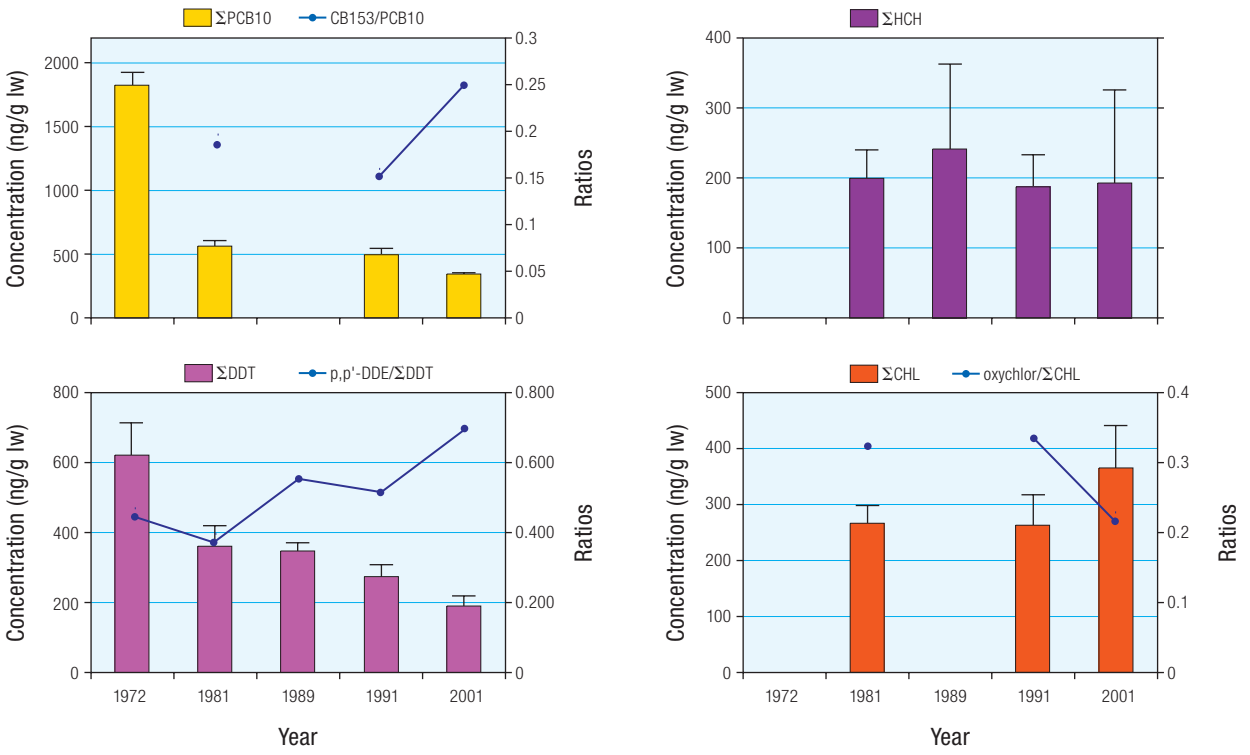

FIGURE 4.3.3

Trends in concentration and proportions of major components in blubber of adult female ringed seals from Holman Island, from 1972–2001.

technical chlordane, *cis*- and *trans*-nonachlor, increased in concentration by 1.4- and 1.7-fold, respectively, from 1982 to 1997, while only *cis*-CHL showed any significant decline. Overall, a 1.2-fold increase in the concentrations of ΣCHL was observed. Oxychlordane, the principal metabolite of *cis*- and *trans*-chlordane, second only to *trans*-nonachlor as the most abundant chlordane-related residue in the SE Baffin beluga blubber, did not change significantly over this 15-year period.

No clear trends were evident in total toxaphene and toxaphene congeners 26 and 50 (Figure 4.3.5). A 2.1-fold increase in age-adjusted mean concentrations of endosulfan (sulfate) and a 1.4-fold decline in dieldrin were also observed over the 15-year period (Stern, 1999). For coplanar PCB congeners, significant declines ranging from 1.7-fold for CB81 to 2.8-fold for CB126, were observed (Figure 4.3.5). Non-*ortho* PCB TEQs (CB77, 126 and 169) have declined from 16 to 6.1 pg/g (2.6-fold) over the 15-year period from 1982 to 1997. Age-adjusted concentrations of major PCB homolog groups (hexa, heptachloro-) did not show a consistent decline over the 15-year period (Stern, 1999; Stern and Addison, 1999).

Stern and Addison (1999) also examined temporal trends of endosulfan, chlorobenzenes and dieldrin in the same beluga blubber samples discussed previously (Figure 4.3.5). Endosulfan sulfate, a metabolite of endosulfan, increased two-fold over the 15-year period. Tetrachlorobenzene (1,2,3,4-TeCBz) also increased 2-fold over the same period. HCB concentrations declined between 1982 and 1992 but were higher in 1996. Dieldrin declined 1.4-fold over the 15-year period.

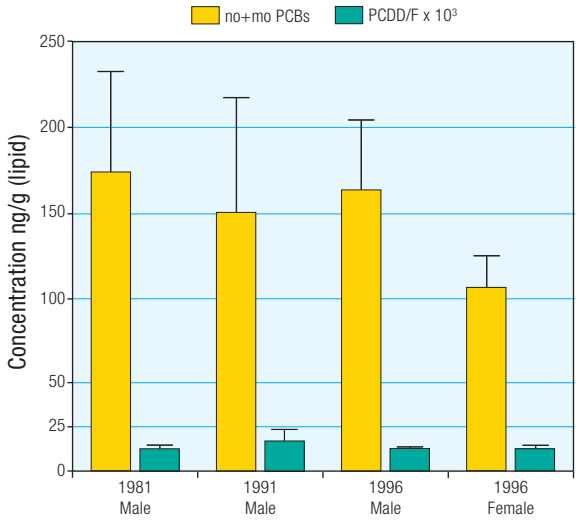

FIGURE 4.3.4

PCDD/Fs and non-*ortho* (coplanar) PCBs and mono-*ortho*-substituted PCBs in Holman Island ringed seals (Stern and Addison, 1999).

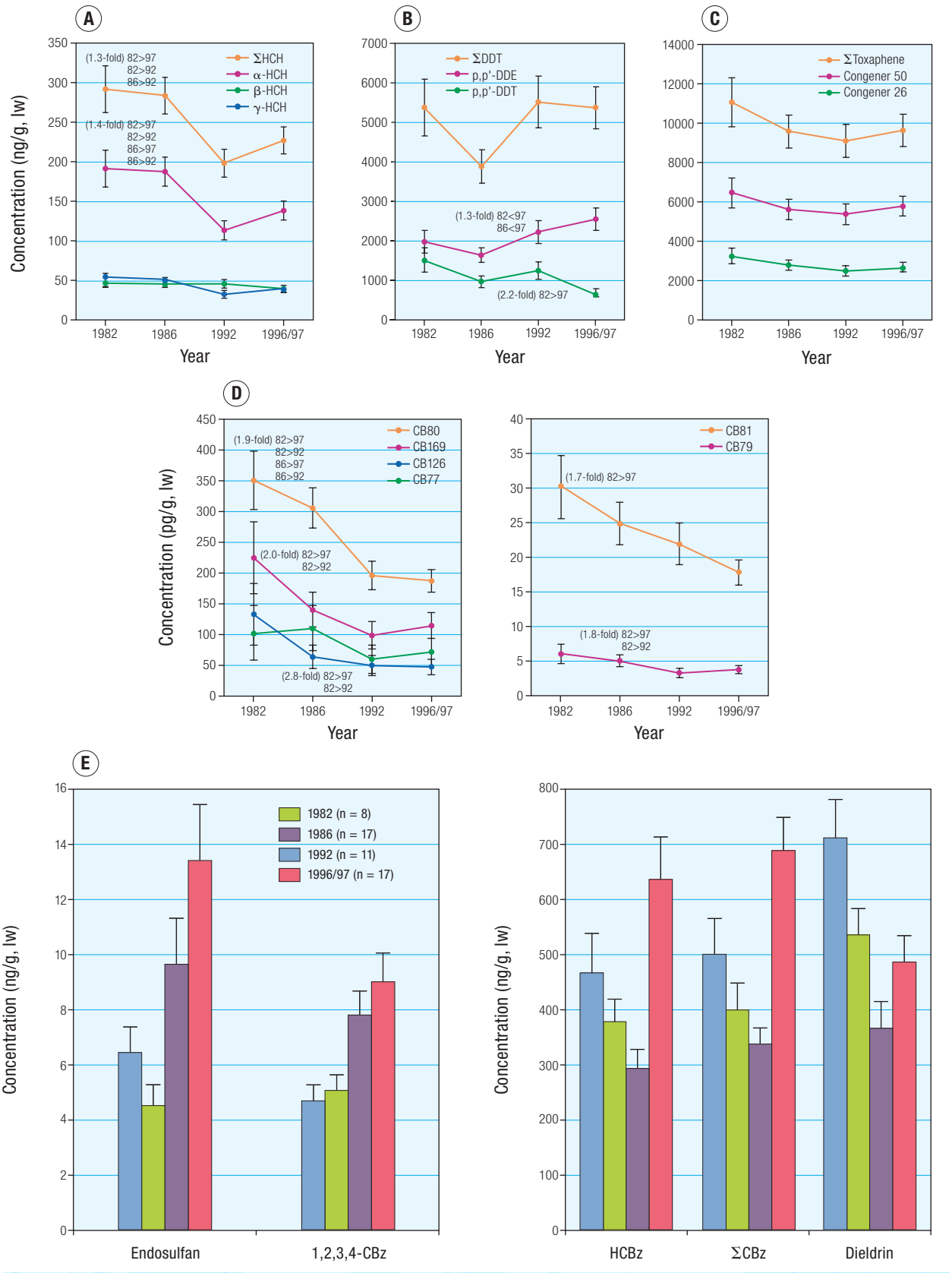

FIGURE 4.3.5

Temporal trends of age-adjusted concentrations in blubber of male beluga from Pangnirtung in the eastern Canadian Arctic of: a) ΣHCH, α-HCH, β-HCH and γ-HCH; b) ΣDDT, *p,p'*-DDE and *p,p'*-DDT; c) Σtoxaphene and congeners 50 and 26; d) PCB congeners 77, 79, 80, 81, 126 and 169; and e) endosulfan, 1,2,3,4-chlorobenzenes, hexachlorobenzene (HCB), sum of tetra-, penta- and hexa-chlorobenzene (ΣCBz), and dieldrin (Stern and Addison, 1999). Concentration differences are significant by t-test at $p < 0.05$.

Narwhal

Results for OCs in blubber of narwhal (*Monodon monoceros*) from the Baffin Bay-Lancaster Sound region (Pond Inlet) of the Canadian Arctic were available from previous reports (Muir *et al.*, 1992b) and recent data (Stern, 2001). Only combined results for 1992 and 1999 were available for the assessment (Annex Table 10); therefore, only a qualitative assessment of temporal changes in legacy OCs in narwhal from this region over a 12–16-year period (1982–83 to 1999) was possible. Male narwhal were selected for temporal trend comparison because, unlike seals, they show little trend in PCB contamination with age (Muir *et al.*, 1992b). Comparison of the combined 1992–1999 data with previous results (Muir *et al.*, 1992b; 1997) suggests no major changes in concentrations of ΣPCB, ΣDDT, ΣCHL, or toxaphene. This lack of change in concentrations of ΣDDT, ΣCHL, ΣPCB and toxaphene in narwhal agrees with results for beluga in the southeastern Baffin Island. One difference between these two odontocetes is the decline in ΣHCH in beluga, while no decline was found in narwhal.

Polar bears

Temporal trends of the major organochlorine groups found in polar bears from the Churchill area of western Hudson Bay are presented in Figure 4.3.6. Biopsy samples from adult female polar bears was chosen to study temporal trends because there is no significant effect of age on any organochlorine. There is a significant effect (increase with age), however, of highly chlorinated PCBs in male polar bears (Norstrom *et al.*, 1998; Bernhoft *et al.*, 1996). Bears less than 5 years old were also excluded because levels of some organochlorines were higher from high exposure in milk during the first two years of life. The trends in adult female polar bears are more likely to represent the actual changes in the polar bear food chain. Biopsy samples were collected opportunistically nearly every year throughout the 1990s (Norstrom, 2001). Adipose tissue samples from 1968, 1984 and 1989 were included to extend the time period for comparison. Because there were no consistent upward or downward trends from 1968–1989 for several organochlorines, only trends in the 1990s were analyzed statistically. There were no statistically significant increasing trends in female bears over the whole nine years. There were statistically significant downward trends of total chlorobenzenes, α-HCH and ΣPCB between 1991 and 1999. There were no significant changes in chlordanes, DDTs (mostly 4,4′-DDE), dieldrin, β-HCH and ΣHCH.

There was a consistent (and significant) decrease of ΣDDT throughout the whole 1968–1990s. Such strong trends are unusual in Arctic biota, suggesting that the phenomenon may be restricted to Hudson Bay. Spraying of DDT for biting insect control in communities and military bases in the Hudson Bay area in the 1950s and 1960s may have contributed a significant load to Hudson Bay during this period. Levels of DDT likely declined after the DDT ban and closing of the large military base at Churchill, and may not have affected other nearby areas.

The overall trend for ΣCBz appears to be an increase between 1968 and 1984, followed by a consistent downward trend since that time. Most of the decline in ΣCBz was due to HCBz, which had a half-life of approximately nine years during the 1990s. HCBz and 1,2,4,5-TeCBz were each roughly half of the total, with a minor contribution from PnCBz. The proportion of 1,2,4,5-TeCBz peaked at 53% between 1995 and 1997, and then decreased to values similar to the pre-1995 period, 40–45%.

The downward trend of ΣHCH in Hudson Bay polar bears in the 1990s was not significant (Norstrom, 2001), but it was significant if 1984 and 1989 were considered (Figure 4.3.6). However, ΣHCH trends are relatively meaningless due to differences in temporal trends of α-HCH and β-HCH. Gamma-HCH was less than 2% of ΣHCH and could not be reliably quantitated in most samples. The apparent half-life of α-HCH in the 1990s was approximately 10 years, very similar to that of HCBz. The decrease in the 1990s appears to be part of a general trend. Levels of α-HCH were about 2–3 times higher in 1968 and 1984 than the average in the 1990s. By contrast, β-HCH concentrations were lower in 1968 than in any subsequent year, and the overall trend in the 1980s and 1990s is very flat. As a consequence, a significantly higher proportion (approximately 50%) of present day ΣHCH in polar bears from Hudson Bay is β-HCH compared to 1984 (25%) and 1968 (17%). A parallel trend is observed in ringed seals the major prey of polar bears.

ΣPCB decreased fairly steadily throughout the 1990s, but with a long half-life of approximately 18 years. The shift in composition of the PCBs was subtle over the decade, but there was a clear tendency for the proportion of less chlorinated congeners to increase, and the highly chlorinated congeners to decrease. Thus CB99 increased from about 10% to 12% of ΣPCB, CB153 was relatively stable at about 35%, and CB180 decreased from 17% to 14%. The trends in these three congeners indicate that the half-life of CB153 (19 years) was similar to that of ΣPCB. The half-life of CB180 (13 years) was shorter and of CB99 (> 50 years, not significant) was longer than ΣPCB. Thus, the decreasing trend in ΣPCB is driven by loss of the highly chlorinated congeners. There is less than a factor of two difference in ΣPCB levels through the 1968–1999 period, and no long term trend is apparent. Concentrations in the early

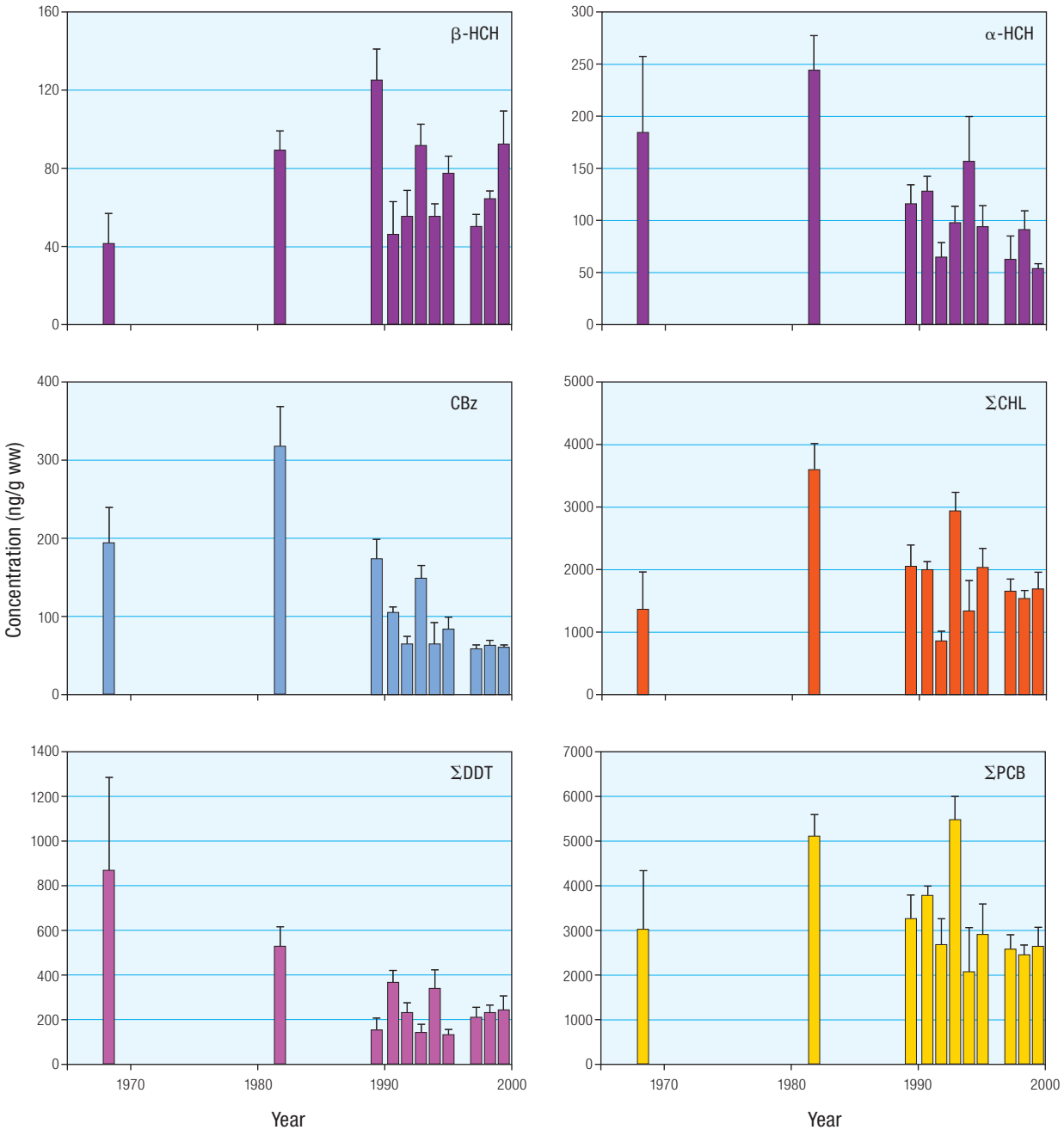

FIGURE 4.3.6

Temporal trends of major organochlorines in Hudson Bay polar bear adipose tissue (Norstrom *et al.*, 2001). Samples are from the Churchill area of western Hudson Bay from 1968–1999. Samples from 1991–1999 are fat biopsies but earlier samples are adipose tissue.

1990s in Hudson Bay were similar to those in the late 1960s, in sharp contrast to the situation in areas such as the Great Lakes or the North Atlantic, where PCBs in herring gull eggs were in the order of 10 times higher in the late 1960s and early 1970s than at present (Hebert *et al.*, 1997).

Shorter term temporal trends in polar bears from five other areas in the Canadian Arctic:

I. Queen Maude Gulf in the western Canadian archipelago
II. Barrow Strait in the central archipelago
III. northern Baffin Bay in the eastern archipelago
IV. Davis Strait in the eastern archipelago
V. north Hudson Bay

were reported by Muir and Norstrom (2000). In that comparison adult male polar bears, 8–10 individuals per area in each year of 1984 and 1989/90, were used. Changes in PCDD/Fs were also determined in pooled samples. Organochlorine concentrations are presented in Norstrom (1997) and, Muir and Norstrom (2000). There were significantly lower concentrations of ΣHCH, ΣDDT and dieldrin in area I, a significant decrease in dieldrin in area II, no changes in area III, a significant increase in ΣCHL in area IV and a significant decrease in ΣCBz, ΣDDT and ΣPCB in area V between 1984 and 1989.

Results for TCDD and PnCDD concentrations in pooled samples from polar bear areas I, II, III and IV in the Canadian Arctic archipelago in 1984 and 1990 are given in Annex Table 13 (Norstrom, 1997). PCDDs and all PCDFs other than 2,3,7,8-TCDD and 1,2,3,7,8-PnCDD were at sub-ng/kg concentrations. Concentrations of TCDD were low and tended to be more evenly distributed in 1990 (0.8–3.1 ng/kg) than in 1984 (2.0–15 ng/kg), as was the case for the other organochlorines. Because pooled samples were analyzed, the significance of the differences cannot be established. Nevertheless, concentrations of TCDD in areas I and II were a factor of 4–5 lower in 1990 than in 1984. The results are consistent with the findings of Norstrom *et al.* (1990) in which ringed seal, beluga and polar bears were analyzed. It is apparent that PCDD/PCDFs are at very low concentrations in the polar bear food chain, and are unlikely to be of toxicological significance.

4.3.2 New chemicals in marine mammals

Ikonomou *et al.* (2002) reported exponential increases in total (Br_2-Br_7)-PBDEs (ΣPBDE) in male ringed seals aged 0–15 years from Holman in the western Canadian Arctic over the period 1981 to 2000 (Figure 4.3.7). ΣPBDE increased nine-fold over this period. Penta- and

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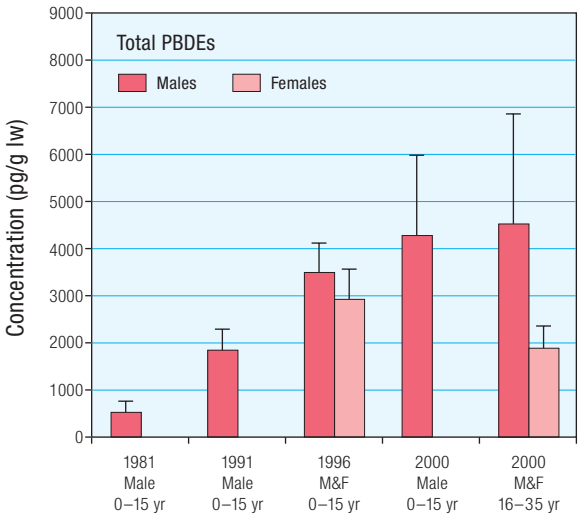

FIGURE 4.3.7

Increasing concentrations of total Br_2-Br_7 PBDE congeners in ringed seal blubber from Holman in the western Canadian Arctic (Ikonomou *et al.*, 2002).

hexa-BDEs were found to be increasing at approximately the same rate (t_2 = 4.7 years and 4.3 years, respectively) and more rapidly than tetra-BDEs (t_2 = 8.6 years) while tri-BDEs showed no increase in this age/sex grouping. The three most prevalent PBDE congeners: BDEs 47, 99, and 100, all increased over the 19-year period; however, only BDEs 47 and 100 increased in parallel with ΣPBDE. BDE99 increased exponentially in a similar manner to ΣPBDE and BDEs 47 and 100 from 1981 to 1996; however, the 2000 samples show the levels of BDE99 stabilizing. This suggests a shift in sources or change in composition of PBDE products. No difference in PBDE levels (both total and of individual congeners, p = 0.98 for ΣPBDE) were observed between younger (0–15 years) and older (16–35 years) male seals in 2000, suggesting that recent PBDE accumulation dominates potential historic accumulation for the older seals.

Stern and Addison (1999) and Stern and Ikonomou (2000) studied temporal trends of polybrominated diphenyl ethers (PBDEs) and polychlorinated diphenyl ethers (PCDEs) in beluga blubber samples from SE Baffin Island in the eastern Canadian Arctic. Levels of the total PBDEs (Br_2–Br_7) and major congeners increased significantly in the SE Baffin beluga over the period 1982–1997 (Table 4.3.1). Age-adjusted concentrations of BDE47 (2,2′,4,4′~TeBDE), the most predominant PBDE congener, increased 6.5-fold over this 15-year period while BDE154 increased 30 times (Figure 4.3.8). Over the 15 year period, contributions of the tribromo- homologue group and PBDE47 to total PBDEs declined by 7% and 3%, respectively. Conversely, pentabromo- and hexabromo-DE contributions have

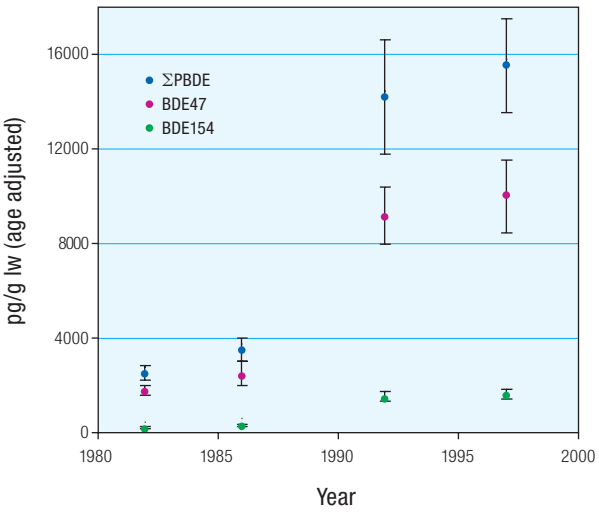

FIGURE 4.3.8

Increasing concentrations of PBDE congeners 47 and 154 along with total Br_2-Br_7 PBDE congeners in beluga blubber (SE Baffin Island, 1982–1996) (Stern and Ikonomou, 2000).

increased by 20% and 80%, respectively. This change in the beluga could be related to the shift in composition of commercial PBDEs to more highly brominated mixtures (DeBoer *et al.*, 2000).

PCDE concentrations declined in the same beluga samples that were analyzed for PBDEs (Stern and Addison, 1999; Stern and Ikonomou, 2000). Maximum concentrations were found in samples from 1982. The two most abundant congeners, CDE 99 (2,2′,4,4′,5-CDE) and CDE 154 (2,2′,4,4′,5,6′-CDE), declined 2.5-fold and 1.8-fold, respectively over the 15-year period. These congeners are prominent contaminants in pentachlorophenol wood preservative and the decline most likely reflects the ban on PCP use in Canada and Scandinavia and use restrictions in the USA.

4.3.3 Metals in marine mammals

Seals

In the previous assessment the data available to 1996 suggested increasing concentrations of mercury in seals, beluga, and narwhal (Wagemann *et al.*, 1996; Muir *et al.*, 1999b) but the rates differed in the eastern and western Arctic, at least for beluga. Results for mercury in seal liver were available for several communities in the eastern Arctic [Qausuittuq (Resolute), Mittimatalik (Pond Inlet), Pangnirtung, Inukjuaq, Kangiqsujuaq (Wakeham), Kangiqsualujjuaq (George River)] and the western Arctic (Holman, Sachs Harbour) from the 1970s to 1990s; however, they were presented only on a regional basis (Wagemann *et al.*, 1996).

Recent (1998–2001) collection of ringed seal tissues from the same locations as those sampled in the 1970s, 80s and early 1990s has enabled comparison of temporal trends of mercury and other metals. Results for both male and female ringed seals were used for mercury in liver because statistical analysis showed no effect of gender of the animals on mercury concentrations. Mercury concentrations ranged widely in adult seals and were highly skewed, however, log transformation reduced skewness and yielded normally distributed data.

The most complete data set was available from Holman, NWT where detailed mercury concentrations along with ages were available for 1972, 1974 and 1977 from the original work of Smith and Armstrong (1978) as well as from Wagemann *et al.* (1996) and Muir *et al.* (2002). Mean concentrations of mercury in seals from Holman aged 5–15 years are shown in Figure 4.3.9. Age-adjustment of the data was also conducted using analysis of covariance but it yielded essentially the same results as selecting only the 5–15 year age group. Average mercury concentrations in Holman ringed seals varied significantly over the 30-year period. Higher concentrations were found in 1974 and 1977 compared to 1993 and 1996. Results for 2001 were also significantly higher than in 1993.

Results for mercury in liver of ringed seals from four other locations are shown in Figure 4.3.10. Geometric mean concentrations from the work of Wagemann *et al.* (1996, and related publications) were combined with

TABLE 4.3.1 Factor increases of major PBDE homologues and congeners over selected time periods for eastern (Pangnirtung) and western Arctic (Eskimo Lakes and Hendrickson Island) beluga and western Arctic (Holman Island) ringed seal. Italicized numbers indicate that the age adjusted mean concentration differences were significant by *t*-test at $p < 0.05$ (beluga only)

	Western Arctic beluga	Eastern Arctic beluga[1]			Holman ringed seal[2]
	1989–95	1982–1986	1986–1992	1992–1997	1991–96
BDE47	*1.5*	*1.6*	*3.7*	1.1	1.9
BDE99	*1.8*	1.6	*6.9*	0.9	1.9
BDE100	*1.9*	1.5	*4.8*	1.1	2
BDE154	*3.6*	*2.1*	*14.1*	1.1	1.9
TrBDE	1.3	1.1	*2.1*	0.9	1.2
TeBDE	1.2	*1.6*	*3.7*	1.1	1.8
PeBDE	*1.9*	1.5	*5.2*	1.1	1.9
HxBDE	*3.8*	*3*	*13.6*	1.1	1.9
ΣPBDE	*1.3*	1.5	*4.1*	1.1	1.8

[1]Stern and Ikonomou (2000); [2]Ikonomou *et al.* (2002).

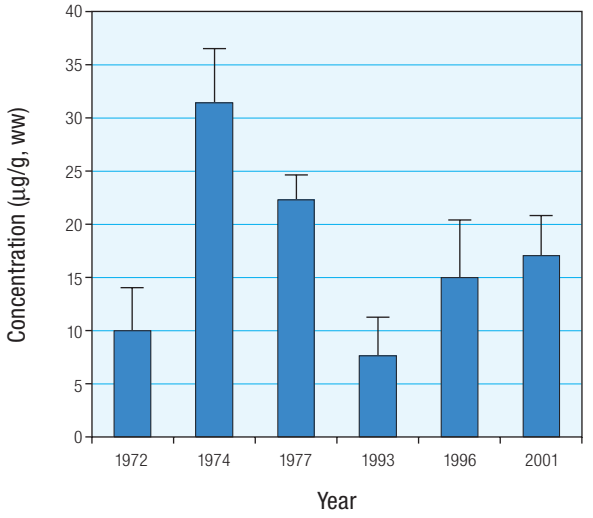

FIGURE 4.3.9

Temporal trends of mercury in ringed seals aged 5–15 years from Holman, NWT (1972–2001). Bars represent geometric means and vertical lines represent upper 95% confidence intervals.

results from 1998–2000 (Muir *et al.*, 2000a; 2001c). The geometric means were not available for results from the 1970s. Therefore, for Mittimatalik (2000) and Qausuittuq, the arithmetic mean concentrations of mercury in adult seals (5–15 years) reported by Smith and Armstrong (1978) were used. The concentrations at Mittimatalik were three-fold higher in 2000 than in 1976, confirming previous conclusions by Wagemann *et al.* (1996) of increasing mercury over time at this location. No significant temporal trends in mercury, however, were found for the other three locations shown in Figure 4.3.10 and no significant trends were found at Ikpiarjuk (Arctic Bay) or Sachs Harbour (Muir *et al.*, 2001b). The year-to-year variation seen at several sites e.g., Holman and Ungava suggests that ringed seals can vary annually in mercury levels in liver. Thus, the Mittimatalik data from 1976 could represent one of those low exposure years presumably due to dietary shifts and it appears that there is no clear increasing trend of mercury in ringed seals.

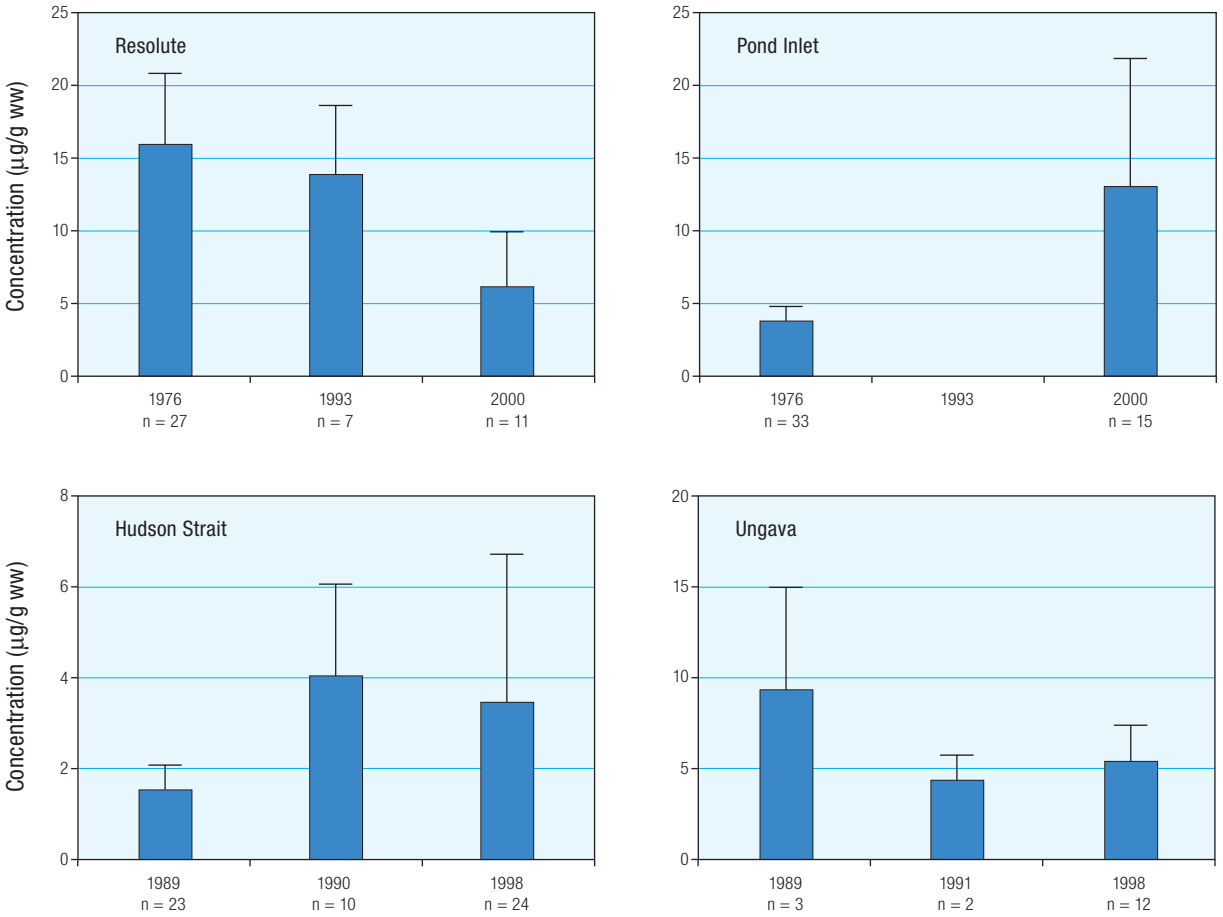

FIGURE 4.3.10

Temporal trend analysis of mercury in ringed seal liver from four locations in the eastern Canadian Arctic. Bars represent geometric mean concentrations and vertical lines represent upper 95% confidence intervals. Results for 1976 from Pond Inlet and Resolute are arithmetic means reported by Smith and Armstrong (1978). For all other results, geometric means were calculated from the original data of Wagemann *et al.* (1996; and related publications) and Muir *et al.* (2000a; 2001b).

No increases in cadmium were found in ringed seal or beluga by Wagemann *et al.* (1996). Cadmium concentrations in liver of ringed seals from Ikpiarjuk from 1974, 1983 and 2000 were very similar (8.35 µg/g ww, 8.63 µg/g ww and 6.16 µg/g ww, respectively) and thus no further assessment of temporal trends of cadmium was done.

Results for lead in seal tissues were near detection limits and too limited to carry out temporal trend assessment.

Beluga

The teeth of Beaufort Sea beluga, harvested in the Mackenzie Delta in 1993 as part of the traditional Inuit hunt, contained significantly higher concentrations of mercury than archeological samples dated 1450–1650 AD (Outridge *et al.*, 2002; Figure 4.3.11). The concentration increases were age-dependent, with older beluga exhibiting larger increases than younger animals, a pattern which matches that predicted by previous modelling work (Bernhard and Andrea, 1984). The increases ranged from four-fold higher in 10-year old beluga, to 17-fold higher in 30-year old animals. Because tooth mercury in the modern beluga was significantly and linearly correlated with soft tissue mercury (including in the muscle and muktuk that traditional Inuit consume), it is likely that soft tissue mercury increased to a similar degree.

While these increases are much larger than those reported in abiotic media anywhere in the Canadian Arctic, modeling also predicted that higher-order consumer species such as beluga would display much greater mercury increases than in the ambient environment, owing to biomagnification and bioaccumulation (Bernhard and Andrea, 1984). The mercury increase could not be explained by changes in diet (shown by stable C and N isotope analyses), sex of the harvested sample, or any other natural phenomenon. The modern beluga also exhibited significant decreases in stable lead isotopes compared to pre-industrial animals, with the trend direction being attributable to the introduction of Eurasian lead (Outridge *et al.*, 1997). In contrast to the Beaufort Sea beluga, walrus at Igloolik in Foxe Basin contained similar concentrations of mercury, cadmium and lead in pre-industrial and modern teeth (Outridge *et al.*, 1997; 2002). Stable lead isotope ratios were also identical between modern and pre-industrial samples, suggesting an absence of industrial lead inputs in this species. This finding supports the bivalve shell data reported by Outridge *et al.* (2000) for other high Arctic sites.

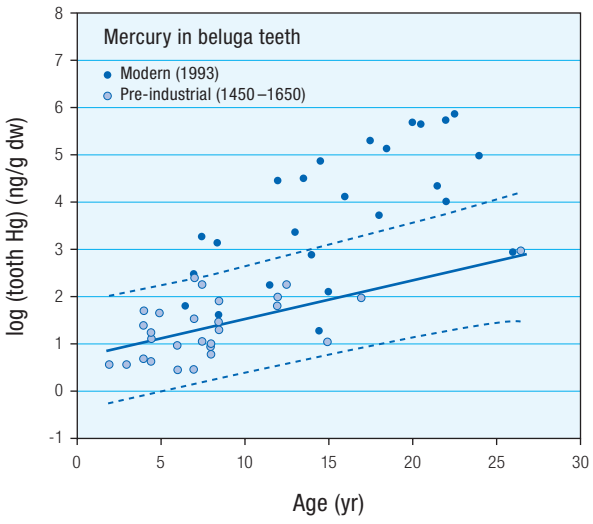

FIGURE 4.3.11

Mercury in modern and pre-industrial teeth of Beaufort Sea beluga whales from the Mackenzie River delta (Outridge *et al.*, 2002).

Wagemann *et al.* (1996) showed that mercury in livers of belugas increased significantly in both the western and eastern Canadian Arctic from 1981–1984 to 1993–1994. Similarly, livers of narwhal sampled from Pond Inlet in the eastern Canadian Arctic in 1992–1994 had significantly higher mean concentrations of mercury than the narwhal sampled from the same site in 1978–1979 (Wagemann *et al.* 1996). There was no indication of any temporal trend of cadmium in livers or kidneys of belugas in the eastern or western Canadian Arctic, nor in narwhal from Pond Inlet (Wagemann *et al.*, 1996).

Mercury data from the late 1990s and 2001 are now available for beluga from several sites in the Canadian Arctic, extending the time series, particularly for the Beaufort coast (Annex Table 9). Most of the samples showed a significant correlation with age and mercury concentration in liver, with the older whales having higher levels of mercury (Stern *et al.*, 2001b). There was little difference between levels in males and females and therefore, all the whales from a collection could be treated as a single data set. Robust regression equations were used to calculate the level of mercury in a 15-year-old whale from each collection (Figure 4.3.12). There was a dramatic four-fold increase in mercury levels in beluga from the Beaufort coast in the western Canadian Arctic between 1981 and 1996 with most of the increase occurring in the 1990s. Whales in the eastern Canadian Arctic did show a somewhat consistent pattern of increase in mercury from the 1980s to the 1990s. Beluga from Arviat (western Hudson Bay) showed much higher levels in 1999 than in 1984 with the age of the whales in the two years being similar. Mercury levels in Pangnirtung beluga did not vary between 1993 and 1997 but both years were greater than levels measured in 1984. Samples from Coral Harbour (northern Hudson Bay) were taken in 1993 and 1997, and from Sanikiluaq (southeastern Hudson Bay) in 1994 and 1998. In both cases, in an

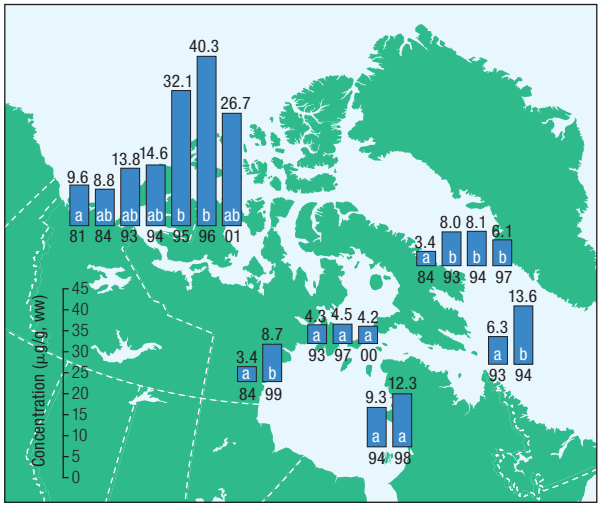

FIGURE 4.3.12

Temporal and spatial trends of total mercury (µg/g ww) in beluga whale liver (Stern *et al.*, 2001b). Figures at the bases of the bars are years when samples were collected and figures at the tops of the bars are means adjusted for the effects of whale age. Letters on the bars indicate statistically significant differences based on Duncan's test. Sanikiluaq showed no trend over time.

interval of only four years, no change in mercury levels were found. At the two locations where samples were taken 13 years or more apart (Beaufort coast, Arviat), an increased rate of mercury uptake was evident in the more recently collected animals (Figures 4.3.13 and 4.3.14). At the third location (Pangnirtung), however, no increase was evident. Belugas showing the greatest changes in uptake of mercury were collected in areas influenced by large freshwater drainages (Mackenzie River, Nelson River, James Bay rivers). This suggests that changes in uptake of mercury by the belugas may be related more to freshwater inputs to these water bodies rather than to direct atmospheric inputs (Stern *et al.*, 2001b). No temporal trends in cadmium concentrations in either beluga or narwhal were found.

Polar bears

In a long-term study using historical samples, mercury levels in the hair of modern polar bears in the Canadian Arctic were reported to be several times higher than in pre-industrial samples recovered from archeological sites (Wheatley and Wheatley, 1988). Possible differences in age (which is a significant factor in the Hg content of many mammals) between modern and pre-industrial populations could not be assessed, and may have influenced the results. No new study of temporal trends of mercury or other heavy metals in polar bears has been conducted.

4.3.4 Modelling temporal trends of organochlorines in pinnipeds and cetaceans

Evaluating temporal trends of OCs in marine mammal populations, and linking them to trends in other biota or environmental media can be confounded by factors that affect tissue concentrations such as age, life span, sex, reproductive activity and blubber thickness. OC concentrations in marine mammals also reflect their exposure history over many years, which could result in a significant lag in response to changes in their exposure levels. These factors can be accounted for quantitatively using species-specific dynamic bioaccumulation models as has been shown in an examination of the history of PCB contamination in the St Lawrence beluga whale population (Hickie *et al.*, 2000). Similar individual- and population-based models developed for ringed seals (Kingsley and Hickie, 1993) are used to reconstruct the history of accumulation of select POPs (ΣPCB, ΣDDT, ΣCHL, α-HCH, β-HCH) and to predict their potential future trends in Arctic populations. The results for males (Figure 4.3.15) are compared to temporal trend data for ringed seals (1972–1991) from Holman Island, from 1972–1991 (Addison and Smith, 1998) and from 2001 (Hoekstra *et al.*, 2002c).

For these simulations, the entire diet of ringed seals was assumed to consist of Arctic cod. Average concentrations reported for Arctic cod from several locations in the Canadian Arctic in the 1990s (Fisk *et al.*, 2001a; Muir *et al.*, 1988; Hargrave *et al.*, 1992) were used to calculate

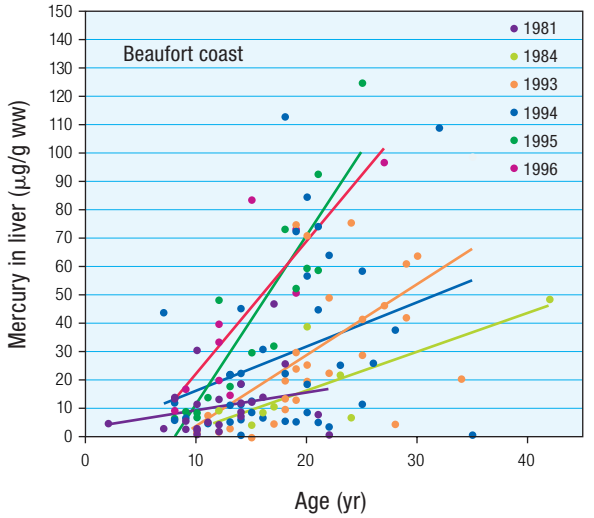

FIGURE 4.3.13

Scatter plot of levels of mercury in liver of beluga whales collected in different years from the Beaufort coast (Lockhart *et al.*, 2001). Least squares regression lines are shown.

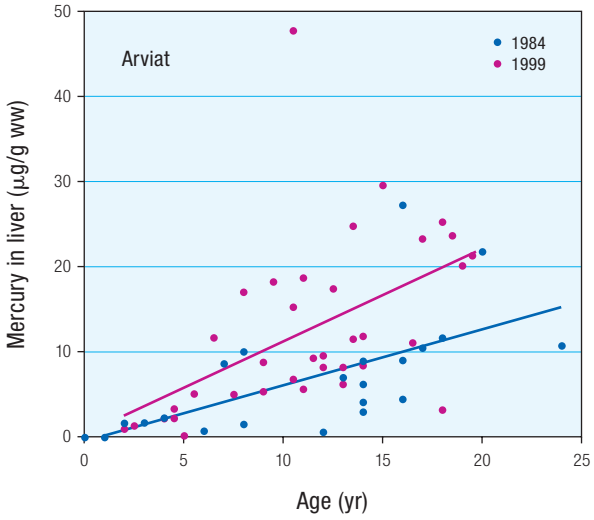

FIGURE 4.3.14

Scatter plot of levels of mercury in liver of beluga whales collected in different years from Arviat on the western coast of Hudson Bay (Lockhart *et al.*, 2001). Least squares regression lines are shown.

baseline average concentrations (*circa* 1996) for use in simulations that extended over the period 1970 to 2010 (Hickie, 2002). Resulting baseline concentrations were 4.7 ng/g ww for ΣPCB, 3.8 for ΣDDT, 4.5 for ΣCHL, 2.1 for α-HCH and 0.32 for β-HCH. Since temporal trend data are lacking for Arctic cod in the Canadian Arctic, trends back to the early 1970s were estimated using temporal trends derived from the Lancaster Sound seabird egg monitoring program (Braune *et al.*, 2001a) using log-linear regressions. Where discrepancies in temporal trends were noted between the three bird species examined, trends from thick-billed murres were used owing to their year-round Arctic distribution. Significant declines were noted for ΣPCB (-5.4%/yr) and ΣDDT (-5.6%/yr) from 1975–1998, while β-HCH levels increased over time (4.2%/yr). Although trends for ΣCHL and α-HCH were not statistically significant, resulting slopes (-1.1%/yr and -0.4%/yr, respectively) were used in model simulations. These trends were assumed to remain in effect in simulations to predict possible future concentrations up to the year 2010. Chemical elimination half-lives for seals used in these simulations were 4.1 years for ΣPCB, 6.9 for ΣDDT, 3.3 for ΣCHL, 2.4 for α-HCH and 1.8 for β-HCH. These were derived from model calibration exercises using independent data sets (Hickie, 2002; unpublished data). The half-life estimate of ΣPCB was based on the weighted sum of half-life estimates for the 20 most abundant PCB congeners.

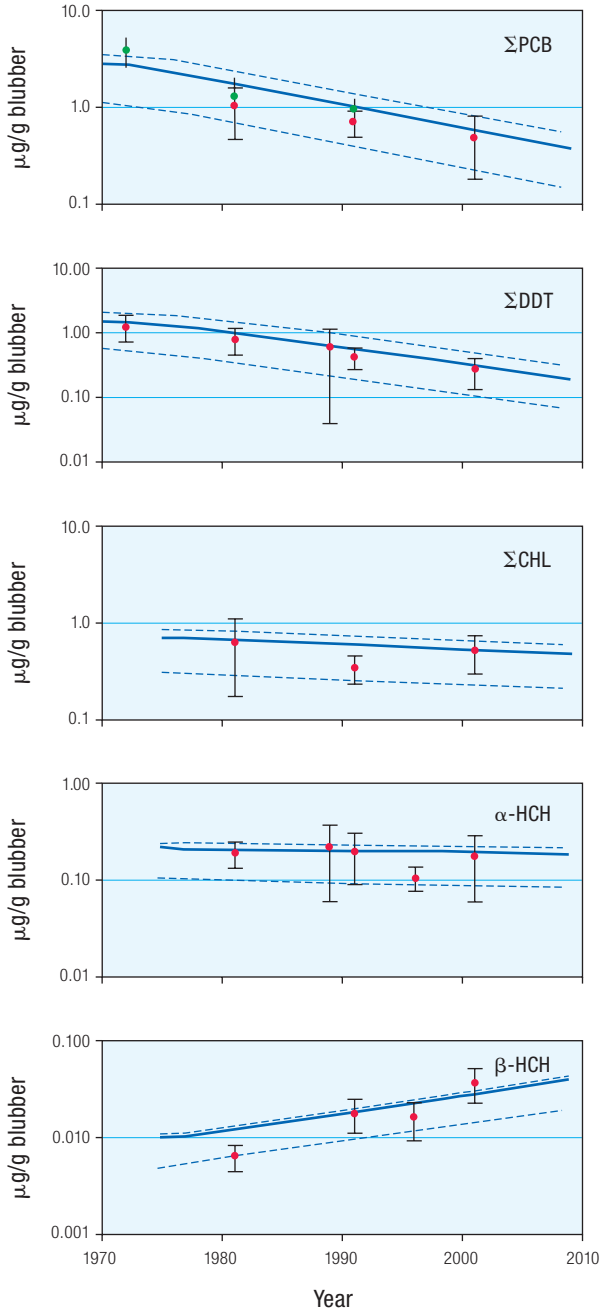

FIGURE 4.3.15

Observed and predicted trends of POPs concentrations (µg/kg blubber) for ringed seals from Holman Island, NWT (Hickie, 2002). Temporal trend data are mean values (± SD) reported by Addison and Smith (1998) from 1972–1991 and by Hoekstra *et al.* (2002c). Simulation results are presented as the mean (solid line) and range (dashed lines) of predicted concentrations for males ranging in age from 2 to 20 years. Data for PCBs are shown as both Aroclor 1254 (green symbols) and ΣPCB congeners (red symbols).

Overall, the simulations showed good agreement with observed temporal trends in the Holman Island seal population for all five chemicals examined. They also demonstrate that the temporal trends evident in ringed seals are consistent with those in seabirds, and likely reflect changes throughout lower trophic levels in the marine ecosystem. The results also suggest that contaminant levels in ringed seal populations do not show any appreciable lag in response when contaminant exposure concentrations change gradually over time, as appears to be the case in the Arctic. The lack of a lag in response can be attributed to the relatively rapid elimination rates for the chemicals examined combined with the effect of population turnover. The strong agreement between the simulations and monitoring data serves to validate the model, and demonstrates that it can be used with some confidence in forecasting responses to potential future loading scenarios. Concentrations of ΣPCB and ΣDDT are predicted to decline by about 40% between 2000 and 2010 to ranges of 0.15–0.50 μg/g for ΣPCB and 0.06–0.30 μg/g for ΣDDT for the scenarios presented herein.

4.3.5 Overall assessment of temporal trends of contaminants in marine biota

A critical question in the assessment of OCs in the Arctic is whether concentrations are increasing or decreasing. The previous CACAR assessment found limited information on temporal trends of OCs in biota. As of 2001, the temporal trend information is much improved. Previous studies have been extended so that a 25- to 30-year perspective is available for polar bears, seabirds and ringed seals and new studies covering a 10–15-year period are available for burbot, lake trout and beluga whales. New chemicals such as the PBDEs have been added to the list of chemicals for which temporal trends are available in marine biota.

There are a number of temporal trends data sets available for ringed seals that cover a large range of years (up to 30 years) and provide good spatial coverage. Significant declines of PCBs and DDT have been observed in all ringed seal populations examined to date. This is consistent with results in other organisms and media from the Canadian and circumpolar Arctic. Increases in the proportion of DDE to ΣDDT suggest that the DDT is weathered or old. There have been limited changes in coplanar PCBs in the Holman Island population. HCH concentrations have not declined from the 1970s to 2001 due to the continued use of these chemicals in Asia until very recently. As well, increasing proportions of β-HCH may play a role in observed trends. The chemical-physical properties of β-HCH differ significant from the other HCH isomers and there is a lag time for the delivery of β-HCH compared to the other isomers to the Arctic. Chlordane appears to be increasing in some ringed seal populations but not others.

Temporal trends of OCs in beluga from southeastern Baffin Island are similar to what has been observed in ringed seals. DDT and PCBs, including coplanar PCBs, declined in these beluga between 1982–1997. This increase in chlordane in this population is consistent with some populations of ringed seals. A significant decline in α-HCH has also been found in these beluga and no trend was seen in toxaphene.

No significant trends in most OCs have been found in polar bears between 1968 and 1989. The exception is DDT, which steadily declined in Canadian polar bears up to 1989. Recent analyses of temporal trends in western Hudson Bay polar bears between 1991 and 1998 have found declining concentrations of chlorobenzenes, α-HCH, and PCBs but no decline in concentrations of chlordanes, DDT, β-HCH and ΣHCH for the same time period.

Information on temporal trends of PBDEs is available for Holman ringed seals and southeastern Baffin Island and western Arctic beluga. Dramatic increases in PBDE concentrations have been observed in both populations. A nine-fold increase in ΣPBDE between 1981–2000 was found in the ringed seals. A 6.5-fold increase in age-adjusted concentrations of PBDE 47 between 1982–1997 was found in the southeast Baffin Island beluga. Similar trends are seen in the western Arctic beluga.

Use of ringed seal bioaccumulation models has provided a future forecast for OC concentrations in ringed seals. This model generated concentration data in ringed seals that was very consistent with measured levels suggesting that it has good potential to predict future concentrations. The model found that there is no lag between OC concentrations measured in ringed seals and changes in the level of exposure. That is the ringed seal provides a good measure of local contamination. The model predicted that DDT and PCB levels in ringed seals will decline by 40% between 2000 and 2010.

Mercury data were available for seabirds, beluga and ringed seals from a number of populations from the 1970s through to 2000. This assessment focussed almost exclusively on mercury because of its toxicological significance and because preliminary analysis suggested no changes in cadmium or lead concentrations in ringed seals. The trends for mercury vary with species and are summarized in Table 4.3.2 along with trends for other metals. Over the long term, i.e., comparing modern (1993) and pre-industrial (1450–1650) samples, there is

little doubt about increased levels of mercury; however, the short-term trend is more difficult to assess.

A three-fold increase in mercury concentrations was observed in Mittimatalik ringed seals between 1976–2000 but no significant increase has been found in six other ringed seal populations including at Holman where the longest time trends were available. Given the year-to-year variation seen at several sites, e.g., Holman and Ungava, which suggests that ringed seals can vary annually in mercury levels in liver, the results for Mittimatalik in 1976 could represent a low exposure year, presumably due to dietary shifts. It appears, therefore, that there is no clear increasing trend of mercury in ringed seals.

The temporal trends for mercury, however, could be localized. Braune (1999) found a significant, approximately two-fold, increase in mercury in thick-billed murre eggs from Lancaster Sound from 1975–77 to 1998, which is also in accord with the results from Mittimatalik. Results for ringed seals from Avanersuaq in northwest Greenland also show a significant increase in mercury from 1984 to 1998 (Riget, 2002). Mercury has also been shown to be increasing in narwhal collected at Mittimatalik between 1978–79 and 1992–1994.

The increase in mercury levels in beluga liver is also localized with significant increases only for the Mackenzie Bay animals and at Arviat. Analysis of age-mercury concentration slopes in beluga suggest that rate of accumulation in beluga has been increasing from the early 1980s until recently. In the southern Beaufort Sea these increases are not paralleled in ringed seals from the same region, e.g., Sachs Harbour and Holman in the western Arctic, although seals from the Mackenzie River delta have not been analyzed.

Rivers are probably a significant transport vehicle of mercury into the Arctic marine environment although there are few measurements on large river systems to confirm this. Dietz *et al.* (1998) summarized mercury measurements in north flowing rivers and reported concentrations of 5–8 ng/L (total mercury) at the mouth of the Mackenzie River. Mercury concentrations in runoff are known to increase with flow. The large increases in mercury concentrations that have been observed recently in the Mackenzie Bay belugas coincide with a warming trend in the western Arctic. Thus, melting of permafrost and movement of organically bound mercury and its accumulation in the form of methylmercury might explain the observed trends; however, much additional work needs to be done to confirm this.

GNWT/RWED/Brett Elkin

TABLE 4.3.2 Summary of temporal trends of mercury and other metals in the Canadian Arctic

Biota		Location	Collection years	Time period (yrs)	Hg	Cd	Se	Pb
Terrestrial & freshwater								
Caribou	Kidney	South Baffin	1992, 1999	8		nc		
Lake trout	Muscle	Lake Laberge	1993, 1996, 1998	6	decr/nc?			
Lake trout	Muscle	Quiet Lake	1992, 1999	8	nc			
Lake trout	Muscle	Great Slave Lake	1979–2001	23	inc			
Burbot	Liver	Lake Laberge	1996, 1999	4	nc			
Burbot	Muscle	Fort Good Hope	1985, 1993, 1995, 1999, 2000	16	incr			
Burbot	Muscle	Great Slave Lake	1995, 1996, 1999	5	nc			
Char	Muscle	Resolute Lake	1992, 1997, 1999, 2000, 2001	10	nc			
Char	Muscle	Char Lake	1992, 1999, 2000, 2001	10	incr/nc?			
Walleye	Muscle	Great Slave Lake	1987–1998	21	decr			
Pike	Muscle	Great Slave Lake	1988–2001	23	decr			
Marine long-term								
Mussel		Axel Heiberg & Cornwallis Is	1000–9000 BP, 1980s–1990s	~9000	nc	nc		nc
Mussel		SE Hudson Bay	1000–9000 BP, 1970s	~9000	incr	nc		incr
Walrus	Teeth	Igloolik	1200–1500, 1987–1988	~700	nc	decr		nc
Polar bear	Hair		300BC–1500AD, 1930–1977	~2000	incr			
Beluga	Teeth	Mackenzie Delta	1450–1650, 1993	~500	incr	decr		decr?
Marine short-term								
Northern fulmar	Eggs	Lancaster Sound	1975–1977, 1987–1988, 1993, 1998	23	incr			
Black-legged kittiwake	Eggs	Lancaster Sound	1975–1977, 1987–1988, 1993, 1998	23	nc			
Thick-billed murre	Eggs	Lancaster Sound	1975–1977, 1987–1988, 1993, 1998	23	incr			
Beluga	Liver	Beaufort coast	1981, 1984, 1993, 1994, 1995, 1996, 2001	20	incr			
Beluga	Liver	Arviat	1984, 1999	16	incr			
Beluga	Liver	Pangnirtung	1984, 1997	14	nc			
Beluga	Liver	Coral Harbour	1993, 1997, 2000	7	nc			
Beluga	Liver	Sanikiluaq	1994, 1998	5	nc			
Ringed seal	Liver	Holman	1972, 1974, 1977, 1993, 1996, 2001	29	incr/nc?			
Ringed seal	Liver	Mittimatalik	1976, 2000	25	incr			
Ringed seal	Liver	Resolute	1976, 1993, 2000	25	nc			
Ringed seal	Liver	Hudson Strait	1989–1990, 1998	10	nc			
Ringed seal	Liver	Ungava Bay	1989, 1991, 1998	10	nc			
Ringed seal	Liver	Sachs Harbour	1987, 1988, 2001	14	nc			
Ringed seal	Liver	Ikpiarjuk	1983, 1993, 2000	17	nc	nc		

ITK/Eric Loring

Biological Effects

5.1 Introduction

The first CACAR concluded, "*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 levels*". The conclusion was reached by assessing the limited number of studies that were available at the time and to a lesser extent by comparing levels with threshold effects. The report further concluded that a lack of information on biological effects in Arctic organisms was a major knowledge gap. Since this report there have been very few studies that have examined the possible effects of contaminants on Arctic biota (Muir *et al.*, 1999b).

Examination of possible biological effects in Arctic organisms is a difficult task. There are inherent problems with any study of wildlife in the Canadian Arctic due to the difficulty and expense of travel and fieldwork. Beyond this, the linkage of biological effects to anthropogenic contaminants is difficult. Biological effects can be measured at different levels of biological organization, from the molecular level to the ecosystem level. Biomarkers measurable at a molecular level respond early, but are not readily interpreted ecologically, while measures with established ecological relevance, such as population declines or reduced reproductive rates, respond too late to have diagnostic or preventative value.

There are two basic approaches in assessing the possible biological effects of anthropogenic contaminants in wildlife. The first involves comparison and extrapolation. The possible effects of contaminants on Arctic species are assessed by comparing levels of a contaminant of interest to levels known to be detrimental, with this knowledge coming from laboratory studies, semi-field studies, or from observations on affected animals in the wild. These types of comparisons have inherent weaknesses. Laboratory animals are most often exposed to single organic contaminants (OCs) or metals, or to technical products at high doses for short periods of time, and it is difficult to extrapolate the toxic effects seen at high acute doses to possible adverse effects at lower, but chronic exposures. Wildlife are exposed to weathered mixtures due to the change in composition of many OC mixtures caused by abiotic degradation, metabolism and subsequent filtering up through the food web. For example, marine mammals at high trophic levels will be exposed to very different PCB compositions expressed as ΣPCB, than is seen in a PCB technical product. The sensitivity to the effects of OCs and metals also varies among species and the extrapolation of effects levels developed for laboratory animals is a confounding problem. Several Arctic species, for example, have delayed implantation (e.g., seals, walrus, polar bears), which may make them more sensitive to the reproductive effects of contaminants than tested laboratory animals without delayed implantation (Sandell, 1990). Very little is known about the sensitivity of Arctic species, particularly marine mammals, to the effects of OCs (de Wit *et al.*, 2003; in press) and metals (AMAP, 2003; in press). Wild animals are generally exposed to lower concentrations of OCs, but they are exposed to mixtures of contaminants and other stressors, and they are exposed over their entire lifetime.

The second approach studies biological effects by measuring biological responses that are related to contaminant exposure, often called biomarkers. At present, use of biomarkers is one of the only methods available to test the hypothesis that trace contaminants are acting biologically on the animals. Almost any biological change, from molecular to ecological, can serve as a biomarker; however, the term most often refers to changes at sub-cellular levels (McCarthy and Shugart, 1990; Huggett *et al.*, 1992; Peakall, 1992). Biomarkers typically are measures of normal processes that take on abnormal values as a result of exposure to chemicals of interest. The sensitivities of biomarkers have generally been established using laboratory animals and their applicability to Arctic wildlife is little studied. It is also not possible to determine causality, only that a statistical association has been found between a biomarker and the contaminant in question. Most contaminants co-vary and thus it is not possible to state equivocally that the biomarker response has been caused by a particular contaminant. There may be other contaminants not analyzed that are just as significant.

This assessment examines the small number of biological effects of OCs studies carried out on Arctic wildlife since the first CACAR (post 1996). It then draws upon threshold levels established for OCs and metals that were chosen for the upcoming AMAP assessments of OCs and metals in the circumpolar Arctic to assess the potential effects of current levels of contaminants in Arctic wildlife (AMAP, 2003; in press; de Wit *et al.*, 2003; in press).

5.2 Effects studies

There have been four known studies that were carried out to assess the possible biological effects of OCs and metals in Canadian Arctic wildlife since the first CACAR was published (i.e., post 1996).

5.2.1 Eider ducks

Populations of eider ducks in Alaska and the Canadian Arctic have declined precipitously in the past several decades (Gratto-Trevor *et al.*, 1998; Robertson and Gilchrist, 1998; Suydam *et al.*, 2000). The causes of these declines have not been identified but contaminants may be one of several risk factors for North American sea duck populations (CWS *et al.*, 1997). Trace elements, in particular cadmium and selenium, have been found at elevated concentrations in eider ducks in Arctic and subArctic areas (Norheim, 1987; Nielsen and Dietz, 1989; Henny *et al.*, 1995; Dietz *et al.*, 1996; Trust *et al.*, 2000; Wayland *et al.*, 2001).

A paucity of useful information exists concerning the possible toxic effects of trace elements on sea ducks. Most information comes from laboratory-based, captive-feeding experiments wherein surrogate species such as the mallard (*Anas platyrhynchos*) were exposed to relatively high levels of single trace elements (DiGiulio and Scanlon, 1985; Heinz *et al.*, 1989; Bennett *et al.*, 2000). The results of such studies are difficult to interpret in terms of potential effects on sea ducks for the following reasons:

1. Sea ducks and experimental species (mainly the mallard) may differ in their sensitivities to a contaminant.
2. Sea ducks in the wild are exposed simultaneously to varying levels of multiple trace elements, some of which may reduce (Magos and Webb, 1980) or increase (Gochfeld, 1997) the toxic effects of others. Typically, such variation is not represented in laboratory studies.
3. Natural environmental stressors, of the type that wild animals routinely encounter (e.g., adverse weather), may act together with contaminants to produce physiological impairment (Forsyth, 2001). Such natural stressors are rarely considered in laboratory-based studies.

It is important to examine trace element concentrations while simultaneously examining the health of these birds under natural conditions to minimize the risk of incorrectly extrapolating the results of laboratory-based studies to wild sea ducks . Health-related biomarkers that may have a bearing on population dynamics would be especially valuable in this regard. One such biomarker is body condition. In waterfowl, various measures of body condition have been linked to reproductive effort (Milne, 1976) and success (Blums *et al.*, 1997) and to survival rates (Bergan and Smith, 1993). If exposure to elevated concentrations of certain trace elements is related to reduced body condition, as has been alluded to by Henny *et al.* (1991), then population dynamics could ultimately be affected. The incidence and severity of disease may also affect wild populations of sea ducks. Poor health and large die-offs of common eiders have been attributed to infestations of acanthocephalan parasites (Persson *et al.*1974; Hollmén *et al.*, 1999). Thus, parasitic infestations could affect eider populations. It has also been shown experimentally that exposure to certain trace elements can increase the severity of parasitic infestations (Boroskova *et al.*, 1995), providing a possible mechanism by which such exposure could affect eider populations. Other biomarkers that may be beneficial for evaluating the health of sea ducks in relation to exposure to trace metals include immune function and stress response. Changes in the immune system, which can affect susceptibility to disease, may provide sensitive, early warning signals of the toxic effects of contaminants, including metals (Lawrence, 1985). The response to acute stress, as evaluated by measurements of corticosteroids, is another biomarker that may be affected by some trace metals (Hontela, 1997). Corticosteroids regulate processes related to energy metabolism, salt gland function and immune function (Hontela, 1997), and therefore are important in the maintenance of homeostasis in animals. Finally, because exposure to trace elements has been associated with tissue damage in some seabirds (Nicholson *et al.*, 1983), histopathological assessment of certain organs can provide useful information concerning animal health.

Body condition in relation to trace elements

In a multiple regression with mercury, cadmium and selenium as independent variables, body weight was inversely related to hepatic mercury concentrations in common eiders in three out of four years in which this relationship was examined. Similarly, abdominal fat mass (AFM) was negatively related to hepatic mercury concentrations in two of four years (Figure 5.2.1). After adjusting for the effects of mercury, cadmium was negatively

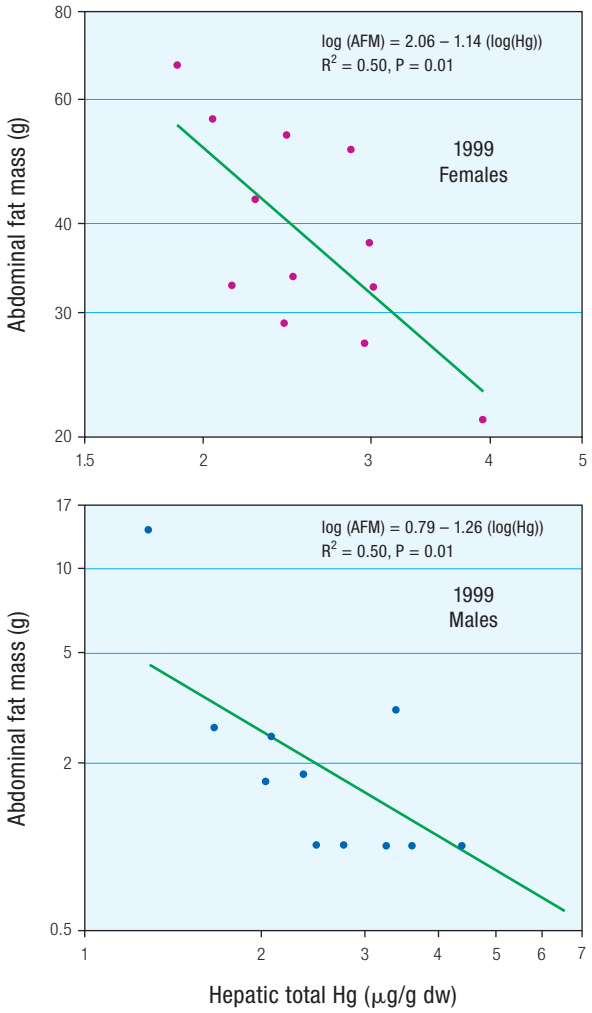

FIGURE 5.2.1

Abdominal fat mass in relation to hepatic total mercury concentrations in common eiders sampled at the East Bay Migratory Bird Sanctuary, Southampton Island, 1999–2000.

related to body weight in females sampled in 2000 and to abdominal fat mass in females sampled in 1999.

It is now widely accepted that body mass and fat stores are important determinants of fitness and survival in ducks (Milne, 1976; Bergan and Smith, 1993; Blums *et al.*, 1997). Because of their importance to fitness and survival, Henny *et al.* (1991) postulated that the negative relationship between cadmium concentration and body mass in sea ducks on the Pacific coast may be a cause for concern. Implicit in their concern was that there may have been a cause-effect relationship between cadmium and body mass in those birds. In the intervening years, other field studies also have reported negative relationships between metal concentrations in various tissues of sea ducks and indices of their body condition (Ohlendorf *et al.*, 1991; Hoffman *et al.*, 1998; Franson *et al.*, 2000). However, these findings have generally not been supported by experimental studies, wherein captive birds have been fed a range of concentrations of mercury or cadmium. For example, in a multi-generation study, mallards that were fed 0.5 mg/g methylmercury in their diets accumulated approximately 1.5 mg/g ww mercury in their livers. Body weights of adults that were fed the mercury-containing diet did not differ from those of adults receiving a control diet (Heinz, 1979). Similarly, body weights of mallards with hepatic mercury concentrations of 22 mg/g ww after being fed 10 mg/g dietary methylmercury for about 2.5 months were not different from those of mallards that received a control diet and had hepatic mercury concentrations less than 1 mg/g (Heinz and Hoffman, 1998). Body weights of ducks fed cadmium at 2–200 mg/g (White and Finley, 1978; Di Giulio and Scanlon, 1985; Bennett *et al.*, 2000) did not differ from those of control ducks. Dietary cadmium at 300 mg/g was associated with body mass loss and other toxic effects (Bennett *et al.*, 2000). It is extremely unlikely, however, that the food of wild sea ducks would have such high concentrations of cadmium (Dietz *et al.*, 1996).

The lack of agreement between the results of experimental feeding studies and field studies may indicate that cadmium and mercury can affect wild birds through indirect mechanisms that are not operational in captive feeding studies. A more probable explanation is that in the wild, normal, seasonal changes in body and organ mass directly affect concentrations of relatively immobile trace elements such as cadmium and mercury. For example, during periods of fasting with consequent loss of body and organ mass, cadmium and mercury may become increasingly concentrated in kidney and liver. In contrast, when birds are gaining weight, as occurs during the pre-laying period in eiders, cadmium and mercury may become increasingly diluted in the rapidly expanding tissue pool. It is possible, therefore, that the inverse relationships between body/organ mass and mercury/cadmium concentrations in this study simply reflect the relative degree to which individual birds are gaining or losing weight at the time they are sampled, rather than an effect of the metals on metabolic pathways affecting body and organ mass.

Parasite burdens in relation to trace elements

The relationship between counts of gastrointestinal parasites and tissue trace element concentrations was examined in 1997 (Wayland, 1999). Nematodes were found in 18 of 20 king eiders and cestodes were found in 17 of 20 king eiders. Nematodes and cestodes were found in 30 of 33 common eiders. Acanthocephalans were found in eight king eiders and 25 common eiders. In both species, after adjusting for differences among sampling

locations, residuals of cestode and acanthocephalan numbers were not negatively correlated with those of trace element concentrations. However, in common eiders, but not in king eiders, residual total and organic mercury concentrations were positively correlated with residual nematode numbers (Figure 5.2.2).

In an experimental study on guinea pigs, the number of nematodes was 15% higher in animals dosed with mercury for 28 days and infected with eggs of the nematode, *Ascaris*, than in infected animals not dosed with mercury (Borošková *et al.*, 1995). The higher number of parasites was attributed to suppressed lymphocyte production in the lymphoid organs caused by mercury. For free-living homeotherms, information concerning the possible effects of contaminants on parasite infestations is scarce. Notably, the number of gastrointestinal nematodes was positively correlated with PCB concentrations in livers of glaucous gulls (*Larus hyperboreus*) from Svalbard, Norway (Sagerup *et al.*, 2000). Dead loons in poor body condition had higher concentrations of mercury and greater numbers of intestinal trematodes than those in good body condition (Daoust *et al.*, 1998). However, those authors were unable to ascertain whether the high numbers of trematodes could be directly attributed to high mercury levels. The findings of those studies, together with the results herein, suggest that parasite infections in free-living birds may be affected by contaminants.

Acanthocephalan parasites, but not nematodes, have been associated with die-offs or poor health of eiders ducks (Persson *et al.*, 1974; Hollmén *et al.*, 1999). Counts of acanthocephalans were not correlated with trace element concentrations in either species. Thus, it cannot be concluded that exposure of eiders to trace elements is likely to increase parasite-induced mortality in eiders.

Other biomarkers

In 1999 and 2000, the relationship between trace element exposure and certain aspects of the immune response was examined in common eiders at East Bay on Southampton Island (Wayland, 1999). Two immune function assays were conducted: (1) the skin-swelling response to an intradermal injection of phytohemagglutinin-P (PHA-P), and (2) the antibody titer in response to an intraperitoneal injection of sheep red blood cells (SRBC). The former assay elicits a simple and rapid cell-mediated immune response while the latter elicits a humoral immune response. Tissue trace element concentrations did not significantly dampen the immune responses of eider ducks in this study. In 1999, selenium concentrations were positively related to the PHA-P response but were not significantly related to the antibody titer.

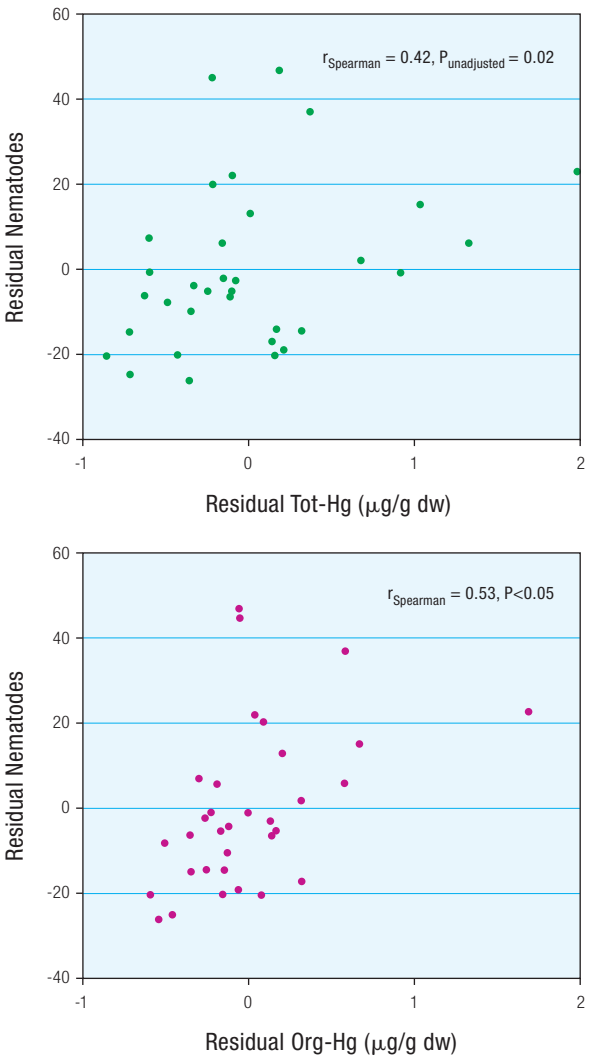

FIGURE 5.2.2

Gastrointestinal nematode numbers in relation to hepatic total and organic mercury concentrations in common eiders sampled at three Arctic locations (Holman on Victoria Island, East Bay Migratory Bird Sanctuary on Southampton Island, and the Belcher Islands) in 1997. To account for differences among sampling locations, residuals of parasite counts and mercury concentrations were determined by subtracting the mean value from individual values at each location.

The PHA-P skin response is a T-cell dependent immune response that is characterized by inflammation of the skin at the injection site. The inflammation results from localized leucocyte proliferation (Goto *et al.*, 1978; McCorkle Jr. *et al.*, 1980). Inflammation may also be related to the build-up of free radicals that are produced by the respiratory burst associated with phagocytosis (Peretz, 1989). While the well-known, stimulatory effects of selenium on cell-mediated immunity (Reilly, 1996) are consistent with the enhanced PHA-P skin response, its anti-inflammatory capability (Peretz, 1989)

should reduce inflammation, in apparent contrast to the results reported in this study. The effect of selenium on the PHA-P skin response likely depends on the extent to which the response reflects T-cell dependent leucocyte proliferation and the extent to which it reflects oxygen-free radical cell damage. The former effect will likely be enhanced by selenium within the range of nutritional adequacy while the latter effect will be dampened.

No evidence was found to indicate that exposure of eiders to mercury and cadmium affected their immune systems, yet these trace metals are known to be immunotoxic in experimental animals (Descotes, 1992; Pollard and Hultman, 1997). Either the immune function assays that were used herein were ineffective in demonstrating immunomodulation by mercury and cadmium exposure in these eiders or the concentrations of these trace metals to which the eiders were exposed were below threshold effect levels. In experimental mammals, immune function assays using PHA-P and SRBC as antigens have demonstrated that mercury and cadmium have the capacity to affect the immune system, usually by suppressing lymphocyte proliferation and hemagglutination (Blakley *et al.*, 1980; Hirokawa and Hayashi, 1980; Krzytyniak *et al.*, 1987; Dan *et al.*, 2000). Therefore, use of these antigens in immune function assays should have enabled effects to be detected on cell-mediated or humoral immunity in eiders by mercury and cadmium, if indeed, they occurred. It is probable that mercury and cadmium exposure in these birds was below levels that would likely affect skin swelling responsiveness to PHA-P and hemagglutination titers to SRBC. In support of the contention regarding mercury, the skin swelling reaction to PHA-P and antibody responses to bovine serum albumin and eastern equine encephalitis virus did not differ between captive great egret (*Ardea albus*) nestlings that were fed diets containing either high, medium or low concentrations of mercury (Spalding *et al.*, 2000a). Hepatic mercury concentrations averaged 15 mg/g, ww, in the medium-mercury group and 140 mg/g in the high-mercury group (Spalding *et al.*, 2000b), which are substantially higher than the concentrations of mercury in eiders in this study.

5.2.2 Black guillemot nestlings at Saglek, Labrador

Saglek Bay, on Canada's Labrador coast, has been the site of a military radar station since the late 1950s. Before remediation in 1997–1999, PCB-contaminated soil at the site was a source of contamination to the surrounding land as well as the inshore marine environment. PCB concentrations are elevated in sediments as well as the local benthic-based marine food web (ESG, 1999). The sediment PCB contamination decreases in concentration by several orders of magnitude with distance from the station. PCB concentrations in the benthic-based marine food web, including invertebrates, fish and diving seabirds, are highly correlated and relate directly to the concentrations in sediments (Kuzyk *et al.*, 2002). In 1999, an ecological risk assessment was initiated to provide a scientific basis from which to evaluate the need for sediment remediation (Kuzyk *et al.*, 2002).

A diving seabird, the black guillemot, was chosen as one focus of the assessment. Liver biomarkers and ΣPCB concentrations in 31 nestlings from three PCB exposure groups: reference group (range: 15–46 ng/g liver ww), moderately exposed Islands group (24–150 ng/g), and highly exposed Beach group (170–6,200 ng/g). Livers of female Beach nestlings were enlarged by 36% compared to livers of reference females. In both sexes, Beach nestlings had liver EROD (7-ethoxyresorufin-o-deethylase) activities elevated by 79% and liver retinol concentrations reduced by 47%. Retinyl palmitate concentrations were also reduced by 50% but only among female nestlings. Island nestlings also exhibited EROD induction (57%) and reductions in retinol and retinyl palmitate concentrations (by 28% and 58%, respectively). Liver lipid content increased with ΣPCB in both sexes, and correlated with liver mass in males. Malic enzyme activity and porphyrin concentrations showed minimal associations with ΣPCB. Although similar associations between liver biomarkers and OC exposure in fish-eating birds are well documented, exposures typically involve multiple contaminants and there is uncertainty about specific PCB effects.

The findings of this assessment indicate that liver biomarkers respond to relatively low PCB exposures (~73 ng/g, liver) in guillemots. These results are very significant and show that regional contamination in the Canadian Arctic can have an important impact on the physiology of local wildlife. Additional research is warranted at Saglek and other locations where PCBs, or other OCs, may be at levels greater than what is normally observed across the Canadian Arctic.

5.2.3 Mercury toxicity in beluga

Since the 1980s it has been known that levels of mercury in some beluga organs (e.g., liver, kidney) are high relative to human consumption guidelines (see Wagemann *et al.*, 1996; for data from the 1980s and 1990s). Risk of methylmercury poisoning in humans is assessed from levels measured in blood or hair. There has been no prior information for Arctic whales on the levels of mercury in the blood, or levels of methylmercury, a neurotoxin, in brain or spinal cord tissue. The first indication that blood and brain levels of mercury were high was reported by Hyatt *et al.* (1996) and Lockhart (1997). Subsequent measurements have confirmed the high levels. If whales

respond as other species (e.g., cats; Charbonneau *et al.*, 1974), there is enough mercury in some whales to cause biological injury. Methylmercury is the neurotoxic form of mercury and whale brain and spinal cord contain relatively little methylmercury as a proportion of total mercury. Since methylmercury is the predominant form in blood and since the source of mercury to brain and spinal cord is likely to be blood, it appears that the neurological organs must change the form of mercury from that delivered to that stored.

The tight correlation between mercury and selenium in most organs other than blood and muktuk suggests that whale organs may store mercury as a selenide. Hyatt *et al.* (1996) and Lockhart (1997) reported that liver slides contain dark staining objects in macrophages and these resemble deposits of mercuric selenide identified in other species (Nigro, 1994). Whales with high mercury levels contain more of these than whales with low mercury levels. The most complete description of the speciation of mercury in Arctic marine mammals was reported by Wagemann (1999) working with liver. Based on a fractionation procedure, this study identified mercuric selenide as a dominant form of mercury.

The data show that the whales accumulate high levels of mercury in several body organs including brain and spinal cord. However, the species of mercury present suggest that the whales may detoxify mercury by demethylating it and storing it as an inert mineral form, namely mercuric selenide. More definitive proof of this will have to be derived from experimental studies of the metabolic processing of methylmercury by whale organs. If this hypothesis is correct, it may represent an adaptation of the whales to allow survival in spite of accumulations of mercury. The most disturbing aspect of the data is the apparent increase in the amounts of mercury in the whales. If a detoxification mechanism exists in whale organs, then at what level does it become saturated? Also, is this mechanism present in other animals, even people?

5.2.4 Toxicity in polar bears

The polar bear has the highest concentrations of PCBs and chlordanes of any terrestrial mammal because of its almost exclusive diet of marine mammals. One of the more sensitive effects of PCBs in mammals is on retinol and thyroid hormone homeostatsis (Brouwer *et al.*, 1989). Vitamin A is important in growth and development, and in differentiation of epithelial cells (Morriss-Kay *et al.*, 1999). Thyroid hormones play a similar role in growth, differentiation and development, especially of the central nervous system and skeleton (Eberhardt *et al.*, 1980). It is therefore important that thyroid hormone and retinol toxicology be studied in the polar bear, especially since cub survival appears to be lower in areas with high PCB contamination. Many of the effects, such as induction of thyroid hormone conjugating and metabolizing enzymes and body stores of Vitamin A, are detectable only in the liver. However, blood is the only (relevant) tissue that is easily collected in living mammals, so considerable effort has been made to use circulating thyroid hormone and retinol levels in animals as a biomarker of both exposure and effects. Blood is the most suitable tissue for investigating interference with thyroid hormone transport by hydroxy-PCBs and other phenolic metabolites by competitive binding to thyroid hormone transport proteins (Lans *et al.*, 1994).

Concentrations of the thyroid hormones, T3 and T4, and retinol were determined in plasma of polar bears from Resolute Bay in Canada and Svalbard, which have among the lowest and highest OC concentrations, respectively, in polar bears (Norstrom *et al.*, 1998). Free T3 (FT3) and free T4 (FT4) indices were also determined. These indices are the inverse of the relative binding capacities of the plasma samples for these hormones, and have been shown to correlate highly with the free hormone concentrations in fish (Eales and Shostak, 1985). The OC and hydroxy metabolite concentrations were also determined in these plasma samples.

Resolute bears had significantly higher total T4, and FT4 index, and lower total T3 and free T3 index than Svalbard bears. Retinol concentrations were not significantly different between regions (Figure 5.2.3). None of the biological measures were significantly related to age, even when separated into region and gender categories.

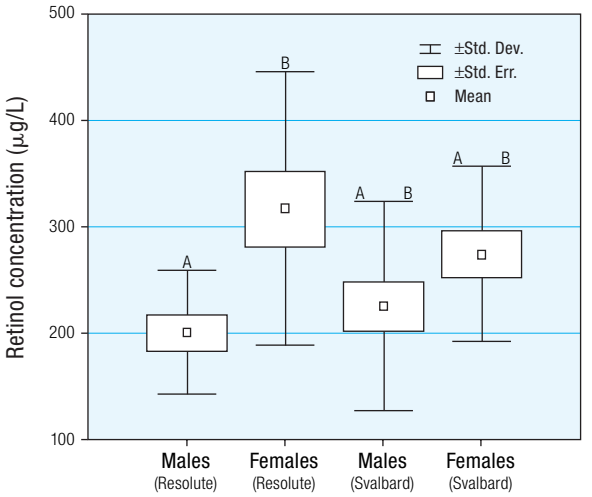

FIGURE 5.2.3

Box plots showing gender differences in retinol concentrations in polar bear plasma from Resolute Bay in the Canadian Arctic in April–May 1997, and from Svalbard in April–May 1998. Adult males and females, n = 12–15 in each category. Categories not sharing a letter (a,b) have significantly different concentrations, $p < 0.01$.

This is in contrast to another study which found that total T4, FT4, total T3 and FT3 are associated with age in male polar bears (Skaare *et al.*, 2001). Gender differences in retinol and thyroid hormone measures for each of the populations are also given in Table 5.2.1. Females had higher total T4 than males in both populations (Figure 5.2.3). Total T3 was higher in females from the Svalbard population and FT3 index was lowest in Resolute females. T4 concentrations reported by Leatherland and Ronald (1981) in male polar bears were much higher, mean of 74 nmol/L. This could be due to seasonal differences, since the latter value was for male bears in October, whereas the present samples and those of Skaare *et al.* (2001) were taken in March. Leatherland and Ronald (1981) indicated that seasonal differences were small; thus, there appears to be disagreement between the current and earlier results for T4.

Correlations among thyroid hormone concentrations, FT3 and FT4 indices, retinol concentrations and the complete suite of POP concentrations in adult polar bear plasma (N = 60) from Resolute Bay, Canada and Svalbard were examined by principal component analysis. Because hydroxy compound may affect circulating levels of free and bound T4 due to competitive binding of hydroxy metabolites and T4 to TTR, all chemical concentrations were converted from mass to molar concentrations before statistical analysis.

Four statistically significant principal components (PCs) were identified explaining 65% of the variance. Compounds which loaded highly on one PC ($r > 0.70$) and low on any other ($r < 0.30$) were considered uniquely related to that principle component. The principal components were clearly related to chemical class. PC1 was modeled entirely by concentration of persistent PCBs, PC2 by OH-PCBs, PC3 by non-persistent PCBs (i.e., that have a low biomagnification factor relative to CB153), and PC4 by chlordanes (decreasing order of explanation of variance).

Using the whole data set (N = 60), retinol concentrations were negatively correlated with PC1 (persistent PCBs) and positively correlated with PC2 (OH-PCBs) (Figure 5.2.4). If only Resolute bears (n = 25) were included in the analysis, retinol was more highly correlated with PC1 and PC2. These results suggest that plasma retinol concentrations are more likely to be affected by the influence of persistent PCBs on retinol metabolism and storage in liver than by interference of OH-PCBs with the transport of retinol via Retinol Binding Protein:TTR dimer formation.

Total T4 plasma concentrations were negatively associated with both PC1 (persistent PCBs) and PC3 (non-persistent PCBs), but not with any other contaminant group, including OH-PCBs. Since both PC1 and PC3

TABLE 5.2.1 Thyroid hormone concentrations, free hormone indices and retinol concentrations in polar bear plasma from Resolute Bay in the Canadian Arctic (1997) and Svalbard (1998)

	Resolute cubs (N = 3)		Resolute juveniles (N = 5)		Resolute male adults (N = 12)		Resolute female adults (N = 13)	
	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Age (y)	(0–2)		(3–4)		10	4	14	7
T3 (nmol/L)	0.21	0.21	0.08	–	0.19	0.19	0.15	0.08
FT3 Index	4.57	1.74	2.91	0.95	5.19	1.34	2.81	1.59
T4 (nmol/L)	8	2.75	6.25	0.41	4.93	4.05	6.62	2.06
FT4 Index	1.08	0.09	1.17	0.03	1.13	0.04	1.15	0.04
Retinol (μmol/L)	1.16	0.33	1.17	0.5	0.7	0.2	1.08	0.46
	Svalbard cubs (N = 3)		**Svalbard juveniles (N = 2)**		**Svalbard male adults (N = 18)**		**Svalbard female adults (N = 15)**	
	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Age (y)	(0–2)		(3–4)		14	6	14	4
T3 (nmol/L)	0.23	0.13	0.69	0.67	0.34	0.3	0.59	0.36
FT3 Index	6.18	0.15	6.24	0.05	6.31	0.08	6.25	0.09
T4 (nmol/L)	1.15	1.7	0.39	0.25	2.33	1.87	3.88	4.82
FT4 Index	0.79	0.11	0.75	0.14	0.93	0.18	0.84	0.18
Retinol (μmol/L)	1.22	0.26	0.88	0.43	0.79	0.34	0.96	0.29

are PCB related, this correlation suggests a common mechanism of action of all PCB congeners in reduction of plasma T4 concentrations. Total T4 concentrations were also negatively correlated with concentration of ΣPCB. PCBs have been shown to reduce total plasma T4 concentrations in rats (Goldey *et al.*, 1995), probably due to increased peripheral T4 metabolism (Morse *et al.*, 1993). T4 concentrations in plasma can be influenced by OH-PCBs at the thyroid hormone transport protein level since many OH-PCBs have been shown to bind to TTR with high affinity relative to T4 (Lans *et al.*, 1993). There was no evidence of this in polar bears.

The FT4 index was negatively correlated to PC2 (OH-PCBs) using the whole data set. When separated by gender, only male bears showed significant correlation between the FT4 index and PC2. When separated by region, the FT4 index was no longer correlated to PC2. The index is an indirect measure of FT4 concentrations that depends on preservation of TTR binding capacity in the sample rather than preservation of T4 itself. There was significantly more scatter in the FT4 index in the Svalbard samples that had been handled several times and stored frozen under unknown conditions. The Resolute samples were stored immediately at -40°C or colder from the time of sampling until they were thawed

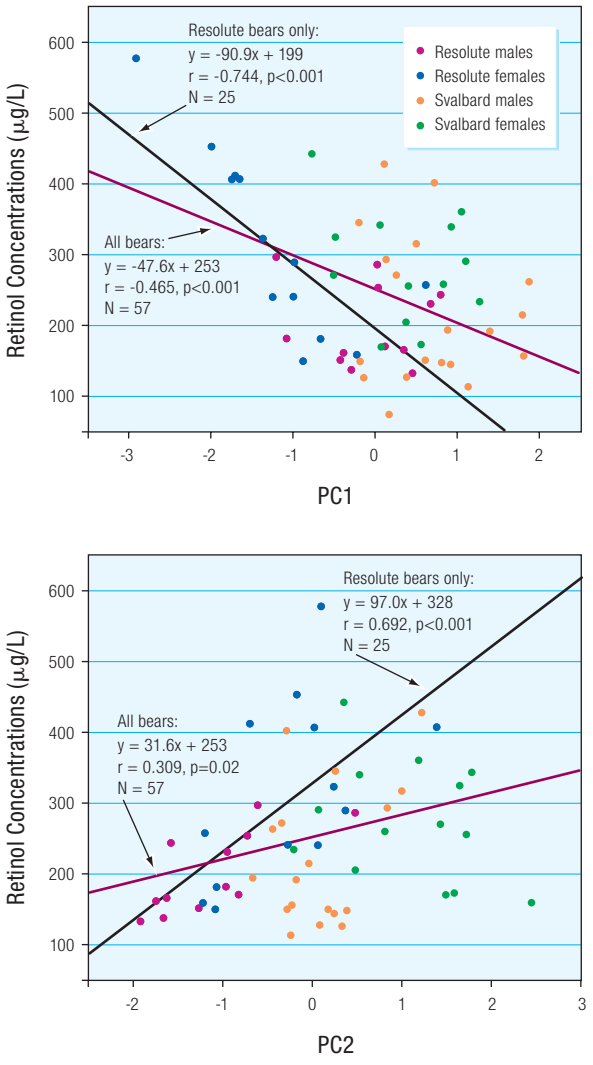

FIGURE 5.2.4

Correlation between retinol concentrations (µg/L) and the first two principal components from analysis of POP concentrations in polar bear plasma from Resolute Bay in April–May 1997, and from Svalbard in April–May 1998. PC1 represents persistent PCBs and PC2 represents OH-PCBs.

for analysis. The correlation between the FT4 index and PC2 may therefore be an artifact of sample preservation.

Decreasing plasma retinol concentrations with increasing PCB exposure seems to be a well-established effect for many species in the literature. The polar bear is also subject to this effect. There is little consistency in interpretation of circulating thyroid hormone concentrations and correlation of POP exposure, or even different studies in the same species. This is undoubtedly due to the complex nature of thyroid hormone homeostasis, e.g., production of thyroid-stimulating-hormone (TSH) activity (induction and inhibition) of hormone producing and metabolizing enzymes (iodinases, deiodinases, UDP-GT, sulfotransferases), seasonal variation, and so on. Specific (TTR) or non-specific (globulins) protein transport also varies among species. Although there are some hints from studies to date that there are effects of PCB exposure on thyroid hormone status in polar bears, much more research is needed before any conclusions can be drawn.

5.3 Comparison of thresholds of biological effects and concentrations observed in Canadian Arctic biota

5.3.1 Organochlorine concentrations and threshold levels of effects

An extensive review of relevant threshold levels for effects of OCs in Arctic biota was carried out for the second AMAP report on POPs (de Wit *et al.*, 2003; in press). This report also compared all circumpolar data for Arctic organisms to these thresholds, including Canadian data. The amount of data for Canadian species is more limited than what is available for the circumpolar Arctic both in terms of the organisms examined and the chemicals measured. Relevant threshold levels available for PCBs, DDT and dioxin-like chemicals for Arctic fish, seabirds and marine mammals are provided in Table 5.3.1. These chemicals were chosen because they are generally found at the highest concentrations in Arctic biota and the greatest amount of threshold level information has been generated for these chemicals. The species examined have been limited to those with the greatest OC concentrations. Levels of OCs in terrestrial animals and invertebrates are generally low and considered to be at levels that are below concern for wildlife health.

Fish

Despite a large amount of research on PCBs and DDT in fish there are few government guideline levels for assessing effects on the fish based on their tissue levels. There are a number of guidelines based on sediment concentrations, which are difficult to apply in this assessment because of very limited data at sites where fish or marine samples were collected. Environment Canada has established guideline levels for ΣDDT and ΣPCB for the protection of wildlife that consume fish at 14 ng/g ww and 0.79 ng TEQ/g ww (mammalian diet), respectively (Environment Canada, 2002). For the purposes of this assessment the guideline for PCBs was converted to 15 ng/g ww (for ΣPCB) based on the fraction of TEQs found in Arctic char of 5 x 10^{-5} of ΣPCB (Muir, 2002; unpublished data). However, these guidelines are not for the protection of the fish themselves. Recent work on the effects of PCBs in Arctic char provides a lowest-observed-effect level (LOEL) for EROD induction at 1,000 ng/g ww in liver (Jørgensen *et al.*, 1999). This is similar to levels observed to cause EROD

TABLE 5.3.1 Threshold levels of biological effects for ΣPCB, ΣDDT and TEQs for dioxin-like chemicals for fish, seabirds and marine mammals. These values were compiled for the second AMAP assessment of POPs in Arctic biota (de Wit *et al.* 2002). NOEL – No-observed-effect-level; LOEL – Lowest-observed-effect-level

	Lowest NOEL	Effect	Reference	Lowest LOEL	Effect	Reference
Fish						
ΣPCB				1,000 ng/g ww in liver	EROD induction	Jørgensen *et al.* (1999)
ΣDDT						
TEQ						
Seabirds						
ΣPCB	2.3 μg/g ww	Hatching effect	Giesy *et al.* (1994b), Barron *et al.* (1995)	3.5 μg/g ww	Egg mortality	Giesy *et al.* (1994b), Barron *et al.* (1995)
ΣDDT				15–20 μg/g ww	Egg shell thinning	Peakall *et al.* (1990)
TEQ	4.6 pg TEQ/g ww	Reproduction	Giesy *et al.* (1994b)	10 pg TEQ/g ww	Reproductive effects	Giesy *et al.* (1994b)
Marine mammals						
ΣPCB	4,000 ng/g lw	Vitamin A reduction in seals	Reijnders (1986), Boon *et al.* (1987)	11,000 ng/g lw	Vitamin A reduction	Murk *et al.* (1998)
ΣDDT						
TEQ	84 pg TEQ/g ww	Vitamin A reduction in otter	Murk *et al.* (1998)	490 pg TEQ/g ww in liver	Mink kit survival	Heaton (1992)

induction in Great Lake fish. Luxon *et al.* (1987) observed EROD induction in Lake Ontario lake trout that had ΣPCB concentrations of 3,000 ng/g but not in Lake Huron lake trout which had concentrations of 2,000 ng/g. Further laboratory studies of Arctic char have found that single doses of PCBs as low as 1,000 ng/g body weight affect disease resistance and stress responses in starved specimens (Jørgensen *et al.*, 2002; in press).

There is a fairly substantial database for the effects of dioxin-like chemicals (PCDDs, PCDFs and coplanar PCBs) in fish and in particular for EROD induction. There have been no new data produced for these chemicals, however, since publication of the first CACAR. It was reported in the first CACAR that EROD levels did not vary across the Canadian Arctic in freshwater fish, even in lakes, such as Lake Laberge, that were more contaminated (Muir *et al.*, 1997). These EROD levels were lower than levels measured in Lake Ontario lake trout. Since EROD induction is a conservative threshold for the effects of dioxin-like chemicals it would seem unlikely that these chemicals are having a significant influence on the health of Canadian Arctic fish.

Figure 5.3.1 shows that for the exception of burbot from Lake Laberge and Greenland shark, levels of ΣPCB in Arctic fish are well below the LOEL for EROD induction. Although the levels of PCBs in the burbot and Greenland shark cross this threshold it does not confirm that the health of these fish are compromised by PCBs. Although EROD induction is correlated to toxic effects (Safe, 1994) there are many biological variables associated with induction of EROD (Stegeman, 1979; Addison and Willis, 1982). There is very little information for the Greenland shark on EROD induction and the levels and effects of PCBs and other contaminants on sharks (Fisk *et al.*, 2002a). Further both of these species have high lipid livers, which may reduce the effects of contaminants (Geyer *et al.*, 1993).

Levels of PCBs and DDT in Arctic fish do, in general, exceed the Environment Canada guidelines for the protection of wildlife that consume the fish. This does not suggest that the PCBs and DDT are influencing the health of the fish. As well, these guidelines are conservative and are 5 times lower than levels established by the US Environmental Protection Agency (EPA). Levels of potential prey items of these Arctic fish, generally zooplankton and smaller fish, are well below these guideline levels. The exception may be the Greenland shark that commonly feeds on marine mammals (Fisk *et al.*, 2002a).

In light of the fact that levels of PCBs and DDTs in most Canadian Arctic fish are well below the most conservative levels for possible biological effects, the potential for OCs to cause biological effects in Arctic fish of Canada is low. There are a small number of cases where OCs may be causing some biological effects, in particular Greenland shark, but the threat should not be considered major. There are also a number of lakes, e.g., Lake Laberge, where levels of OCs have been and are higher but population level effects have not been observed.

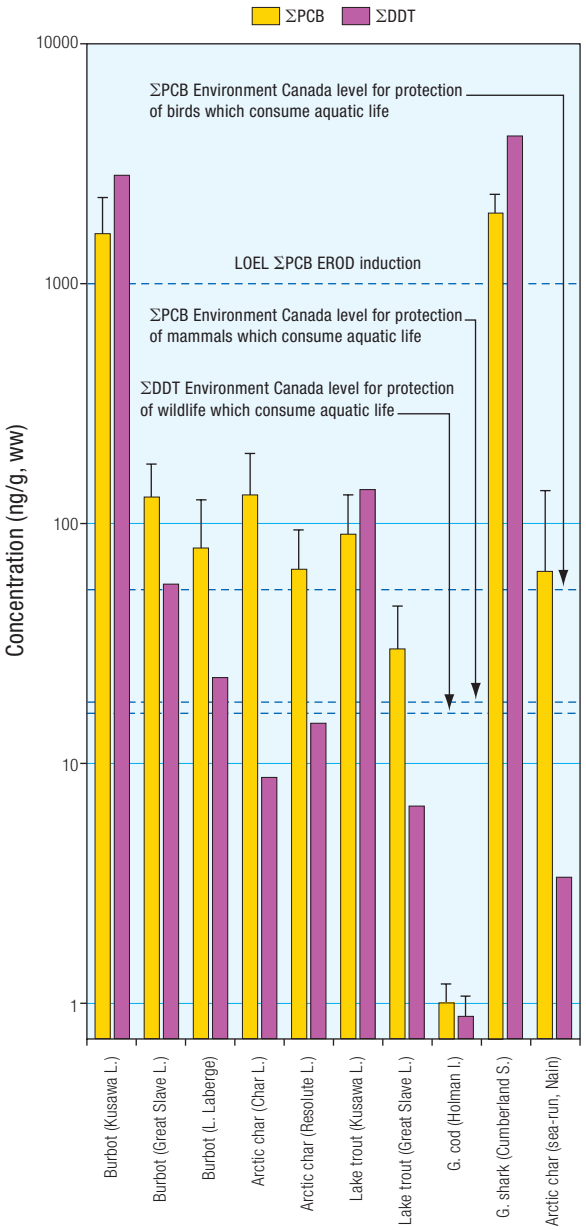

FIGURE 5.3.1

ΣPCB and ΣDDT concentrations in freshwater and marine fish compared with threshold effects levels and Environment Canada guidelines for the protection of wildlife (Environment Canada, 2002). Assumes average PCB TEQ = 5 x 10^{-5} of ΣPCB which is the ratio found in Arctic char (Muir, 2002; unpublished data). LOEL for PCB EROD induction is from Jorgensen *et al.* (1999). Due to numerous limitations in the threshold data, in quantification of PCBs and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

Seabirds

A fair amount of research has been carried out on the effects of OCs in birds. This in part due to the strong effect of DDT on egg-shell thinning and the reduction in the populations of birds of prey due to DDT. There are solid no-observed-effect-levels (NOELs) and LOELs for seabirds. As can be seen in Figure 5.3.2, levels of ΣPCB and ΣDDT in the livers of Arctic seabirds are well below the NOEL and LOEL levels. DDT and PCBs have the highest concentrations of any OCs in seabirds and it would seem unlikely that other OCs are at levels associated with biological effects. The seven species of seabirds studied (Figure 5.3.2) cover the entire range of trophic levels and OC concentrations found in seabirds from the Canadian Arctic.

A recent comparison of OC levels in Arctic seabird eggs with threshold effects levels found that only PCB levels in the eggs of glaucous gulls ranged within the LOEL established for hatching success in white-leghorn chicken (Muir *et al.*, 1999b). This threshold level (approximately 1,000 ng/g ww) is higher than any levels found in the liver of adult seabirds. The chicken is very sensitive to the effects of OCs, and therefore may not be a good surrogate for assessing the effects of OCs in seabirds.

Toxic equivalency factors (TEQs) were recently determined for the liver and eggs of seabird species collected 1975 and 1993 (Annex Table 13). These TEQs were based on concentrations of dioxins (PCDDs), furans (PCDFs) and non-*ortho* PCBs. As can be seen in Figure 5.3.3, TEQ concentrations surpassed the NOEL and LOEL levels established for relevant seabird species including herring gulls and coromorants. In particular, levels in northern fulmar far exceed these threshold levels. As was reported in Section 3.3.2.2, these TEQ values were much higher than levels observed in marine mammals. Further investigation of dioxin-like chemicals in seabirds is warranted.

A second method for assessing the possible effects of OCs in seabirds is to examine levels of OCs in their diet. Environment Canada has chosen guidelines of 14 ng/g ww for ΣDDT and 2.4 ng TEQ/g ww for PCBs as the levels in aquatic organisms that are safe for the protection of birds that consume these organisms (Environment Canada, 2002). For purposes of this assessment, the guideline for PCBs was converted to 48 ng/g ww ΣPCB based on the fraction of PCB TEQs found in Arctic char of 5 x 10^{-5} (Muir, 2002; unpublished data). The US EPA has established less conservative levels that are approximately five times higher. Many of the seabirds, including dovekie, black guillemots, thick-billed murres and black-legged kittiwakes consume zooplankton and small marine fish. Levels of PCBs and DDT in these organisms are well below these guideline levels. The glaucous and ivory gulls and northern fulmar are known to scavenge dead marine mammals and may at times consume food that is above these threshold levels.

The threshold effects levels in tissue chosen for seabirds were among the most conservative and no levels were found that exceed them, it can be concluded that the

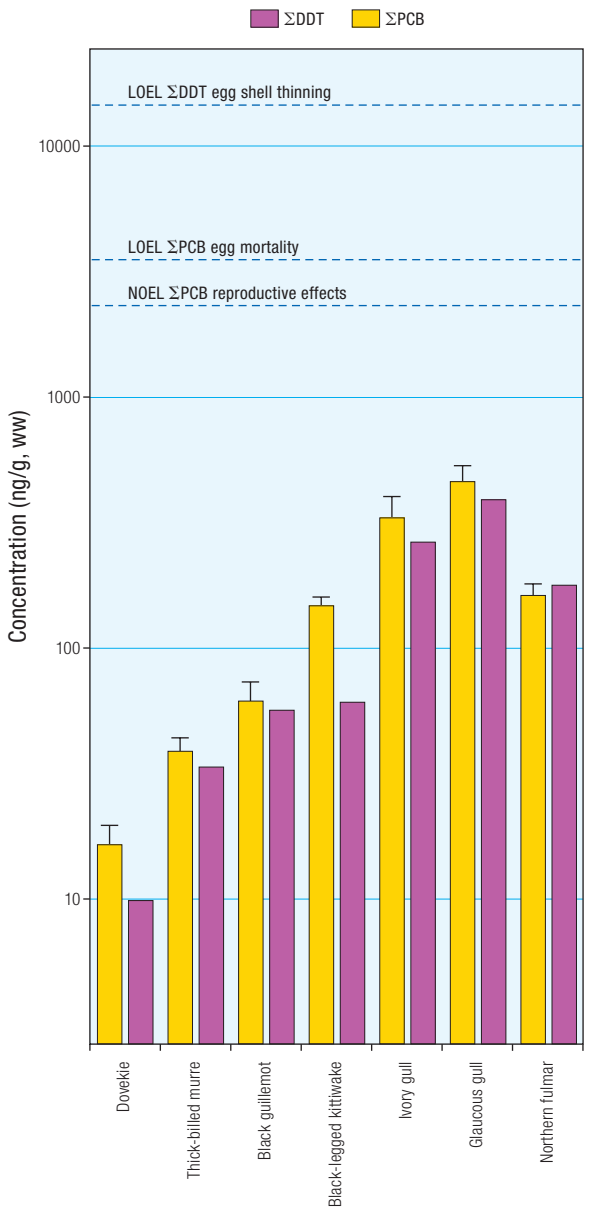

FIGURE 5.3.2

ΣPCB and ΣDDT concentrations in the livers of seabirds compared to threshold effects levels. LOEL for DDT is from Peakall *et al.* (1990), LOEL and NOEL for PCBs are from Giesy *et al.* (1994) and Barron *et al.* (1995). Due to numerous limitations in the threshold data, in quantification of PCBs and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

potential for OCs to cause significant biological effects in seabirds is low. Additional work, however, is warranted on dioxin-like chemicals in seabirds.

Marine mammals

PCBs are the dominant OC found in marine mammals and, similar to seabirds, a number of studies are available for establishing the threshold effects levels for PCBs in marine mammals. In particular laboratory studies have been carried out on the effects of PCBs on captive seals (Reijnders, 1986; Boon *et al.*1987). A number of Arctic marine mammals have levels of ΣPCB in blubber or fat that were near or above the NOEL for vitamin A reduction in seals, effects on reproduction in otter, and effects on visual memory in Rhesus monkeys (Figure 5.3.4). Although this suggests that PCBs may be causing biological effects in some species caution is warranted. No levels were found that exceeded the LOEL for vitamin A reduction in seals or immune effects in Rhesus monkeys (Figure 5.3.4). The effect of a reduction of vitamin A on the health of a marine mammal population is unknown. These threshold levels were established in the liver of the seal, whereas most of the levels presented in Figure 5.3.4 were determined in fat or blubber, which tend to sequester OCs and likely reduce their potential biological effects.

As with seabirds, an assessment of the possible effects of OCs in marine mammals can be made by examining levels of OCs in their diet. Environment Canada (2002) has chosen 14 ng/g ww for ΣDDT and 0.79 ng TEQ/g ww for PCBs (or 15 ng/g ww for ΣPCB assuming the fraction of TEQs in Arctic char as discussed previously) as the levels in aquatic organisms that are safe for the

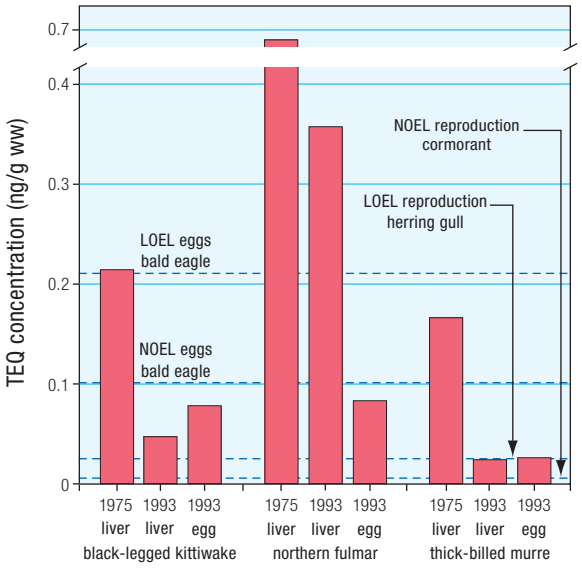

FIGURE 5.3.3

Toxic equivalent (TEQ) concentrations in the livers of seabirds compared to threshold effects levels. LOEL reproduction for herring gulls and NOEL reproduction for cormorants are from Giesy *et al.* (1994), NOEL for bald eagle eggs are from Elliott *et al.* (1996) and LOEL for bald eagle eggs are from Elliott *et al.* (1996) and Kubiak *et al.* (1989). Due to numerous limitations in the threshold data, in quantification of PCBs and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

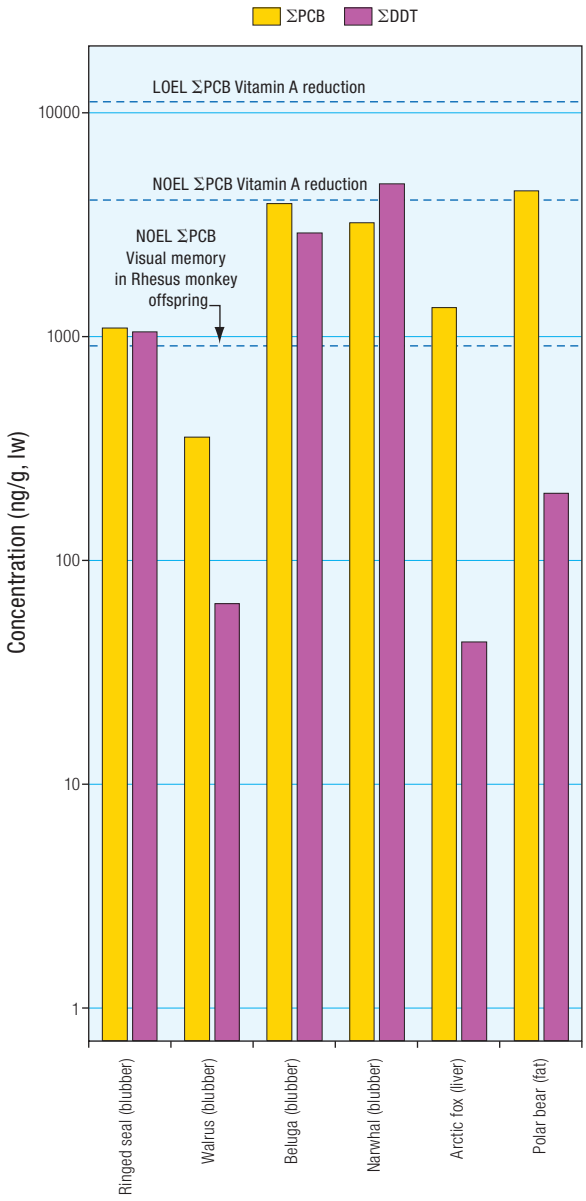

FIGURE 5.3.4

ΣPCB and ΣDDT concentrations in marine mammals compared to threshold effects levels. LOEL for PCB vitamin A reduction is from Murk *et al.* (1998), NOEL for PCB vitamin A reduction is from Reijnders (1986) and Boon *et al.* (1987), and NOEL for PCB visual memory in Rhesus monkey offspring is from Shantz *et al.* (1991). Due to numerous limitations in the threshold data, in quantification of PCBs and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

protection of organisms that consume these organisms. Seals and walrus generally consume zooplankton, benthic invertebrates and small fish (e.g., Arctic cod), although this varies with the species of pinniped. These organisms have OCs concentrations that are below the guideline levels. One exception would be walrus that consume ringed seals. These individuals have been shown to have higher OC levels (Muir *et al.*, 1995). Beluga and narwhal consume zooplankton and small pelagic fish, which have levels of OCs below the Environment Canada guidelines, but also consume benthic fish, of which some species exceed the guidelines. Polar bears are clearly feeding on diet items, i.e., seal blubber, that are well above the Environment Canada guidelines. The greatest concern is for possible biological effects of OCs in polar bears from the Canadian Arctic in light of recent studies suggesting that PCBs may influence their immune and thyroid hormone systems.

5.3.2 Metals concentrations and threshold levels of effects

Threshold levels of effects for heavy metals on Arctic wildlife were established for the AMAP assessment of metals in the Arctic (Dietz *et al.*, 1998). These threshold levels are to be used in the second AMAP assessment of metals in the Arctic (AMAP, 2003; in press) and are summarized in Table 5.3.2. This report has restricted the metals examined to mercury, cadmium and selenium based on the relative amount of data available for the Canadian Arctic and the level of concern for these metals.

Terrestrial animals

Concern has been raised regarding the levels of heavy metals in terrestrial animals, particularly in caribou. Unlike OCs, metals reach levels in terrestrial mammals that are similar or greater than those seen in marine mammals. New data for metals in the terrestrial Arctic is limited to cadmium and mercury in caribou. Mean concentrations of mercury and cadmium in the kidney of five caribou herds are below the threshold levels for biological effects established for Arctic organisms by the second AMAP report (AMAP, 2003; in press) (Figure 5.3.5). The range of cadmium concentrations in two of the caribou herds exceeded the threshold level established for cadmium and warrants continued monitoring. Past assessments of cadmium in kidney and liver of caribou, however, concluded that levels were comparable to findings in other big game species of Canada and should be considered natural given the general elevation on a circumpolar basis (Elkin, 1997).

Fish

As with OCs, few guidelines have been established for the protection of fish from metals. Guidelines are available for the fish consumers, both human and wildlife. Mercury levels in freshwater fish surpass the guidelines for human subsistence or commercial sale in many lakes and for may species (Figure 3.1.4).

TABLE 5.3.2 Threshold levels of biological effects for cadmium, mercury and selenium for birds and mammals as complied by Dietz *et al.* (1998)

Metal	Group	Tissue	Concentration mg/kg ww	Effect	Reference
Cadmium	Birds	Liver	> 40	Cadmium poisoning	Furness (1996)
		Kidney	> 100	Cadmium poisoning	
	Mammals	Liver	> 20–200	Potential renal dysfunction	Law (1996)
		Kidney	> 50–400	Potential renal dysfunction	
Mercury	Birds	Liver	> 30	Lethal level in free ranging birds	Thompson (1996)
		Kidney	> 30	Lethal level in free ranging birds	
		Egg	> 3.0	Detrimental effect upon free ranging bird hatching	
	Marine mammals	Liver	> 60	Liver damage	Law *et al.* (1996)
	Terrestrial mammals	Liver	> 25	Laboratory succumbed animals due to mercury intoxication	Thompson (1996)
		Kidney	> 25	Laboratory succumbed animals due to mercury intoxication	Thompson (1996)
Selenium	Birds	Liver	> 9	Deformed embryos	Heinz (1996)
		Kidney	> 9	Deformed embryos	
		Egg	> 3	Deformed embryos	
	Terrestrial mammals	Liver	> 7	Hepatic lesions	WHO (1987)

Seabirds

A fairly large database on metals in seabirds is available for assessment of potential biological effects. Using the guidelines established by AMAP (Table 5.3.2) levels of cadmium and mercury in the liver of eight seabird species are below threshold levels for biological effects (Figure 5.3.6) (Dietz *et al.*, 1998). Levels of selenium in the livers are at or near threshold levels for causing deformities in embryos, but this should be considered a very conservative threshold level. These results are consistent with the second AMAP assessment of metals in the Arctic, which concluded that there was little evidence of biological effects of heavy metals in seabirds (AMAP, 2003; in press). As well, Wayland (2001, and Section 5.2.1) found no evidence that metals were influencing the health of eiders.

Marine mammals

Metal data are available for a number of marine mammal species, and for ringed seals and beluga from a number of locations. Levels of mercury and cadmium in ringed seals and walrus are below threshold levels established by the AMAP report on metals in the Arctic (Dietz *et al.*, 1998) (Figure 5.3.7).

5.4 Assessment of biological effects work in Canadian Arctic biota

The lack of information on the biological effects of OCs and metals in Arctic biota was identified as a major data gap in the first CACAR assessment (Muir *et al.*, 1997). Since that time, there have been few biological effects related studies on Canadian Arctic biota, most likely because of a low level of concern and the difficulty and expense of carrying out appropriate studies in the Arctic.

There is currently little or no evidence to suggest that OCs or metals are having a widespread negative effect on the reproduction or survival of any Arctic wildlife. It has been shown, however, that where local contamination has occurred, there is the possibility that physiological effects can be observed in wildlife. These effects were observed in seabirds that were only moderately highly contaminated with PCBs. These effects are important and warn against the assumption that anthropogenic contaminants are not influencing the health of Arctic

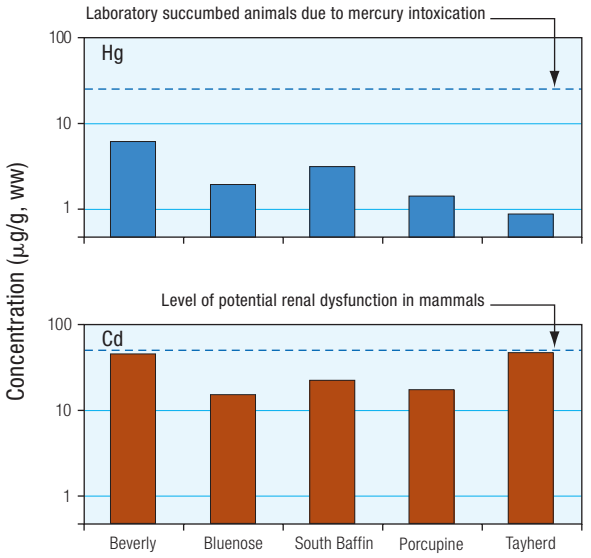

FIGURE 5.3.5

Mercury and cadmium concentrations in caribou compared to threshold effects levels. Mercury and cadmium threshold levels are from Thompson (1996) and Law (1996), respectively. Due to numerous limitations in the threshold data and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

animals. As well, with impending climate warming, and possible effects on contaminant dynamics, and the discovery of a range of new contaminants, several of which are increasing in the Arctic, there is sufficient reason to continue monitoring of the health of Arctic wildlife populations and possible biological effects of anthropogenic contaminants.

A recent study on physiological effects in seabirds of elevated PCBs in the Saglek Bay region demonstrates that elevated levels of PCBs can elicit effects. This study found that seabirds from areas with regional contaminants had PCB levels that were 1 to 2 orders of magnitude higher than reference areas, which caused enzyme induction and changes in the vitamin economy of these birds. This study demonstrates the possible effects of local contamination but also suggests that seabirds with the higher contaminant burdens, such as the glaucous gulls, may be susceptible to the effects of contaminants.

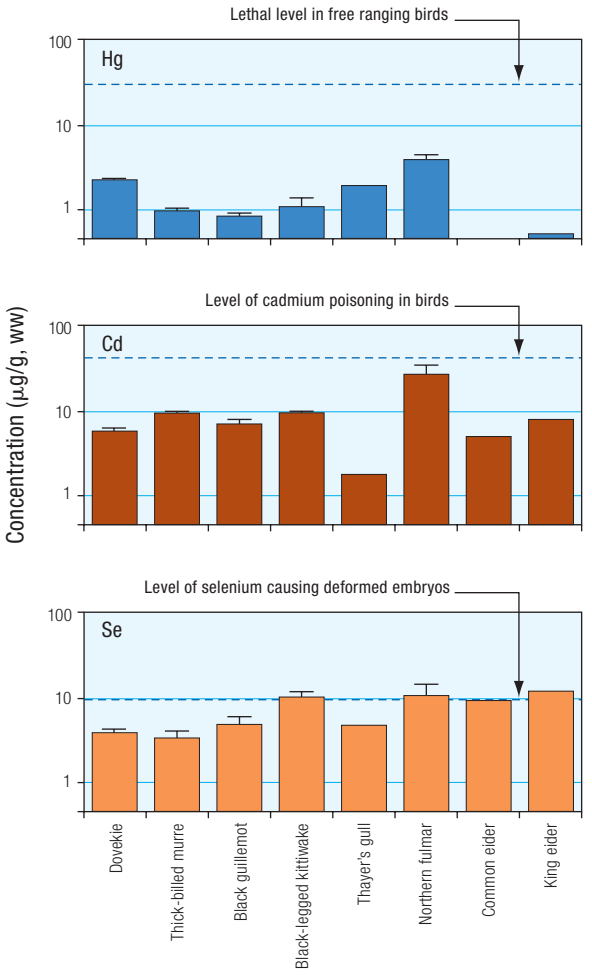

FIGURE 5.3.6

Mercury, cadmium and selenium concentrations in seabirds compared to threshold effects levels. Mercury, cadmium and selenium threshold levels are from Thompson (1996), Furness (1996) and Heinz (1996), respectively. Due to numerous limitations in the threshold data and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

A comprehensive study examined the possible links between trace metal concentrations and health-related biomarkers in sea ducks (common and king eiders) (Wayland, 2001). Concern about the health and size of sea duck populations has increased and the effects of metals have been suggested as a potential factor. The biomarkers tested included body condition, parasitic infestations and immune function but little evidence for a relationship between sea duck health and metal levels was found. Mercury levels in the livers of sea ducks were related to body mass in three of the four years studied and in abdominal fat mass in two of the four years studied. Cadmium levels in liver, after removing the influence of mercury, were related to body weight in one year of the study. However, laboratory studies of cadmium and mercury exposure in ducks did not support these relationships and it was concluded the gain and loss of body weight may have influenced the metal concentrations measured. Mercury and cadmium did not influence the immune system of the sea duck although selenium levels were related to one of the two tests of the immune systems carried out.

Recent studies on levels of thyroid hormones and retinol levels in Canadian (Resolute) and Svalbard polar bears were carried out to assess the possible effects of PCBs and their metabolites. Levels of PCBs are higher in the Svalbard bears than in the Resolute bears providing a good dose-response relationship. Such dose-response relationships are critical for determining if biological effects are related to exposure. Thyroid hormone levels in the blood varied between the Resolute and Svalbard polar bears but retinol levels did not. Retinol was correlated to levels of PCBs and PCB metabolites (mainly OH-PCBs), suggesting some interference in the retinol economy of the polar bears. Blood levels of the thyroid hormone, T4, were correlated to levels of PCBs but not PCB metabolites.

The relationships observed between the PCBs and PCB metabolites and thyroid and retinol economy are a concern and warrant further research. Although effects are likely greater in the more contaminated Svalbard bears, the Resolute bears fall within the relationship. As well, immune and hormone effects related to PCB levels have also recently been reported in Svalbard polar bears (Li *et al.*, 2002a). Recent studies on Canadian polar bears have also suggested that changing ice conditions can affect the recruitment and population characteristics and size of polar bears (Stirling *et al.*, 2000). Forecasted

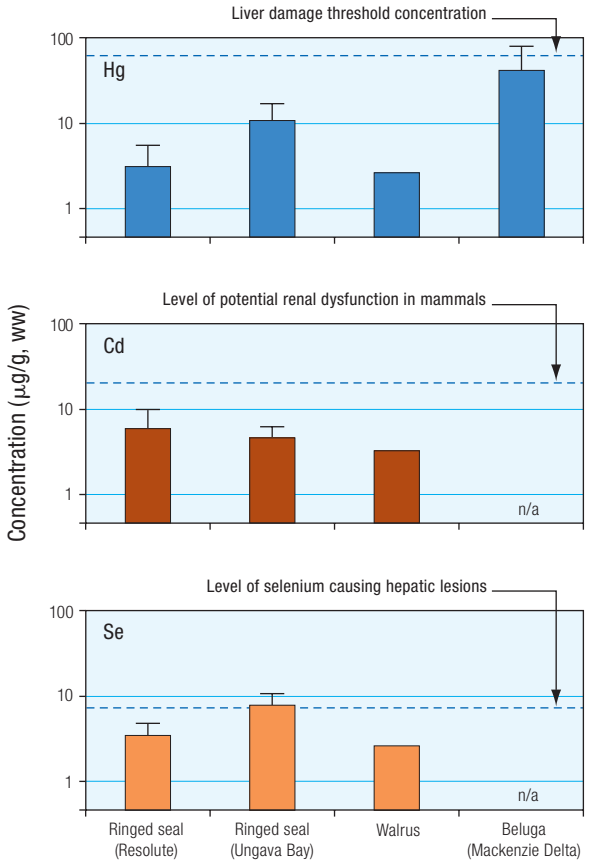

FIGURE 5.3.7

Mercury, cadmium and selenium concentrations in marine mammals compared to threshold effects levels. Mercury and cadmium threshold levels are from Law (1996) and for selenium are from WHO (1997). Due to numerous limitations in the threshold data and problems with extrapolating such data across tissues and species, this comparison should be used with caution.

changes in climate and ice conditions in the Canadian Arctic may result in increased significance of stress related to anthropogenic contaminants in polar bears.

Few examples were found of current levels of OCs in Canadian wildlife exceeding threshold effects levels. In the rare cases where levels in organisms did exceed threshold levels, the threshold levels were conservative and were generally established as NOELs. This would suggest that currently the threat of OCs on the health of Arctic wildlife is low. The second AMAP assessment of POPs in the circumpolar Arctic (de Wit *et al.*, 2003; in press) made a similar conclusion regarding Canadian wildlife. However, levels of dioxin-like chemicals in seabird eggs and liver did exceed a number of threshold levels and warrant further study.

As with OCs, few examples were found of current metals concentrations measured in Canadian wildlife exceeding threshold effect levels. Although levels of metals are sometimes found to be high in Arctic wildlife, particularly marine mammals, evidence of possible biological effects is lacking. In fact studies of Arctic marine mammals have concluded that high levels of cadmium in kidney are not posing a threat to this species (Sonne-Hansen *et al.*, 2002). There is evidence, however, that warmer temperatures can result in higher tissue levels of metals in fish and beluga. Monitoring of Arctic wildlife health, in conjunction with continued monitoring of trace metal concentrations is recommended.

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Conclusions, Knowledge Gaps and Recommendations

A large amount of data on contaminants in Canadian Arctic biota have been produced since the first CACAR was published in 1997. However, data and knowledge gaps remain; including some that were identified in the first CACAR and new ones that have arisen.

6.1 Conclusions

6.1.1 Priority chemicals

There are a large number of priority chemicals measured in Canadian Arctic wildlife, including approximately 100 legacy organochlorines (OCs) (including PCBs, DDTs, chlordanes, HCHs and CBz) and in some studies up to 25 metals. *The "legacy" OCs, as well as mercury, cadmium and selenium have continued to be the focal point of the measurement and monitoring of contaminants in the Canadian Arctic.*

New information on toxaphene has been generated for freshwater fish¸ seabirds and marine mammals, addressing a data gap identified in the first CACAR. Analytical methods for toxaphene have improved with the measurement of individual chlorobornane congeners and lab performance has been assessed as part of the NCP interlaboratory studies. New data for PCDD/Fs and non-*ortho* PCBs have been generated for seabirds and have shown that PCDD/F levels in northern fulmar were higher than those observed in marine mammals.

A series of new chemicals have been found in Arctic biota. The number of new chemicals measured has increased since the first CACAR. These data have provided more comprehensive coverage for Arctic organisms, including temporal trend data sets, compared with the first CACAR but the scope is very limited, in comparison to legacy OCs. In particular, information on polybrominated diphenyl ethers have been generated for both freshwater and marine organisms while measurement of short-chain chlorinated paraffins (SCCPs) and chlorinated naphthalenes were made in beluga and ringed seals. The perfluorinated sulfonic acid, PFOS, has also been found in Canadian Arctic biota, a chemical that was not expected to be found in the Arctic due to its physical-chemical properties. The discovery of such a chemical in Arctic biota at significant concentrations raises questions about sources and pathways of these and other polar chemicals. *Concentrations of these new chemicals are generally lower than legacy POPs (PCBs, DDT, etc.) but there is concern because some are increasing in concentration (PBDEs), others such as PFOS have unique toxicological properties, and some were not expected to be found in the Arctic.*

The NCP QA/QC program has played an important role in improving the quality of contaminant data produced by NCP projects. This has resulted in high confidence levels in data and subsequent conclusions.

6.1.2 Spatial trends

The scope of contaminant data for freshwater fish has grown considerably since the first CACAR assessment. New data for the NWT and Nunavut addresses a data gap identified in this report. The number of species used has increased and data for mercury and OCs is available for lake trout, walleye, northern pike and Arctic char from a wide geographic region. Mercury data for a number of fish species now includes most regions of the Canadian Arctic. *For many lakes concentrations of mercury in top predator freshwater fish exceed guideline levels for subsistence and/or commercial sale. No geographic trends are discernable for the elevated mercury levels.* A number of studies, however, have found that food web length, and age and size of the fish are important variables, while lake size or area do not appear to be as important.

Mercury data has been produced for ringed seals and beluga from across the Canadian Arctic. *Levels of mercury in ringed seals and beluga show no significant geographic trends; however, levels are elevated in beluga in the Beaufort Sea and western Hudson Bay regions.* Higher levels of mercury in the Beaufort Sea region may be due to the effects of increased mercury in the Mackenize River due to higher temperatures. Levels of cadmium in ringed seals and beluga continue to show large differences in levels in liver and kidney between western and eastern Canadian Arctic populations, which is best explained by differences in geology.

Concentrations of most OCs in biota are slightly elevated in the eastern compared to the western Canadian Arctic,

consistent with circumpolar trends. HCHs tend to be higher in the western Arctic because of the influence of recent use in Asia. Levels of OCs, with the exception of HCH, are lower in the Canadian Arctic compared to Greenland and the European and Russian Arctic.

Lake-to-lake variability of OCs in freshwater fish is less than what is seen for mercury but lake trout and burbot in a number of lakes have higher levels. The reason for this is food web length for some lakes but this does not explain the high levels seen in some lakes. In Yukon, glacial melt may influence OCs seen in some lakes and this effect may become more important if increasing temperatures continue. A study in Resolute Lake found that higher water temperatures resulted in higher levels of cadmium in Arctic char liver. The influence of climate change remains an unknown factor in contaminant dynamics in freshwater biota and should receive attention in the future.

Spatial coverage of OCs in ringed seals, beluga and seabirds remains a strength of the Arctic contaminant data set for Canada. Concentrations of OCs in marine mammals and seabirds remain fairly comparable across the Canadian Arctic although subtle differences from west-to-east and south-to-north are found in proportions of various chemicals.

Information on new chemicals and metals other than mercury is most complete for marine mammals compared to other species but it is not currently possible to assess the geographic trends of these new contaminants. New information on spatial trends of OCs and metals in Arctic fox and marine invertebrates has recently been produced.

A number of comprehensive marine food web studies were carried out recently. *These studies have provided information on the pathways of biomagnification of OCs and mercury in marine food webs.* These studies also highlighted the significance of biomagnification on levels of OCs and mercury in higher trophic level organisms.

The contribution of local contamination by POPs has not been addressed very well because of the emphasis of NCP on background sites influenced by long-range transport. Near field- far field comparisons have only been conducted in a few cases, e.g., Saglek Bay (Labrador), and they generally have shown more elevated contaminant levels.

6.1.3 Temporal trends

The most significant development on contaminants in Canadian Arctic biota is that of temporal trend data sets, which have covered a range of species and chemicals (OCs and metals), including new chemicals such as polybrominated diphenyl ethers. Many of these studies

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have provided data sets that stretch from the 1970s to 2000 for legacy OCs and from the 1980s for PBDEs and PCDD/Fs. These temporal trend studies have addressed a major data gap identified for both the Canadian and circumpolar Arctic and have provided data sets for comparison to comprehensive long-term studies of contaminants in fish from the Great Lakes and northern Sweden. Retrospective surveys have also provided temporal trend data sets for new chemicals, only recently discovered in the Arctic. *These results demonstrate the importance of continued monitoring of contaminants and the maintenance and support of tissue archives.*

There is solid evidence that mercury has increased in Canadian Arctic animals from pre-industrial times to the present; however, temporal trend data sets of mercury in these animals over the past 20–30 years show conflicting trends. *In most data sets, which includes freshwater and marine organisms, there has been no change in mercury levels over the past 30 years.* Mercury is increasing in beluga from the Beaufort Sea and western Hudson Bay but not from any other locations. It has been suggested that increased temperatures in the Mackenzie River region have liberated mercury and may explain the increases in mercury concentrations observed in the Beaufort Sea beluga. Mackenzie River burbot are the only freshwater fish species which have shown increasing concentrations of mercury, which supports this hypothesis. Recent changes in climate have been observed in western Hudson Bay and may also play a roll in the increases in mercury observed in beluga from this region. *Therefore, it appears that increasing trends in mercury observed in some Arctic populations may be related to climate change.*

Most "legacy" OCs have significantly declined in Canadian Arctic biota from the 1970s to the 1990s. The one exception to this is ΣHCH levels, which have remained relatively constant in most species. This decline was most rapid in the 1970s and 1980s following the banning of OCs in the USA, Canada and western European countries. The rate of decrease, however, has slowed or stopped in many species, e.g., polar bears of western Hudson Bay. The rate of decline of OCs also varies among species. It is more rapid in ringed seals and seabirds and slower in polar bears and beluga.

The relative proportions of individual chemicals have also changed. More recalcitrant compounds have become more prevalent. For example, PCB 153, DDE and β-HCH all make up a great proportion of their respective groups now than in the past.

Impact of climate change on OC and Hg dynamics is an emerging issue. The dramatic increases in Hg in Mackenzie Bay beluga coincide with a regime shift in the Beaufort sea and with warming of the western Arctic including the Mackenzie valley.

6.1.4 Biological effects

The first CACAR concluded that there was limited information and data on biological effects of contaminants in Canadian Arctic biota and it remains an important knowledge gap. *Although most of the studies to date have failed to find biological effects related to anthropogenic contaminants and threshold levels of effects are generally above current levels found in the Canadian Arctic environment, there is still cause for concern and a need for additional research.* Biological effects have been observed in polar bears in western Hudson Bay and in seabirds from Saglek Bay (Labrador). Climate warming may increase contaminant burdens and stress associated with current and future contaminant burdens may become important. Although the possible effects associated with change in the Arctic clearly goes beyond contaminant effects it is important that they be considered and studied in the future.

Recently completed studies of Churchill and Svalbard (Norway) polar bears have concluded that the immune systems of polar bears are compromised by PCBs and potentially by other OCs. Although the Churchill bears have lower concentrations than those from Svalbard, they did fall within the relationship and thus remain a concern. The influence of these altered systems on the health of polar bear populations is unknown.

Near field-far field comparisons conducted in Saglek Bay have shown elevated contaminant levels and related biological effects in black guillemots near sites of former PCB use.

TCDD toxic equivalent (TEQ) concentrations in livers of northern fulmars, black-legged kittiwake and thick-billed murres from Lancaster Sound surpassed no-observed-effects-levels and lowest-observed-effects-levels established for fish-eating birds such as herring gulls, bald eagles and cormorants. In particular, levels in northern fulmars far exceed these threshold levels. These TEQ values were much higher than levels observed in marine mammals, which generally do not exceed thresholds for effects based on TCDD TEQs. Further investigation of dioxin-like chemicals in seabirds is therefore warranted.

6.2 Knowledge gaps and recommendations

6.2.1 Priority chemicals

Although identification and measurement of new organic contaminants in Canadian Arctic biota is one of the major accomplishments since the first CACAR was published, the amount of data is limited. Data for toxaphene in the terrestrial environment continues to be lacking. New data for PCDD/Fs and non-*ortho* PCBs have been generated for seabirds but spatial trends remain limited, particularly in the freshwater environment.

The amount of information on metals other than mercury, cadmium and selenium has increased since the first CACAR, but remains limited. Knowledge of the speciation and isotopic ratios of metals, which are useful for assessing exposure, pathways and sources of metals continues to be a knowledge gap. The contribution of natural geological and anthropogenic sources to the body burdens of mercury and other toxic heavy metals continues to be elusive, as does the possible biological consequences of these different sources.

The influence of climate warming on levels of OCs and metals in Arctic biota is currently difficult to assess. Some studies have found that changes in climate may result directly in higher levels of metals in biotic tissues.

The contribution of local contamination by OCs, e.g., from burning of municipal waste in northern communities, or from former DEW-line sites to levels seen in biota has not been addressed very well because of the emphasis of NCP on background sites influenced by long-range transport. There is evidence from studies of invertebrates in harbours in the eastern Canadian Arctic, and in Norway and Greenland that harbours are more contaminated than far-field sites. Such near-field, far-field comparisons have only been conducted in a few cases e.g., Saglek Bay (Labrador), and Cambridge Bay, Nunavut, and they generally have shown more elevated contaminant levels.

- Information on new chemicals has been generated for a number of Arctic biota, including some temporal trend data sets, but the scope is still very limited. These chemicals include PBDEs, PCNs, SCCPs and PFOS. There is a need to assess these chemicals in a wider range of species, their food web behaviour and possible biological effects.

6.2.2 Spatial trends and geographic coverage

High lake-to-lake variability of mercury in fish continues to be inadequately explained. New chemicals and non-mercury metals have been measured in freshwater fish but there are not sufficient data to assess the spatial trends of these pollutants. The influence of climate change remains an unknown factor in contaminant dynamics in freshwater biota and should receive attention in the future.

Levels and trends of other metals and elements (other than Hg, Cd, Se, Pb and As) in Arctic biota remain largely unstudied. The platinium series metals, especially palladium, have been found to be increasing in glacial snow cores from Greenland. There is very little information on these or other metals and their dynamics in the Arctic environment.

Very little information has been generated on contaminants in the terrestrial environment. The first CACAR concluded that spatial coverage prior to 1996 was sufficient and that levels were low. Analysis of 25 metals in several caribou herds covering most of their ranges showed some geographic trends for a number of metals. The reason for this variability may be geological but some questions remain about these trends and possible consequences. However, levels and concern about contaminants in the terrestrial environment is low.

There is limited data on levels of metals in marine invertebrates. Knowledge of the dynamics of contaminants in lower trophic level is a key component to assess food web dynamics and spatial trends in the ocean.

No new metal data have been produced for polar bears since the early 1990s and OC data have only been generated for the western Hudson Bay population. Ongoing NCP studies, however, funded in 2002–2003, are addressing this gap. There are limited new data on OCs in narwhal, walrus and other seal species. No data have been generated on the threatened eastern bowhead whale population but new data on western Arctic bowhead suggest levels are likely to be low due to the low trophic level of this species.

- The reasons for variable levels of mercury found in fish from across the Arctic continues to be a knowledge gap.
- Levels and trends of non-standard metals (other than Hg, Cd, Se, Pb and As) continue to be a knowledge gap. There is very little information on these other metals and their dynamics in the Arctic environment.
- The role of abiotic-biotic interfaces at the base of food webs needs to be understood better in both marine and freshwater environments. There is a need, for example, to better understand the link between abiotic Hg deposition in snow and its relationship to marine, freshwater and terrestrial food webs. There have been no studies linking Hg in meltwater to uptake in Arctic food webs. This applies to other metals as well. This is particularly important if climate and ocean current changes distribute or alter food webs and delivery of contaminants.
- The influence of climate change on levels of OCs and metals in Arctic biota is currently difficult to assess. Some studies have found that climate warming may result in directly in higher levels of metals in biotic tissues.
- Data on new chemicals in Arctic biota have increased significantly since the first CACAR but there is insufficient information to assess species differences, spatial trends or food web dynamics.
- It is strongly suggested that statistical analysis and interpretation of all OC and metals data in biota incorporate all relevant and available biological data. New monitoring programs should include a suite of biological measurements such as stable isotopes, age, sex, etc.
- Much of the data used in this assessment were available only in NCP synopsis reports and not subjected to detailed statistical analysis or interpretation. Peer-reviewed publication of this NCP funded work should be encouraged and become an important criterion for continued and future funding.

6.2.3 Temporal trends

Temporal trend data sets of contaminants for terrestrial animals, aquatic invertebrates and marine fish are lacking. With the exception of PBDEs, there is limited information or data on temporal trends of new chemicals. Contrasting temporal trends of mercury across species and regions in the Canadian Arctic is the most serious temporal trend data gap.

- Continued monitoring of legacy OCs, new chemicals and metals has provided valuable information on the effects of banning chemical use and the discovery of new chemicals in Arctic biota. Recently, declines in the rate of decline of legacy OCs in some Arctic species and variable rates between species suggests that long-term monitoring of contaminants should continue in a number of species. Continued support of long-term monitoring studies and tissue archives is strongly recommended.
- Temporal trends of mercury in freshwater and marine biota are contradictory. Some studies have found significant increases and others have found no change in mercury levels. Therefore it is not currently possible to determine if anthropogenic mercury is increasing in Arctic biota. It is also not possible to evaluate where increases in mercury observed in some biota are due to increased anthropogenic inputs are due to environmental change, e.g., climate warming. Temporal trends of mercury continue to be a major gap that warrants further attention and investigation.
- The influence of climate warming on mobilizing metals warrants further attention.
- There is limited data to assess temporal trends of metals other than mercury.

6.2.4 Biological effects

There have been few studies on the biological effects of contaminants in Arctic organisms from Canada or the circumpolar Arctic. Some biochemical effects have been found in more contaminated species but the importance of this on population dynamics is unknown. There is a lack of basic biological information for Arctic organisms that hinders efforts to assess potential changes caused by contaminants, or any anthropogenic stresses.

Most threshold effects established for contaminants have generated using non-Arctic animals. Arctic organisms often have unique biological strategies and systems to deal with the harsh Arctic climate; therefore, comparison of threshold effects levels to current levels in Arctic organisms is confounded.

- A future area of effects studies and risk assessment should focus on thresholds for Arctic species so that more effective comparisons can be made.
- Physiological changes were observed in seabirds with higher levels of PCBs due to local contamination. The impact of these changes in population dynamics should be studied. The health of wildlife near other sources of local contaminants should also be assessed.
- Biological effects of new chemicals, as well as persistent, toxic metabolites such as hydroxy-PCBs and methylsulfone PCBs. Can the effects of the newly measured chemicals like the PBDEs be discerned in the presence of much higher levels of PCBs? Do other chemicals, particularly PFOS, with unique properties have different effects that can be measured?
- Biological effects in species other than polar bears need to be studied. More knowledge about starvation effects is needed in birds and mammals as well. Interpretation of correlative hormone studies is hampered by lack of information on what other variables may affect these wild populations. This makes drawing conclusions from some biomarker studies tenuous.
- Biomarkers for toxaphene exposure need to be developed and biological effects monitoring undertaken in those species with high levels.
- The implications of compromised immune systems in polar bears warrant further study. Clear effects of PCBs were found in individual polar bears, the influence of this on the populations is unknown. It may be that bears with elevated PCBs are more susceptible to viral infection similar to the situation with seals in the North Sea in the 1980s.

PART II

Pat Roach

GNWT/RWED/Mitch Taylor

Interlaboratory Quality Assurance for the Northern Contaminants Program

7.1 Introduction

The NCP, like all research and monitoring programs, requires an ongoing quality assurance (QA) program. Such a program provides assurance to managers of the quality, reliability, and comparability of measurement results being generated for their research projects. At the same time, it should also meet the diverse quality assurance/quality control (QA/QC) needs of the researchers and analysts by providing them with appropriate diagnostic tools for their analyses and offering guidance and support toward corrective measures, if needed.

The main objective of the NCP QA program is to provide information to its science managers on the overall quality of NCP measurement data to assist them in making informed decisions on the sources of contaminants and their effects on the Arctic environment and on human health. As a result, these decision makers would be assured that their contributions toward establishing international agreements and controls to protect the health of the Arctic ecosystem and its inhabitants are based on a scientifically sound database of information.

A second objective of the QA program is to assure research managers of the NCP-funded studies of the quality, reliability, and comparability of measurement results produced by laboratories contributing data to their research. This is primarily achieved by conducting intercomparison exercises on various contaminants of concern, such as persistent organic pollutants (POPs) and heavy metals.

This chapter provides information on the overall quality of the NCP's analytical data from 1998–2002, and focusses on issues such as accuracy, precision, bias, and data comparability among laboratories. The results of a series of intercomparison studies are summarized and recommendations are made for future QA activities. Data quality issues associated with the human heath studies are addressed in "Quality Assurance Aspects of the Human Health Studies" in the Human Health report of this CACAR II series (Van Oostdam *et al.*, 2003).

7.2 Background

Phase I of the NCP (1993–1998) devoted considerable attention to identifying contaminants and pathways, monitoring trends, and establishing controls on substances of concern in the Arctic environment. The focus of Phase II was shifted more toward immediate human health and safety issues associated with these contaminants in traditional/country foods for northern people. At the onset of Phase II of the NCP, it became clear that other external QA programs were already competently addressing the quality of data for many NCP parameters and some matrices of interest. In many cases, this was achieved through accreditation to international standards for environmental, nuclear, and health/hygiene laboratories. Two surveys were therefore conducted in 1998 to assess and prioritize the data quality needs of the NCP, particularly in terms of analytes and matrices of interest.

The first survey identified the organizations that were contributing scientific data to the NCP, reviewed their analytical programs and capabilities, and assessed their existing quality control measures (Stokker and Gomes, 1999a). Table 7.2.1 lists the measurement laboratories that participated in the NCP Interlaboratory QA Program over the last 5 years. Approximately half of these laboratories contributed measurement data to various NCP research studies during Phase II of the program. Many of these facilities were either accredited by, or in compliance with, regulations established by external agencies, indicating that a sound quality management system was in place. These agencies included the Standards Council of Canada (SCC), the Canadian Association for Environmental Analytical Laboratories (CAEAL), the American Industrial Hygiene Association (AIHA), and the Atomic Energy Control Board (AECB). It was determined from the survey that existing quality control measures, including participation in external interlaboratory programs, adequately assured the data quality for determinations of trace metals, organochlorine pesticides (OCs), polychlorinated biphenyls (PCBs), and nutrients in water, for the measurement of radionuclides, and for

analyses of human health tissues (e.g., blood, urine, hair). Due to the lack of available intercomparison programs employing appropriate matrices with suitable target analyte concentrations, however, the following were determined to be the highest priority for annual NCP intercomparisons:

1. heavy metals in sediments and biota;
2. methylmercury in biotic tissues;
3. OCs and PCBs in biotic and abiotic samples; and
4. toxaphene in biotic tissues.

The second, concurrent survey reviewed the suitability of a number of external interlaboratory QA studies that would complement or could even substitute for NCP-run intercomparison studies. These external intercomparisons were of particular interest for the less commonly analyzed parameters such as organotins, polynuclear

TABLE 7.2.1 Laboratories participating in the Northern Contaminants Quality Assurance Program

Organization	Interlaboratory study participation
Analytical Service Laboratories Ltd., Vancouver, B.C.	Trace metals
Atomic Energy of Canada Limited Whiteshell Labs, Pinawa, Man.	Trace metals in sediments
Aurora Laboratory Services Ltd., Vancouver, B.C.	Trace metals
Axys Analytical Services Ltd., Sidney, B.C.	OC/PCBs, toxaphene
Bureau of Chemical Safety, Food Research Division, Health Canada, Ottawa, Ont.	OC/PCBs
Centre de Toxicologie du Québec, Ste-Foy, Que.	Trace metals, OC/PCBs, toxaphene
Centre for Indigenous Peoples' Nutrition and Environment, Ste-Anne-de-Bellevue, Que.	Trace metals and methylmercury, OC/PCBs, toxaphene
Enviro-Test Laboratories, Edmonton, Alta.	Trace metals and methylmercury, OC/PCBs
First Nations and Inuit Health Branch, Health Canada, Ottawa, Ont.	Trace metals, OC/PCBs
Flett Research Ltd., Winnipeg, Man.	Trace metals and methylmercury
Freshwater Institute, Fisheries and Oceans Canada, Winnipeg, Man.	Trace metals and methylmercury, OC/PCBs, toxaphene
Frontier Geosciences Inc., Seattle, Washington, USA	Trace metals and methylmercury
Great Lakes Institute for Environmental Research, University of Windsor, Windsor, Ont.	Trace metals
Great Lakes Laboratory for Aquatic Sciences, Fisheries and Oceans Canada, Burlington, Ont.	OC/PCBs, toxaphene
Great Lakes Science Center, United States Geological Survey, Ann Arbor, Michigan, USA	Toxaphene
Indiana University, Bloomington, Indiana, USA	Toxaphene
Institut des sciences de la mer de Rimouski (ISMER), Université du Québec à Rimouski, Rimouski, Que.	Organotins
Institut Maurice-Lamontagne, Fisheries and Oceans Canada, Mont-Joli, Que.	OC/PCBs, toxaphene
Institute of Ocean Sciences, Fisheries and Oceans Canada, Sidney, B.C.	OC/PCBs
McMaster University, Hamilton, Ont.	Trace metals
National Laboratory for Environmental Testing, Environment Canada, Burlington, Ont.	Trace metals and methylmercury, OC/PCBs, toxaphene
National Water Research Institute, Environment Canada, Burlington, Ont.	Trace metals, toxaphene, organotins
National Wildlife Research Centre, Environment Canada, Hull, Que.	Trace metals, OC/PCBs
Northwest Atlantic Fisheries Centre, Fisheries and Oceans Canada, St. John's, Nfld.	Organotins
Norwest Soil Research Ltd, Surrey, B.C.	Trace metals
Nunavik Research Centre, Makivik Corporation, Kuujjuaq, Que.	Trace metals
Ontario Ministry of Environment, Toronto, Ont.	OC/PCBs, toxaphene
Philip Analytical Services, Bedford, N.S.	Trace metals
School of Public Health, Environment and Occupational Health, University of Minnesota, Minneapolis, Minnesota, USA	Toxaphene
SRC Analytical, Saskatoon, Sask.	Trace metals
Taiga Environmental Laboratory, Yellowknife, N.W.T.	Trace metals
Université du Québec (INRS-Eau), Sainte-Foy, Que.	Trace metals
University of Guelph, Guelph, Ont.	Trace metals
Wastewater Technology Centre, Burlington, Ont.	OC/PCBs
Wellington Laboratories, Guelph, Ont.	OC/PCBs

TABLE 7.2.2 External interlaboratory programs in which NCP laboratories participate

Program & Organization	Analytes	Matrices
CAEAL Proficiency Testing Program Canadian Assoc. for Environmental Analytical Laboratories, Ottawa, Ont.	Trace metals, nutrients, OCs/PCBs, PAHs	Water, sediment/soil, oil, air filters
CFIA Mercury Quality Assurance Program Canadian Food Inspection Agency, Winnipeg, Man.	Hg	Fish
CFIA Fish Check Sample Program Canadian Food Inspection Agency, Mississauga, Ont.	PCBs	Fish
CTQ Interlaboratory Comparison Programs Centre de Toxicologie du Québec, Ste-Foy, Que.	Trace metals	Blood, serum, urine
InterLaB WatR, InterLaB Soil, Environmental Resource Associates (ERA), USA	Trace metals, nutrients, OCs/PCBS, PAHs, dioxins/furans	Water, soil (fortified materials only)
Integrated Atmospheric Deposition Network (IADN) Environment Canada, Downsview, Ont.	Trace metals, OCs/PCBs, PAHs	Standard solutions, water
IAEA-AQCS (Analytical QC Services) Intercomparison Exercises International Atomic Energy Agency, Austria	Radionuclides, trace metals, OCs/PCBs, methylmercury	Sediment, fish, lichen, algae, coral sand, soil, water
Hair Mercury Quality Control Program First Nations and Inuit Health Branch (FNIHB), Ottawa, Ont.	Hg	Human hair
NOAA Intercomparison Exercise for Trace Metals in Marine Sediments and Biological Tissues, National Oceanographic and Atmospheric Admin., USA	Trace metals	Sediment
NOAA Intercomparison Exercise Program for Organic Contaminants in the Marine Environment, Maryland, USA	OCs/PCBs, PAHs	Sediment, mussels, fish
NWRI Ecosystem Quality Assurance Program National Water Research Institute, Burlington, Ont.	Trace metals, major ions and nutrients, Hg	Water
QUASIMEME International Laboratory Performance Studies and Development Exercises, Scotland	Trace metals, PAHs, nutrients, chlorinated organics, VOCs, PCBs, dioxins/furans, toxaphene, organotins	Seawater, soil, sediment, fish, shellfish, standard solutions and extracts

aromatic hydrocarbons (PAHs), dioxins and furans, as well as chemicals of emerging concern such as the brominated flame retardants (BFRs) and polychlorinated naphthalenes (PCNs). Table 7.2.2 lists several of these external interlaboratory programs whose regularly scheduled studies were recommended as supporting the data quality needs of the NCP QA Program (Stokker and Gomes, 1999b). Recommendations were also made for participation in certain international development exercises that were unique, one-time-only intercomparisons.

Each year, prior to scheduling the new intercomparison studies, a list of the analytes and matrices being measured in the research studies was compiled along with updated information on the NCP laboratories and their performance in previous studies. This allowed the most appropriate series of intercomparisons for the QA program to be designed in order to accommodate shifting priorities and new matrices, as well as to focus on data quality issues revealed in previous studies.

7.3 Interlaboratory comparison studies

Table 7.3.1 provides a list of interlaboratory studies that were conducted under Phase II of the NCP. The studies were designed to identify sources of measurement uncertainty and variation of analytical results. By serving as a diagnostic tool for the participating analysts, these studies also provided a means of continually improving the measurement process. As outlined above, the studies were specifically designed to address emerging issues and a variety of matrices, and to monitor QA issues that were revealed in earlier intercomparisons. For the OC/PCB and toxaphene studies, the test samples became progressively more complex in each intercomparison.

7.4 Data assessment techniques

Interlaboratory data were assessed for accuracy, precision, and bias, and where possible, Z-scores were calculated (Thompson and Wood, 1993). To evaluate accuracy, the submitted results for standard solutions and matrix test samples were compared to their target concentrations. In some studies, certified reference materials (CRMs) were used with known target concentrations, while in other cases, the real matrix samples had target values determined by consensus from data submitted in the study. Where sufficient data were received, accuracy and comparability were also evaluated by Z-scores after rejecting outliers using a Grubb's test at the 5% significance level (Grubbs, 1969). Precision was evaluated by means of replicate analyses, by percent difference on blind duplicate

TABLE 7.3.1 Interlaboratory studies conducted from 1998–2002, during Phase II of the Northern Contaminants Program

Study No.	Target analytes	Test samples
NCP II-1	Heavy metals	Great Lakes sediment CRMs
NCP II-2	Heavy metals methylmercury total organic mercury	Whole lake trout (northern Québec) Ringed seal muscle (northern Québec) Mussel homogenate CRM, fish muscle CRM
NCP II-3	21 organochlorines 30 PCB congeners 4 coplanar PCBs	Standard solutions of OCs, PCBs and coplanar PCBs Whole lake trout (Great Lakes) Dried mussels CRM
NCP II-4	Total toxaphene Toxaphene congeners	Technical toxaphene solutions (Hercules standard) Mix of 13 toxaphene congeners Lipid-free burbot liver (Yukon, Northwest Territories)
NCP II-5	Heavy metals Methylmercury Total organic mercury	Narwhal muktuk (Nunavut) Land-locked char fillets (Nunavut) Burbot liver (Yukon, Northwest Territories)
NCP II-6	Organotins (specifically MBT, DBT and TBT)	Standard solutions Dried sediments, dried fish CRM, dried mussel CRM
NCP II-7	Heavy metals Methylmercury Total organic mercury	Caribou liver (Northwest Territories) Ringed seal liver and kidney (Baffin Bay) Greenland shark muscle
NCP II-8	22 organochlorines 30 PCB congeners 12 WHO PCBs	Mixed OC/PCB standard solution Solution of 12 WHO PCB congeners Polar bear blubber (Alaska), ringed seal blubber (Baffin Bay) Lake Ontario Coho salmon, Lake Superior siscowet
NCP II-9	Total toxaphene Homologue totals Toxaphene congeners	Technical toxaphene, mixture of 15 toxaphene congeners Lake Superior siscowet Beluga whale blubber (Nunavut)
NCP II-10 (proposed for 2002)	Heavy metals Methylmercury	Polar bear liver, seabird tissue, whole walleye sediment
NCP II-11 (proposed for 2002)	Toxaphene	Technical toxaphene and congener mix solutions Seal blubber, burbot liver extract

samples, or by graphical interpretation of Youden Pair plots (Youden, 1959; 1960).

Bias, which is an indication of systematic error, was determined by a modified Youden-ranking procedure (Youden, 1962; 1963; 1969), or by graphical interpretation of Youden Pair plots. A set of results was said to be biased when the set exhibited a tendency to be consistently higher or lower than the results from the other participants. This ranking procedure and the criteria employed in testing for bias have about one chance in 20 of deeming a set of results biased, when in fact it was not (i.e., $\alpha = 0.05$). When bias was found, it suggested the presence of a systematic error that should be identified and corrected by the laboratory.

Z-scores were reported only for analytes for which sufficient data were received in the study. They were calculated using the original QUASIMEME-specified target standard deviation of 12.5% for real matrix test materials (Cofino and Wells, 1994). This target variability represents what would be achievable if the participating analysts were able to distinguish between two samples that differed by 50% in concentration. For the trace metal studies, a graphical presentation of "% satisfactory Z-scores" showed which laboratories generated the most reliable data with the least number of outlying results relative to their peers (Wells *et al.*, 1997).

A complete data summary was provided at the close of each study as a diagnostic tool for the participants to apply corrective action, as needed. Where possible, the data review also included a comparison of the overall study results and conclusions of the results of other similar external intercomparisons. Therefore, each study also provided a snapshot of data quality to the science managers of the NCP along with an overview of the capabilities and comparability of the NCP laboratories conducting these measurements.

7.5 Results of the intercomparison studies

In all intercomparisons, participants were asked to use their own in-house methodologies. In order to include as many laboratories as possible, both standard solutions and real matrix samples were used as check samples. By comparing the results submitted on standard solutions, the performance of abiotic and biotic measurement laboratories could be compared because their differences due to the analysis of unfamiliar matrices were minimized. Most of the real matrix samples used as test materials in these studies were biotic in nature, reflecting the emphasis on traditional/country foods of the North in Phase II of the NCP. Although several laboratories that do not generate NCP measurement results were welcomed into the intercomparison program, the remainder of this discussion will focus primarily on the performance of the NCP laboratories.

7.5.1 Toxic heavy metals in sediments

Several NCP Phase I research studies measured loadings of metals such as mercury, cadmium, and lead in Arctic and sub-Arctic lakes (Lockhart *et al.*, 1995; Barrie *et al.*, 1997). Additional studies during Phase II continued to address temporal trends and spatial distribution of mercury and other metals in sediment cores of eastern and western Arctic lakes (Lockhart, 2000; Cheam *et al.*, 2001). Therefore, the first heavy metal intercomparison, NCP II-1, was conducted to assess the ability of the NCP-funded laboratories to measure heavy metals in sediments. Four freeze-dried reference sediments, including one

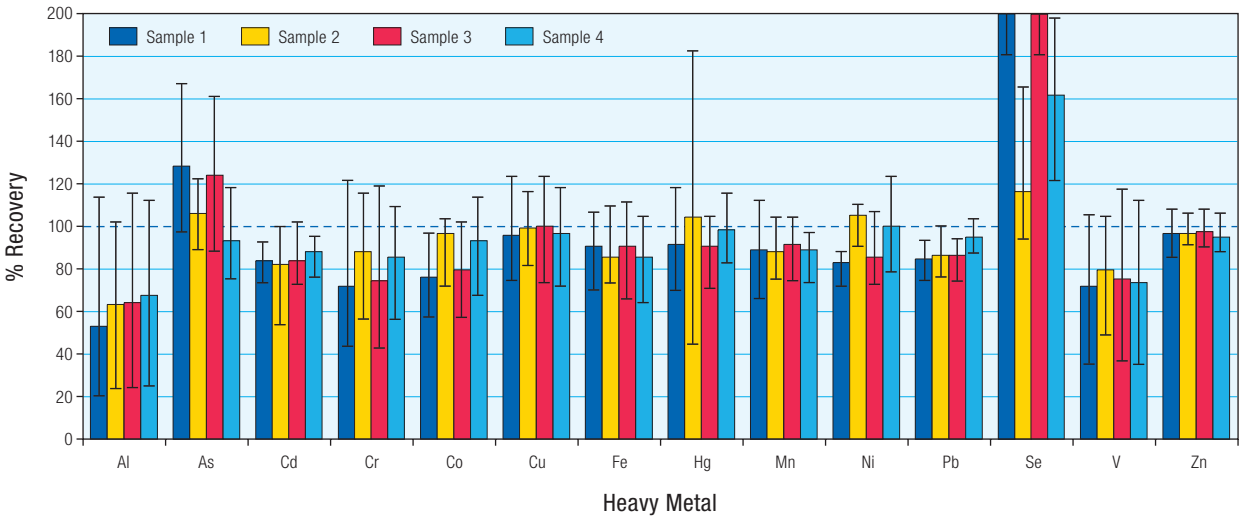

FIGURE 7.5.1

Mean % recovery of heavy metals from sediments in Study NCP II-1. Error bars mark the full range of recoveries by the study participants.

paired set of blind duplicates, were used as the test materials. The participating laboratories submitted results for at least 10 heavy metals, and in some cases, up to 23 metals.

A comparison to the reference and certified reference values of the test sediments showed that accurate data were being generated for most metals in sediment. However, several laboratories employed methods that did not include the use of hydrofluoric acid (HF), which is known to yield lower recoveries for many of the silicate minerals (Cook *et al.*, 1997). This was particularly evident in this study for aluminum, chromium, thallium, and vanadium. For most of the remaining metals, the data were quite comparable between laboratories, showing interlaboratory coefficients of variation of less than 25%. Accuracy and comparability in this study are shown graphically in Figure 7.5.1, which plots the mean % recovery for each metal in each of the four sediment CRMs. The error bars mark the highest and lowest % recovery of each metal in the sediment test samples. It can be seen from Figure 7.5.1 that the most accurate and comparable data were generated for cadmium, cobalt, copper, iron, manganese, nickel, lead and zinc.

Intralaboratory precision, which was calculated as the percent difference between results for the blind duplicate samples, was very good for all but one of the participants. On the other hand, each participating laboratory demonstrated bias for at least one metal in this study. This suggested that systematic errors were present and should be addressed by each participant in this program. Using the results for 11 metals and a target standard deviation of s = 12.5%, all participants achieved better than 70% satisfactory Z-scores (i.e., $Z \leq 2$) on their analyses of heavy metals in sediments.

7.5.2 Heavy metals and methylmercury in biotic samples

Several research studies conducted during Phase I of the NCP revealed alarming trends of mercury levels in the environment as well as in many traditional food species, such as fish, birds, and marine mammals (Barrie *et al.*, 1997). In addition, contamination of marine biota by heavy metals showed significant regional differences (Wagemann *et al.*, 1996) and additional monitoring was recommended to address specific information gaps. Annual intercomparisons (studies NCP II-2, NCP II-5 and NCP II-7) were therefore conducted to assess the measurement capabilities of NCP-funded laboratories to analyze biotic tissues for heavy metals such as mercury, cadmium, and lead, as well as for methylmercury.

In order to assess accuracy, the first biotic intercomparison for heavy metals included several internationally certified reference materials (CRMs). Many of the submitted results were well within the 95% tolerance limits listed for the CRMs' reference values and nearly all data were within ±25% of the certified reference values. The mean % recoveries ranged from 92–101% for all metals except aluminum (84%), chromium (72%), and nickel (71%). At methylmercury concentrations greater than 0.4 μg/g, most laboratories achieved recoveries that were also within ±25% of the reference values.

As listed in Table 7.3.1, the remaining test samples in studies NCP II-2, -5, and -7 were homogenized biota samples from Northern Canada. The naturally low levels of the heavy metals in the Arctic biota caused a wider spread to the data than that found for the sediment samples of NCP II-1.

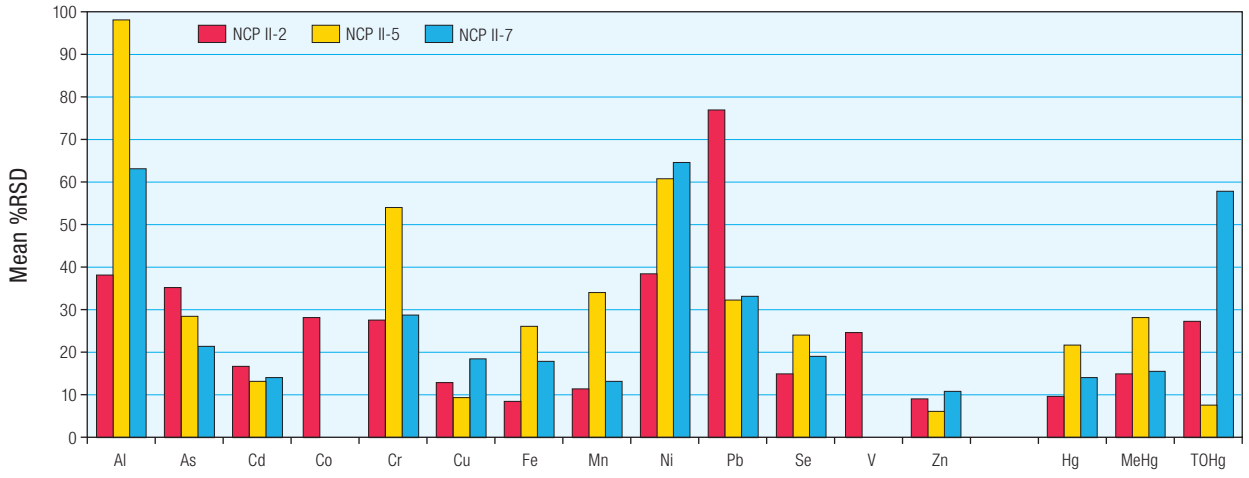

FIGURE 7.5.2

Relative standard deviations (% RSDs) of heavy metals and methylmercury for three intercomparisons on Arctic biota samples. The test samples for NCP II-5 (muktuk, char, and burbot liver) had very low concentrations of most metals.

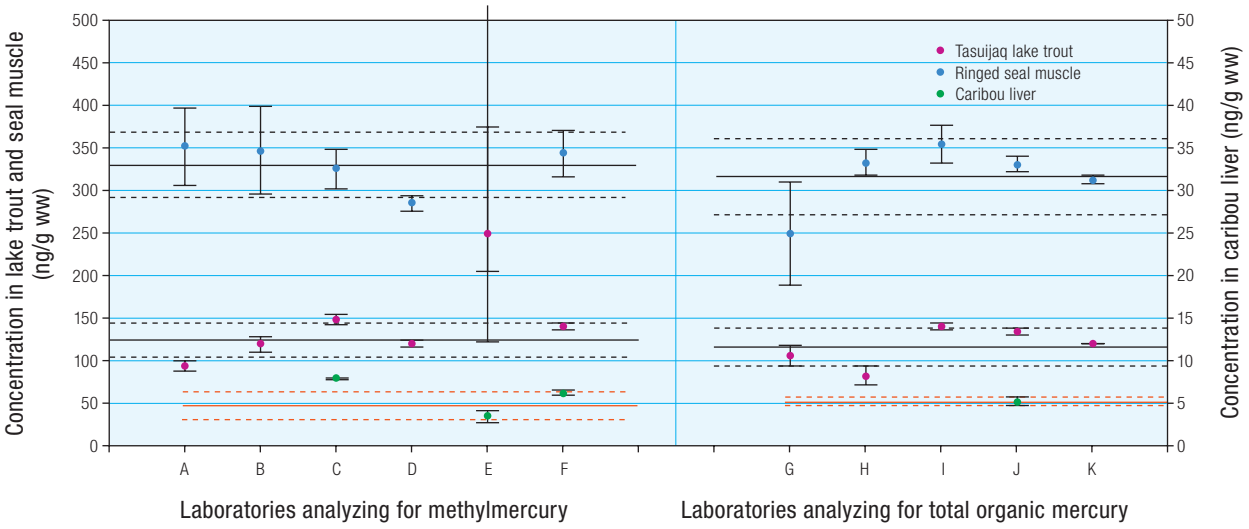

FIGURE 7.5.3

Intralaboratory mean values for methylmercury (six laboratories) and for total organic mercury (five laboratories) plotted on the y-axes. The respective standard deviations of the means of triplicate analyses are shown as error bars. Horizontal lines represent the interlaboratory means ± the interlaboratory standard deviations. The caribou liver sample had levels too low for most laboratories to measure.

Nevertheless, for several metals, the interlaboratory coefficient of variation was better than 30% in all three studies. This is illustrated in Figure 7.5.2, which compares the mean relative standard deviation for each metal in the three studies. The higher variation in NCP II-5 is likely due to the additional challenges of homogenizing and digesting the muktuk, dealing with the high lipid content of the muktuk and burbot liver, and the very low concentration levels in the char. Figure 7.5.2 also shows that the least comparable results were generated for aluminum, chromium, nickel, and lead. Intralaboratory precision for the heavy metals, as determined by triplicate analyses of the seal muscle and whole lake trout samples in NCP II-2 and the caribou liver sample in NCP II-7, was generally very good for all NCP laboratories.

With few exceptions, the NCP laboratories generated excellent methylmercury results in terms of both accuracy (relative to the CRM reference values) and comparability among laboratories (low coefficient of variation), despite the use of very different extraction and analysis procedures at each facility. For total organic mercury, the data were

also fairly agreeable between laboratories, as seen by interlaboratory coefficients of variation of less than 25%.

Figure 7.5.3 illustrates the precision and accuracy demonstrated by each laboratory by plotting the intra-laboratory mean and standard deviation for methylmercury and total organic mercury for each participant on three test samples. (Laboratories B, E, F, and K do not contribute organic mercury data to the NCP program.) Figure 7.5.3 also shows that, despite the very low levels in the caribou liver, those laboratories that were able to measure methylmercury or total organic mercury were precise and comparable.

As a result of the very low levels of the target analytes in many of the samples, there were several outliers in each study, and consequently, several unsatisfactory Z-scores (i.e., $Z \geq 3$). However, most participants achieved better than 70% satisfactory Z-scores. In each study, only a few participants exhibited any metal-specific bias. This is a considerable improvement over the trace metals in sediments study, in which all participants exhibited systematic errors.

The results of these three heavy metal/methylmercury intercomparisons indicate that considerable confidence can be placed in the data being generated for most of the key toxic heavy metals. Aluminum, chromium, and nickel data, however, remain less reliable as the data were significantly less comparable over a wide concentration range. Methylmercury and total organic mercury results remain very reliable for the NCP measurement community. Monitoring of trace metals and methylmercury analyses should continue to be done on an annual basis, incorporating test materials with different concentration levels and varied digestion, analytical, and instrumental challenges.

7.5.3 Organochlorines and PCBs

At the onset of this QA program, there were some concerns about the comparability of PCB measurements, particularly when individual researchers analyzed for and reported data for different sets of PCB congeners. To address this issue, PCB congener concentrations reported in the literature for marine or Arctic samples were compiled in order to tabulate the reported levels for various matrices of interest to the NCP studies. From these data, a list of 30 PCB congeners was recommended for future NCP work, based on their toxicity, frequency of occurrence, and concentration levels relative to the (reported) total PCB levels (Stokker, 2000).

This list of congeners is provided in Table 7.5.1, along with comparisons to similar lists put forward by other national and international environmental monitoring programs including:

1. Arctic Monitoring and Assessment Programme (AMAP) (Murray, 1999);
2. International Council for the Exploration of the Seas (ICES) (ICES, 2001);
3. International Atmospheric Deposition Network (IADN) (Cussion, 1993);
4. National Oceanic and Atmospheric Administration (NOAA, 1993);
5. Gulfwatch Monitoring Program (Gulf of Maine Council on the Marine Environment, 1998a; 1998b; Chase *et al.*, 1998);
6. Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME) (de Boer and Wells, 1997);
7. The Quebec Ministry of the Environment (Lévesque and Moore, 1998); and
8. The Canadian Shellfish Guidelines (Dumouchel and Hennigar, 1995).

In 2001, the subset of PCB congeners of toxicological importance was expanded from the four coplanar PCBs assessed in study NCP II-3 (i.e., PCB 77, 81, 126, and 169) to the 12 PCB congeners provided with a toxic equivalency factor (TEF) by the World Health Organization (WHO) (Van den Berg *et al.*, 1998). Laboratory performance on the analysis of these 12 WHO PCB congeners was evaluated in NCP II-8.

During Phase II of the NCP, two intercomparison studies were conducted on the analysis of OCs and PCBs. Study NCP II-3 utilized separate check sample solutions for OCs and for PCBs, a Great Lakes fish and two biotic tissue CRMs, while study NCP II-8 employed a mixed OC/PCB standard solution and a variety of northern biota as test samples. (Refer to Table 7.3.1 for details.) Injection-ready standard solutions were included as check samples in order to assess the quality of calibration solutions being used and to evaluate the separation and quantitation techniques for both OCs and PCBs without the confounding influence of matrix effects.

The results for the analysis of the 21 OCs in an isooctane solution were extremely good. Most participants were within 25% of the design values, while the interlaboratory coefficient of variation for most parameters was better than 20%. For the analysis of PCB congeners in injection-ready solutions, most of the reported results were within 15% of their design values and interlaboratory comparability ranged from 7–40%. For the mixed

TABLE 7.5.1 Comparison of NCP list of PCB congeners with other international programs

PCB Congener No.	1999 NCP Minimum list	AMAP	ICES	IADN	NOAA	GULFWATCH mussels	QUASIMEME	QUEBEC MOE	Canadian shellfish guidelines
5		(x)		x		(x)		x	
8	√	x/5		x	•	•/5			x
15				x		(x)			x
17								x	
18	√	x	x	x	•	•/15		x	x
28	√	•/31	x	x	•	•	x	x	x
29				x		•			x
31	√	(•)		x			x	x	x
33								x	
44	√	x	x	x	•	•		x	x
49	√	x		x				x	
50						•			
52	√	•	x	x	•	•	x	x	x
66	√	(x)	x	x	•/95	•/95			x
70				x				x	
74	√			x				x	
77	B	x		x	B	•			x
81	B			x					
82								x	
87	√	x		x		•		x	x
90		(•)		x	(x)	(x)			x
95		x/66		x	(x)	(x)		x	x
99	√	x		x				x	
101	√	•/90	x	x	•/90	•/90	x	x	x
105	√	•/132		x	•	•	x	x	x
110	√	x		x				x	x
118	√	x	x	x	•	•	x	x	x
126	B	x		x	B	•			x
128	√	x		x	•	•		x	x
132		(•)		x		(x)		x	x
138	√	•/163	x	x	•/163 /164	•	x	x	x
146		x							
149	√	x		x				x	
151	√	x		x				x	
153	√	•	x	x	•	•/132	x	x	x
154						•			
156	√	x		x			x	x	
157		(x)							
158								x	

continued

Program relevance:

√ – Recommended for NCP work
• – Essential
x – Recommended
B – Indicates congeners of toxicological importance
(•) – Co-elutes with an essential congener
(x) – Co-elutes with a recommended congener

TABLE 7.5.1 Comparison of NCP list of PCB congeners with other international programs

PCB Congener No.	1999 NCP Minimum list	AMAP	ICES	IADN	NOAA	GULFWATCH mussels	QUASIMEME	QUEBEC MOE	Canadian shellfish guidelines
159					(x)				
163		(x)		x	(x)				x
164					(x)				x
169	B	x		x		•		x	x
170	√	x	x		•/190	•/190		x	x
171								x	
177								x	
178									x
180	√	•	x	x	•	•	x	x	x
182					(x)				
183	√	x		x				x	
187	√	x	x		•/182 /159	•		x	x
190					(x)	(x)			x
191								x	
194	√	x		x				x	
195	√	x		x	•	•/208		x	x
199								x	
201	√	x/157							
205								x	
206	√	x	x		•	•		x	x
208						(x)		x	x
209	√	x	x		•	•		x	x
Total # congeners:	30	7 essential 39 (total)	14	84	18–20	25	10	41	35

Program relevance:

√ – Recommended for NCP work
• – Essential
x – Recommended
B – Indicates congeners of toxicological importance
(•) – Co-elutes with an essential congener
(x) – Co-elutes with a recommended congener

OC/PCB solution used in study NCP II-8, the participants continued to produce very accurate and comparable results except for *p,p′*-DDD, *o,p′*-DDT and *p,p′*-DDT. These three parameters had a number of outlying results reported by several participants, resulting in considerably higher coefficients of variation. With few exceptions, PCB congener results reported for the mixed standard solution were consistently within 20% of their design values and showed coefficients of variation up to 20% for the 30 target PCB congeners and up to 30% for the 12 WHO congeners. The widest variation was seen for congeners 66, 95, and 209. A close examination of these data suggests that the close elution of PCB 66 and PCB 95 gave rise to some difficulties in the correct identification of these congeners for a few participants. Collectively, these results provide considerable confidence in the quality of OC and PCB calibration solutions being used by the NCP laboratories. In addition, this indicates that the participating laboratories were in control of the separation, identification, and quantification of individual OCs and PCB congeners.

The results for the tissue CRMs were considerably less accurate than for the standard solutions. This suggests that some analyte losses may be occurring during the extraction or cleanup of the biotic samples. For the certified fish homogenate and the certified mussel tissue, % recoveries of the OCs ranged from 31–137%. Figures 7.5.4a and 7.5.4b illustrate the accuracy and comparability of OCs in standard solutions compared to those in the biotic tissue CRMs. It is clear from these graphs that the interlaboratory means of the OC analytes in the fish and mussel CRMs were considerably less than their respective target values, represented by the diagonal line.

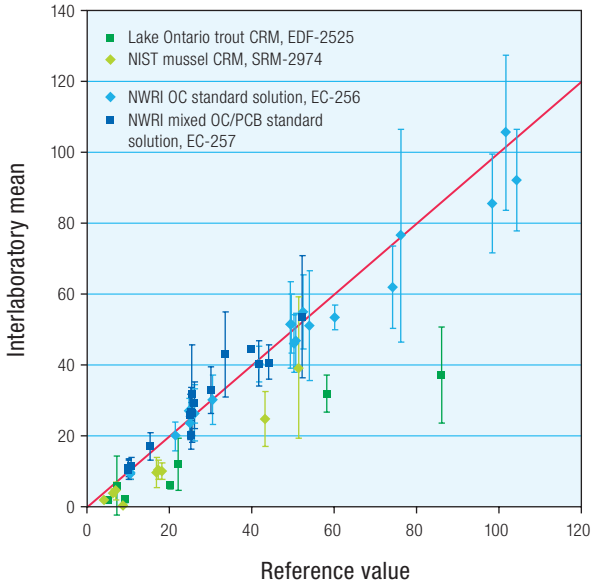

FIGURE 7.5.4a

Accuracy and comparability of OCs in standard solutions (pg/μL) and biota samples (ng/g). Error bars represent the standard deviations of the interlaboratory results. The diagonal line indicates where the interlaboratory mean would equal the reference value if 100% accuracy were achieved.

There was also less comparability among the participants for OC measurements in the biotic tissues than for the standard solutions, particularly where concentration levels were less than 10 ng/g. At concentrations greater than 10 ng/g, the OC coefficients of variation (CVs) generally ranged up to 30%; between 1–10 ng/g, the coefficients of variation ranged up to 50%; and at concentrations less than 1 ng/g, interlaboratory coefficients of variation were as high as 116%. Despite the more generous OC concentrations in the polar bear blubber, seal blubber, and siscowet fish tissue, these high lipid samples had interlaboratory CVs between 15–72%.

Accuracy of PCB congener analysis in the two certified tissues was much better than that for the OCs as the mean recoveries of the individual congeners ranged from 67–104%. For most congeners, the interlaboratory comparability was also good: coefficients of variation ranged from 10–45% in most of the tissue samples, even at levels approaching 1 ng/g. For the black guillemot liver, for which individual PCB concentrations were predominantly below 1 ng/g, some of the interlaboratory CVs ranged as high as 74%. Similar to the OC measurements, somewhat more variation was seen among the data for the high lipid polar bear and seal blubber samples.

In study NCP II-3, nine participants contributed results for the four coplanar PCBs in the injection-ready solution. The agreement to target (accuracy) and the agreement between laboratories (between-lab precision) were

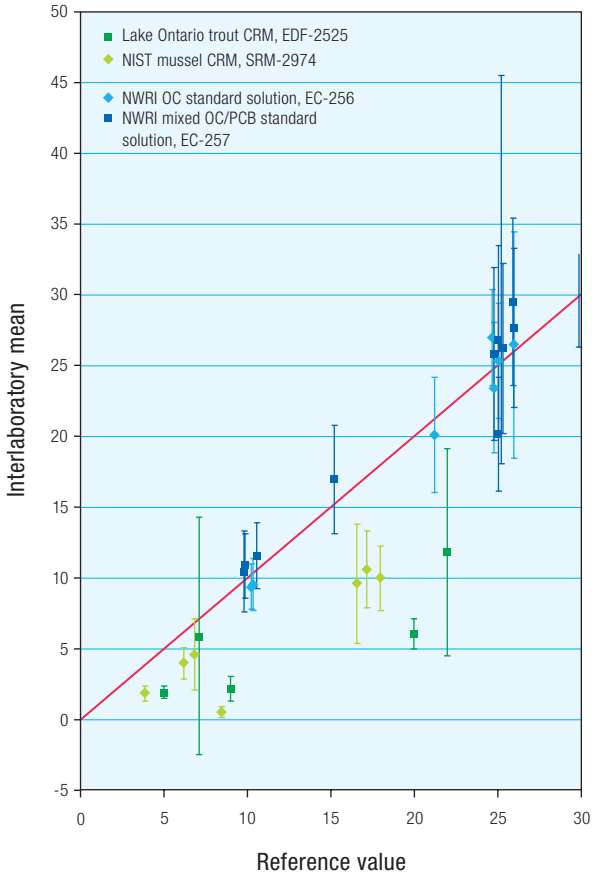

FIGURE 7.5.4b

An expanded view of Figure 7.5.4a showing accuracy and comparability of OCs in standard solutions (pg/μL) and biota samples (ng/g) at low concentration levels.

excellent for this test sample. For the three matrix samples in this study, fewer laboratories were able to quantify the considerably lower concentration levels of these parameters. Nevertheless, with the exception of PCB 81 results from one (non-NCP) laboratory, the coplanar PCB data for the biota samples were in close agreement with each other (better than 30% coefficient of variation for all samples). This is shown in Figure 7.5.5, where the interlaboratory coefficients of variation for the four coplanar PCBs are plotted for the two reference standard solutions and the three biotic samples in study NCP II-3.

Although additional results are still forthcoming for study NCP II-8 at the time of this writing, most of the data received so far for the 12 WHO PCB congeners are also very comparable among laboratories. Many of the interlaboratory coefficients of variation are well below 40%. The most notable exceptions are for the black guillemot liver, which had PCB congener concentrations that were one to two orders of magnitude lower than in the other samples. The statistical results for the data already received are plotted in Figure 7.5.6 along with

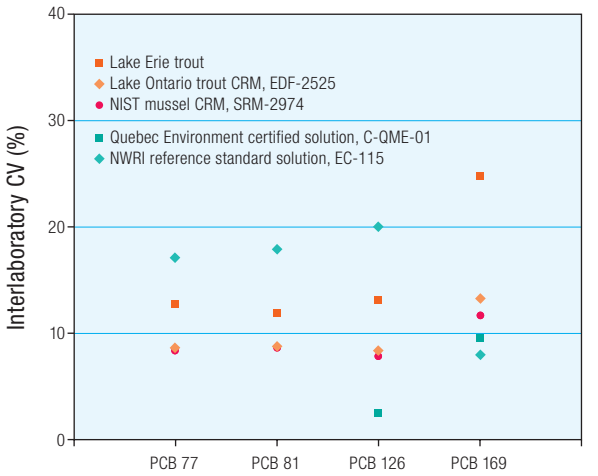

FIGURE 7.5.5

Interlaboratory comparability of coplanar PCBs in two standard solutions and three biota samples.

the nominal concentrations for each congener in the different tissues. It can also be seen from this graph that the data for PCB 77 in the two blubber samples had poor agreement among the laboratories.

In these studies, the OC, congener PCB, and toxic WHO PCB congener analyses were produced using a variety of extraction, cleanup, and instrumental procedures. Although the datasets were too small to draw any firm conclusions about the effect on data quality due to the differences between methodologies, it became apparent that there were considerable differences in reported detection limits. The detection limits reported by the participants varied more than 100-fold for the OCs (from 0.01–5 ng/g) and ranged over four orders of magnitude for some of the PCB congeners (from 0.0001–1 ng/g).

In many Arctic biota samples, methods with high detection limits could have a significant influence on data quality at low concentration levels.

To summarize: the analyses of OCs and PCB congeners in injection-ready solutions were generally quite accurate and comparable. This provides considerable confidence in the quality of calibration solutions being used by the NCP laboratories. The analyses of these same parameters in biotic tissue samples, however, were considerably less comparable, particularly for the OCs, where several extreme values contributed to between-laboratory variations exceeding 100%. This is in agreement with the findings of other international scientists conducting similar intercomparison studies: that currently available analytical methods for congener PCBs and OCs do not allow for the production of very accurate results when analyte concentrations are below 1 ng/g (de Boer and Wells, 1997). The higher lipid content of some of the Arctic biota test samples also appeared to reduce accuracy and comparability among the OC and PCB congener results. On the other hand, analyses of the toxic coplanar PCBs and the 12 WHO PCB congeners in these NCP intercomparisons were much more reliable. Future studies should continue to incorporate a variety of natural materials from the North, in order to accommodate a continuing progression of complexity in the test samples.

7.5.4 Toxaphene

The toxaphene methodology review conducted from 1999–2000 revealed a very diverse set of instrumental and quantitation techniques as well as many types and sources of calibration standards being used by the NCP

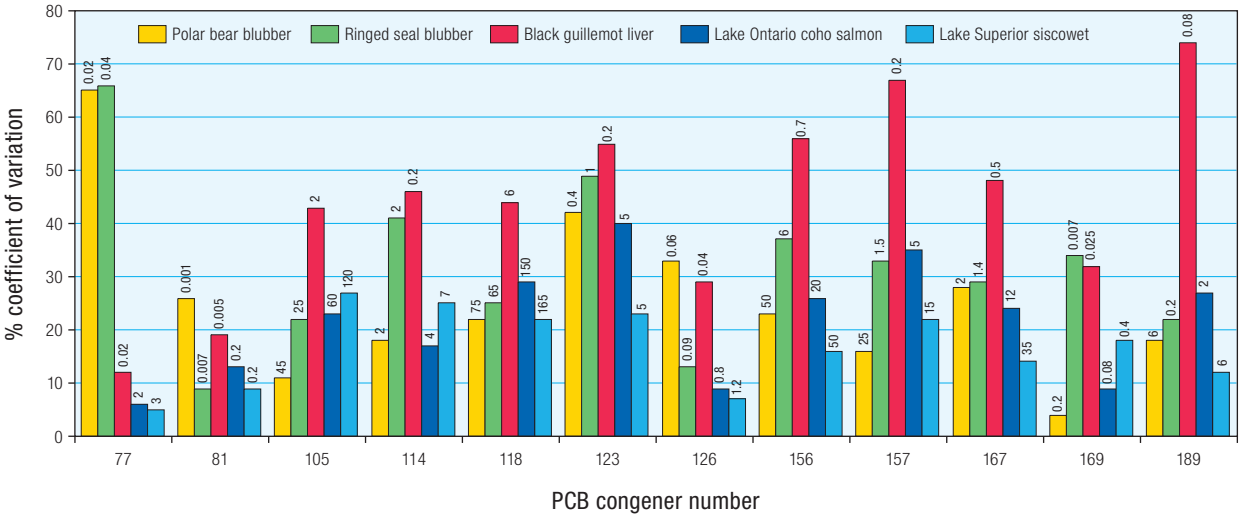

FIGURE 7.5.6

Comparability of WHO PCB congener data for biotic test samples in intercomparison Study NCP II-8. Nominal concentrations for each congener in the different tissues are noted above their respective bar in ng/g wet weight (ww).

measurement laboratories. In order to eliminate differences in standards as the primary source of variation between laboratories, therefore, the first NCP toxaphene intercomparison included the provision of a calibration standard (purchased from Promochem GmbH) which the participants were to use as their calibration solution for the analysis of toxaphene congeners. The four check samples in this study included one test solution of 13 toxaphene congeners, two technical toxaphene standards for total toxaphene analysis, and one lipid-free burbot liver extract for congener-specific and total toxaphene analysis.

The results for this first toxaphene intercomparison were very encouraging, particularly for the congener-specific analyses. The coefficients of variation for the individual (non co-eluting) toxaphene congeners in the blind test solution ranged from 7–25%, showing very good agreement between the ten laboratories. These results compare favourably with the European Research Project MATT where it was determined that "a CV value of about 20% is about the best of what can be obtained at the moment, even for more experienced laboratories" (de Boer *et al.*, 1999). One concern in this study is the magnitude of some of the "false positives" (i.e., results reported for congeners not added to the test sample solution) reported by some laboratories. These are most likely due to system contamination at the laboratory or errors in peak identification by the analysts.

Not unexpectedly, congeners P26, P40, P41, P44, P50, and P62 dominated the toxaphene spectrum for the burbot liver extract. In addition, several participants reported comparable results for significant levels of congeners P38 and P42. Congeners P40 and P41 co-eluted for most analysts in this study and were therefore statistically assessed as one entity. There was good agreement between laboratories for all the above congeners as the interlaboratory coefficients of variation ranged from 18–49%. This is similar to the results seen in the QUASIMEME program where "CV values of 16 to 39% were obtained for the congeners P26, P50, and P62 in cleaned marine mammal and fish extracts" (QUASIMEME, 1998; de Boer *et al.*, 1999).

There was less agreement between laboratories for total toxaphene, although this could partly be due to the provision of a calibration solution for the congener-specific analysis. The two technical toxaphene standards had interlaboratory coefficients of variation of 48% and 43%, and the data for total toxaphene in the burbot liver extract had a coefficient of variation of 56%. Figure 7.5.7 is a Youden Pairs plot (Youden, 1959; 1960) of the total toxaphene data for the two technical toxaphene test samples. This graph clearly shows a bimodal distribution for both of the technical toxaphene standards. The data

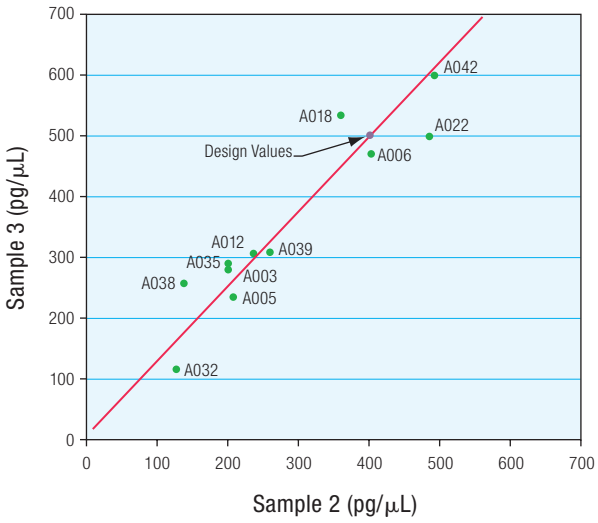

FIGURE 7.5.7

Youden Pairs plot of total toxaphene results for two Hercules standard solutions. The perpendicular closeness of each data point to the diagonal line indicates acceptable intralaboratory precision for all participants.

from one group of laboratories centred about the design values, while a larger group of laboratories reported total toxaphene results at about 50% of the design concentrations. As a result, the interlaboratory means and medians were significantly lower than the target values assigned by the supplier of this standard. Either most laboratories are underestimating their total toxaphene measurements or the reported concentration of this particular commercial standard was incorrectly identified. Figure 7.5.7 also demonstrates precision within the participating laboratories: the increasing perpendicular distance from the diagonal line is proportional to decreasing precision (Youden, 1959; 1960). Despite the poor interlaboratory comparability, therefore, this plot of the two test samples as Youden Pairs revealed that the intralaboratory precision for total toxaphene was very good for all participants.

Our toxaphene methodology survey indicated that several researchers were quantifying total toxaphene against a Hercules technical toxaphene standard, while others employed technical toxaphene standards from the U.S. Environmental Protection Agency or other (commercial) suppliers. Most calculated total toxaphene by using a single response factor against the sum of four to more than 40 peak areas, while two used multiple response factors for some or all of their total toxaphene calculations (Stokker, 2000).

As indicated by the wide-ranging recoveries against the Promochem calibration standard provided, there were significant differences between the participants' congener standards. Further work is needed to identify how much of these differences is due to the calibration standards

and how much to peak identification and the quantitation procedures used by the analysts. Even among the laboratories who reported using the same standard (Dr. Ehrenstorfer's 'Parlar 22'), there was considerable variability between congeners P26, P38, P39, P41, P44, P56, P58, and P62, which could not be attributed to one particular laboratory. Generally, the Parlar source of standards tended to be equal to or somewhat higher in concentration than the Promochem calibration solution provided. Until this variation among in-house calibration standards is reduced, however, it may contribute significantly to a lack of comparability among sample results generated by different laboratories.

In the first intercomparison study, negative ion chemical ionization mass spectrometry (NICI-MS) was the predominant technique used for quantitation. Eight participants analyzed their samples by GC-MS (gas chromatography-mass spectrometry), two used Ion Trap GC-MS-MS, one combined the data from GC-MSD (mass selective detector) and GC-ECD (electron capture detection), and one used GC-ECD with a microbore column. For the individual congeners, most analysts reported detection limits of 0.1–5 pg/μL, while one laboratory reported congener-specific detection limits of 0.01–0.1 pg/μL for their Ion Trap method. Detection limits for total toxaphene ranged from 5–100 pg/μL. Full details of the analytical and quantitation techniques can be found in the study report for NCP II-4.

The overall performance and comparability of the NCP laboratories in the first toxaphene intercomparison were very encouraging. There were also indications, however, that significant differences remain between the laboratories, particularly for total toxaphene data. To ensure comparability with other international programs that address toxaphene, the selection of target parameters for future studies will continue to follow the recommendations put forth by AMAP (AMAP, 1998; p. 312) that

> *"Future monitoring should ... include determination of total toxaphene (by NIMS) for comparison with past work as well as measurements of specific chlorobornane congeners."*

Therefore, the second toxaphene intercomparison (NCP II-9) is focusing on the separation and identification of individual congeners and the quantitation of total toxaphene. In order to include several American laboratories that do not routinely measure toxaphene congeners, homologue totals were also requested of the participants. The study samples include standard solutions to assess accuracy as well as more complex matrix samples: a beluga blubber sample and a high lipid fish homogenate prepared from a large Lake Superior siscowet. This study is still in progress at the time of this writing.

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7.5.5 Radionuclides

Several NCP research projects continue to incorporate the analysis of radionuclides into their work each year. Most of these measurements, however, have been conducted at only two facilities, both of which routinely participate in more than one international program of radionuclide intercomparisons (Stokker, 2000). The first facility has a full QA program in compliance with the Atomic Energy Control Board requirements and ISO Guide 17025 (Standards Council of Canada, 2000). They also participate regularly in interlaboratory programs conducted by the U.S. Environmental Protection Agency (US EPA) National Exposure Research Laboratory (NERL), Chalk River Labs of Atomic Energy of Canada Limited (AECL), and the World Health Organization's (WHO) International Reference Centre for Radioactivity in France. The second institution participates frequently in the intercomparisons offered by the US EPA NERL and the International Atomic Energy Agency (IAEA) in Austria.

7.5.6 Stable lead isotopes

The quality and reliability of stable lead isotope measurements is a growing area of analytical concern. Past NCP studies have reported on these measurements (Lockhart *et al.*, 2000b) and they are becoming increasingly popular in global studies of climate change and contaminant source identification (Kurkjian *et al.*, 2002; Murphy and Katz, 1998).

The measurement of environmental isotopes is a powerful tool that can be used to identify contamination sources and pathways, to investigate climatic changes, and to study environmental forensics (Murphy and Katz, 1998) and human exposure and metabolism (Smith *et al.*, 1996; Smith *et al.*, 1998). This is because isotopes,

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both stable and radioactive, are preserved in various natural archives such as lake and ocean sediments, glaciers, polar ice caps, precipitation and oceans, and even trees. Ratios of stable isotopes are often uniquely associated with a geologic formation, while certain naturally occurring processes can concentrate the abundance of one type of isotope in one location as compared to another. Lead's isotopic abundance is one of the least reproducible because various isotopes are the final products of the radioactive decay of a number of heavy elements (Woolard *et al.*, 1998). The subtle changes in the concentration or type of isotopes can be accurately measured and compared to reconstruct an accurate history or to provide evidence of anthropogenic inputs of contaminants, such as lead from leaded gasoline.

Until the mid- to late-1990s, most studies employing stable lead isotope tracer methods used thermal ionization mass spectrometry (TIMS) to measure isotope abundance. This technique provides excellent isotope measurement sensitivity and precision, but is expensive, labour-intensive, and not well suited for rapid throughput of large numbers of biological samples (Gwiazda *et al.*, 1998). More recently, scientists have explored the use of ICP-QMS (inductively coupled plasma quadrupole mass spectrometry) to measure lead isotopic ratios in environmental work. Recognized limitations to ICP-MS measurements include the inability to measure very low levels of lead and particularly the low abundance isotope ^{204}Pb (Delves, 1999), a relatively noisy sample introduction, spectroscopic interferences, and not enough precision to adequately distinguish environmental sources based upon their isotopic composition (Sardella, 2000). In the case of lead, the isotope ratios of contaminant environmental lead typically vary in a narrow range (e.g., by less than 3 to 7%). For ICP-QMS, the general precision of measurement is considered to be 2–3%, with a precision on isotope ratios of 0.1–1% (Sardella, 2000; Moens and Jakubowski, 1998). The use of high-resolution technology, such as the double-focussing mass spectrometer, which can greatly improve the sensitivity and precision, is now becoming more common. A single-collector double focussing magnetic sector ICP-MS can achieve precisions of 0.05–0.2%, and when a multi-collector detection system is used, precision can be further improved to a level comparable with that of the more costly TIMS (Moens and Jakubowski, 1998). It is becoming increasingly important, therefore, to assess the precision and accuracy of stable lead isotope data by considering the limitations of the instrumentation used (Smith, 2000).

At present, there are no known external quality assessment programs for stable lead isotope measurements. Few certified reference materials (CRMs) are known to be commercially available. The National Institute for Standards and Technology (NIST) has the following three lead (Pb) wire CRMs available:

SRM-981: Common Lead Isotopic Standard;

SRM-982: Equal-Atom Lead Isotopic Standard; and

SRM-983: Radiogenic Lead Isotopic Standard.

Each has certified isotopic compositions for ^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb and for the atomic abundance ratios of ^{204}Pb/^{206}Pb, ^{207}Pb/^{206}Pb, and ^{208}Pb/^{206}Pb.

When stable lead isotope measurements have a greater role in the NCP research studies, it is recommended that method performance be carefully documented with all data. Furthermore, precision and accuracy of such analyses should be established and monitored internally by the laboratories with the above CRMs, and externally by the NCP or other external QA providers, in appropriate intercomparison studies.

7.5.7 Organotins

The toxic impact of tributyltin (TBT) on marine organisms has become well known (Chau *et al.*, 1997) and its discovery in harbour sediments and snails from Norway, Iceland and Alaska, including some from remote regions, has made it a contaminant of increasing concern (AMAP, 1998). It is released into the environment by the leaching of TBT-based anti-fouling paints used on boats and ships. Because the data on TBT levels in the Arctic are limited, the following recommendations were made in the AMAP (Arctic Monitoring and Assessment Program) Assessment Report:

> *"Surveys of TBT in harbour sediments in the Arctic should be carried out to assess the extent of TBT contamination."* and

"The risk exists that TBT biomagnifies and analyses should also be made on bottom-feeding fish, waterfowl, and marine mammals, particularly as these are components of the diet of Arctic peoples." (AMAP, 1998 p. 307).

Although few NCP research projects had proposed to incorporate the analysis of organotins in their work (Stokker, 2001), eight Canadian facilities were invited to participate in intercomparison study NCP II-6. The seven test samples included two standard solutions, three dried sediments, and two dried biota samples. The analysts were asked to report on the concentration levels of monobutyltin trichloride (MBT), dibutyltin dichloride (DBT) and tributyltin chloride (TBT).

There was excellent agreement between laboratories on the sediment samples with slightly more variability on the test standard solutions. This was not unexpected, as these compounds are known to readily swap ligands when stored in mixed solutions. The test standard solutions contained DBT at identical concentration levels: very good precision was shown for the analysis of DBT in these blind duplicates. This study, however, also brought into question the quality of some commercially available MBT standards that were being used by a few of the participating laboratories. As a follow-up to this discovery, direct comparative analyses on these standards were made and the MBT standards in question were discarded.

7.5.8 Chemicals of emerging concern

In addition to the more commonly analyzed chemicals of interest, a number of additional contaminants of emerging concern have been addressed in several recent NCP studies. Included among these analytes are polychlorinated naphthalenes (PCNs), short-chain chlorinated paraffins (SCCPs), haloacetic acids, polychlorinated diphenyl ethers (PCDEs), and brominated flame retardants (BFRs) (Bidleman *et al.*, 2001). The measurement analyses for these contaminants are highly specialized and are carried out in only a few laboratories. The intercomparisons conducted by the NCP QA Program, therefore, have not addressed these contaminants. To ensure the data quality of these measurements, however, continued interchange of samples and standards among the NCP laboratories is encouraged, as well as participation in external development exercises, such as those run by QUASIMEME.

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Among the BFRs, temporal and spatial trends of PBDEs have been the focus for several years of three Canadian laboratories (Stern and Ikonomou, 2001), one of which reports PBDE results to the NCP. From 1999–2000, these three laboratories successfully participated in a special international PBDE intercomparison study conducted by researchers in the Netherlands in collaboration with the Bromine Science and Environmental Forum (de Boer and Cofino, 2001). These same researchers have recently completed a second intercomparison as a development exercise under the auspices of QUASIMEME.

7.6 Overall analytical data quality within the NCP QA program

Each year throughout Phase II of the NCP, approximately 30 research projects with an analytical measurement component have received NCP funding. Most of the laboratories participating in the NCP QA Program also participate in a number of other national and international performance evaluation programs, including certification and accreditation programs. On the environmental side, the Canadian Association of Environmental Analytical Laboratories (CAEAL) has accredited more than half of the NCP laboratories for a number of water and/or sediment procedures. Similarly, the NCP laboratories conducting radionuclide analyses were in compliance with national standards for these measurements.

The reliability of heavy metal analyses has steadily improved over the past four intercomparison studies and remains strongly metal-dependent. Most participants in the studies have demonstrated good comparability for the key heavy metals such as arsenic, cadmium, copper, mercury, and zinc. Aluminum, chromium, and nickel data are less comparable among laboratories. In general, accuracy and comparability have been good where metal concentrations are generous, but deteriorate at the very low levels measured in some Arctic tissues such as land-locked char, burbot liver, and ringed seal muscle. Although bias for various metals was evident among all the participants at the onset of this program, it has improved considerably in the last two trace metal studies, with very few participants now exhibiting any metal

bias. Methylmercury and total organic mercury data have generally been reliable throughout Phase II, with the latter measurements being somewhat limited by their higher detection limits.

The first interlaboratory study on the analysis of OCs and PCB congeners in standard solutions and fish tissue (NCP II-3) showed that the laboratories were generally quite accurate and comparable in their analyses of OCs and PCBs in standard solutions but were considerably less comparable on the fish tissue samples, particularly for the OC measurements. Interlaboratory data for the coplanar PCBs 77, 81, 126, and 169 were very good, both in the standard solutions and in the matrix samples. Results from the second interlaboratory study (NCP II-8) showed similar results: the laboratories generated more accurate and comparable results for PCB congeners than for OCs, and the results for standard solutions were significantly better than for the real matrix samples.

These results suggest that the laboratories have been using good quality calibration standards but may be experiencing some losses and/or contamination problems with their sample preparation steps, particularly for high lipid tissues. Despite these concerns, most of the OC and PCB data have been acceptable at more generous concentration levels and are less reliable at concentrations below 1 ng/g.

The results of the first toxaphene intercomparison study confirmed that the different calibration standards being used contributed significantly to the variability among laboratories. This was particularly evident for total toxaphene measurements, which showed a bimodal distribution in the results. Similar to the findings in other international toxaphene intercomparisons, there was good agreement among the participants for their measurements of the key biotic congeners. High false positives, however, were one of the main concerns for the congener-specific analyses, and should continue to be monitored by the use of blind test mixtures of toxaphene congeners.

7.7 Summary and next steps

NCP-funded research and monitoring studies involve the analysis of a wide variety of different contaminants at trace and ultra-trace levels in various matrices including air, snow, water, sediments, plants, fish, bird tissues and eggs, marine and terrestrial mammals, and human tissue samples. It is such a diverse program that it becomes difficult to assess the matter of comparability of data among the different measurement laboratories and between individual projects.

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The NCP intercomparison studies used a variety of (mostly biotic) test materials representative of the types of samples being analyzed in the NCP research studies. In many cases, standard solutions were also used as check samples in order to include several abiotic testing facilities. Although each interlaboratory data summary represents only a snapshot of the quality of measurements being generated at a particular time, several key findings have emerged from this QA program.

Considerable confidence can be placed in the reliability of data generated for the toxic heavy metals arsenic, cadmium, copper, mercury, selenium, and zinc, while aluminum, chromium, and nickel data show more variability between laboratories.

1. Methylmercury and total organic mercury data have been consistently reliable and comparable among the NCP measurement laboratories, while some participating laboratories, external to the NCP, have demonstrated problems with both accuracy and precision.
2. The quality of the OC and PCB calibration solutions in the NCP measurement community are good, but some losses, particularly for the OCs, have been demonstrated on biotic samples.
3. Organochlorine pesticides (OCs) and polychlorinated biphenyls (PCBs) are generally more accurate and comparable at analyte concentrations greater than 1 ng/g.
4. Because of the diversity in standards and quantitation techniques, toxaphene data should be scrutinized carefully, particularly those of total toxaphene measurements.
5. Toxaphene congener analysis in standards and in biotic samples has generally been accurate and comparable among the NCP laboratories.

6. DBT and TBT measurements are reliable for both sediment and biotic samples, while MBT data are highly dependent on the quality of the calibration standard used.
7. External QA programs, including the assessment of compliance to national and international standards, have supported the data quality of radionuclide measurements for the NCP research projects.

Ideally, the QA program would ensure the reliability and comparability of analytical results for all target contaminants in each matrix and species, as well as among the individual laboratories contributing the measurement data. Unfortunately, such a broad scope would be too costly. Consequently, the approach taken so far within the NCP QA Program has been to assess existing data quality measures in each measurement facility, make recommendations for participation in complementary external intercomparison programs, and then prioritize the remaining analytes and matrices. As a result, the current series of NCP intercomparison studies were designed and conducted to address these gaps in data quality assurance.

Future NCP studies should continue to ensure that acceptable levels of precision and accuracy are generated for the measurement of OCs, PCBs, toxaphene, heavy metals, and methylmercury. As more facilities begin to monitor and contribute data on new persistent contaminants such as the PBDEs and PCNs, additional intercomparisons addressing these emerging issues should be conducted. The ultimate goal is to provide assurance to NCP managers and scientists of a reliable and scientifically sound base for their research and monitoring programs of the North.

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Annex Tables

ANNEX TABLE 1 Mercury, cadmium, arsenic, selenium and lead in freshwater fish from the Canadian Arctic

Species (common name)	Region	Tissue	Year	Age	Length (mm)	Weight (kg)
Salvelinus alpinus (Arctic char) - landlocked	Kaniqsujuaq	muscle	1998	11.9 ± 2.1	-	1.43 ± 0.28
Lota lota (Burbot)	Fort Good Hope	muscle	1999	-	676 ± 107	-
	Fort Good Hope	muscle	2000	-	699 ± 104	-
	Fort Good Hope	muscle	1999	-	735 ± 101	-
	Fort Good Hope	muscle	2000	-	732 ± 127	-
	Fort Good Hope	liver	1999	-	676 ± 107	-
	Fort Good Hope	liver	2000	-	699 ± 104	-
	Fort Good Hope	liver	1999	-	749 ± 77	-
	Fort Good Hope	liver	2000	-	732 ± 127	-
Coregonus clupeaformis (Lake whitefish)	Aubry Lake	muscle	1999	-	507	-
	Cli Lake	muscle	1996, 2000	-	495	-
	Colville Lake	muscle	1993, 99	-	447	-
	Deep Lake	muscle	2000	-	444	-
	Ekali Lake	muscle	1996	-	470	-
	Kelly Lake	muscle	1998	-	495	-
	Little Doctor Lake	muscle	1996	-	407	-
	Mahoney Lake	muscle	1996	-	510	-
	Manuel Lake	muscle	1978, 89, 93, 95, 97, 98	-	505	-
	McEwan Lake	muscle	2000	-	450	-
	McGill Lake	muscle	2000	-	392	-
	Mirror Lake	muscle	2000	-	457	-
	Reade (Unnamed) Lake	muscle	2000	-	427	-
	Sibbeston Lake	muscle	1997	-	422	-
	Tagatui Lake	muscle	1996	-	337	-
	Tsetso Lake	muscle	1997	-	421	-
	Turton Lake	muscle	1996	-	419	-
	Willow Lake	muscle	1999	-	405	-
Coregonus nasus (Broad whitefish)	Peel River	muscle	1999	-	-	-
Stenodus leucichthys nelma (Inconnu)	Kelly Lake	muscle	1998	-	760	-
	Peel River	muscle	1999	-	-	-
Coregonus autumnalis (Arctic cisco)	Bandy Lake	muscle	2000	-	325	-
	Deep Lake	muscle	2000	-	155	-
	Ekali Lake	muscle	1996	-	338	-
	McEwan Lake	muscle	2000	-	197	-
	Sanguez Lake	muscle	1996	-	267	-
Salvelinus namaycush (Lake trout)	Atlin Lake	muscle	1993, 98	-	577	-
	Aubry Lake	muscle	1999	-	576	-
	Belot Lake	muscle	1993, 99	-	618	-
	Cli Lake	muscle	1983, 96, 2000	-	481	-
	Coal Lake	muscle	1999	-	344	-
	Colville Lake	muscle	1993, 99	-	512	-
	Fox Lake	muscle	1998	-	400	-
	Itsi Lake	muscle	2001	-	-	-
	Kelly Lake	muscle	1998	-	598	-
	Kusawa Lake	muscle	1999	-	515	-
	Laberge Lake	muscle	1993, 96, 98, 99	-	518	-
	Little Doctor Lake	muscle	1996	-	547	-
	Loon Lake	muscle	1997	-	600	-
	Mahoney Lake	muscle	1996	-	672	-
	Manuel Lake	muscle	1997	-	485	-
	Mirror Lake	muscle	2000	-	460	-
	Nares Lake	muscle	1996	-	530	-
	Quiet Lake	muscle	1992, 99	-	553	-
	Saturday Night Lake	muscle	1998	-	399	-
	Turton Lake	muscle	1996	-	566	-
	Watson Lake	muscle	1997	-	515	-
	Willow Lake	muscle	1999	-	614	-
Esox lucius (Northern pike)	Bandy Lake	muscle	2000	-	591	-
	Cli Lake	muscle	2000	-	660	-
	Colville Lake	muscle	1999	-	630	-
	Deep Lake	muscle	2000	-	584	-
	Ekali Lake	muscle	1996	-	578	-
	Gargan Lake	muscle	1996	-	668	-
	Kelly Lake	muscle	1998	-	673	-
	Little Doctor Lake	muscle	1996	-	695	-
	Loon Lake	muscle	1997	-	648	-
	Mahoney Lake	muscle	1996	-	660	-
	Manuel Lake	muscle	1978, 95, 97, 98	-	637	-
	McEwan Lake	muscle	2000	-	569	-
	McGill Lake	muscle	2000	-	577	-
	Nares Lake	muscle	1996	-	870	-
	Reade (Unnamed) Lake	muscle	2000	-	584	-
	Sanguez Lake	muscle	1996	-	683	-
	Sibbeston Lake	muscle	1997	-	671	-
	Tagatui Lake	muscle	1996	-	634	-
	Tsetso Lake	muscle	1997	-	763	-
	Willow Lake	muscle	1999	-	634	-
Stizostedion vitreum vitreum (Walleye)	Deep Lake	muscle	2000	-	448	-
	Ekali Lake	muscle	1996	-	410	-
	Little Doctor Lake	muscle	1996	-	474	-
	Mackenzie River	muscle	1997	-	352	-
	McEwan Lake	muscle	2000	-	430	-
	McGill Lake	muscle	2000	-	480	-
	Sanguez Lake	muscle	1996	-	440	-
	Sibbeston Lake	muscle	1997	-	468	-
	Tathlina Lake	muscle	1981, 90, 93, 94, 98	-	393	-
	Tsetso Lake	muscle	1997	-	493	-

"-" = no data available. *References:* 1. Muir *et al.* (2001a). 2. Stern *et al.* (2001). 3. Lockhart *et al.* (2000; 2001a); Lockhart (2002).

Sex	n	Statistic	Hg (μg/g)	Cd (μg/g)	As (μg/g)	Se (μg/g)	Pb (μg/g)	Reference
	7	mean ± SD	0.14 ± 0.03	-	0.01 ± 0.00	0.65 ± 0.17	-	1
m	21	mean ± SD	0.286 ± 0.095	-	0.637 ± 0.637	0.395 ± 0.107	-	2
m	21	mean ± SD	0.345 ± 0.097	-	1.333 ± 1.944	0.478 ± 0.136	-	2
f	21	mean ± SD	0.259 ± 0.108	-	2.626 ± 3.815	0.219 ± 0.104	-	2
f	15	mean ± SD	0.364 ± 0.140	-	1.929 ± 1.621	0.460 ± 0.175	-	2
m	21	mean ± SD	0.046 ± 0.024	-	0.607 ± 0.326	1.071 ± 0.628	-	2
m	21	mean ± SD	0.064 ± 0.026	-	0.585 ± 0.412	1.646 ± 0.733	-	2
f	20	mean ± SD	0.064 ± 0.069	-	1.353 ± 0.811	0.687 ± 0.552	-	2
f	15	mean ± SD	0.094 ± 0.056	-	0.632 ± 0.349	1.203 ± 0.469	-	2
-	14	mean	0.048	-	-	-	-	3
-	43	mean	0.078	-	-	-	-	3
-	24	mean	0.03 (n = 54)	-	-	0.14	-	3
-	28	mean	0.249	-	-	0.157	-	3
-	20	mean	0.082	-	-	-	-	3
-	79	mean	0.165	-	-	0.369	-	3
-	18	mean	0.13	-	-	-	-	3
-	20	mean	0.131	-	-	0.222	-	3
-	27	mean	0.111 (n = 50)	-	-	0.23	-	3
-	30	mean	0.088	-	-	-	-	3
-	15	mean	0.15 (n = 29)	-	-	0.159	-	3
-	107	mean	0.351	-	-	0.345	-	3
-	30	mean	0.146	-	-	0.098	-	3
-	143	mean	0.071	-	-	-	-	3
-	20	mean	0.035	-	-	0.119	-	3
-	102	mean	0.075	-	-	-	-	3
-	12	mean	0.113	-	-	-	-	3
-	167	mean	0.085	-	-	-	-	3
-	1	mean	0.08	-	-	0.73	-	3
-	4	mean	0.396	-	-	0.268	-	3
-	10	mean	0.256	-	-	0.444	-	3
-	19	mean	0.229	-	-	0.253	-	3
-	1	mean	0.26	-	-	0.175	-	3
-	20	mean	0.118	-	-	0.087	-	3
-	35	mean	0.093	-	-	0.154	-	3
-	12	mean	0.158	-	-	0.124	-	3
-	15	mean	0.218	-	-	0.551	-	3
-	88	mean	0.254	-	-	-	-	3
-	19	mean	0.192 (n = 73)	-	-	0.102	-	3
-	59	mean	0.79	-	-	-	-	3
-	12	mean	0.072 (n = 16)	-	-	0.318	-	3
-	10	mean	0.204 (n = 250)	-	-	0.175	-	3
-	2	mean	0.397	-	-	0.145	-	3
-	5	mean	0.086	-	-	-	-	3
-	31	mean	0.482	-	-	0.237	-	3
-	13	mean	0.573 (n = 14)	-	-	0.389	-	3
-	39	mean	0.413	-	-	0.381	-	3
-	10	mean	0.393	-	-	-	-	3
-	6	mean	0.369	-	-	-	-	3
-	20	mean	0.367	-	-	0.268	-	3
-	19	mean	0.299	-	-	-	-	3
-	63	mean	0.606	-	-	0.293	-	3
-	1	mean	0.343	-	-	0.25	-	3
-	14	mean	0.302	-	-	0.635	-	3
-	14	mean	0.221	-	-	0.171	-	3
-	55	mean	0.6	-	-	-	-	3
-	2	mean	0.152	-	-	0.195	-	3
-	32	mean	0.38	-	-	-	-	3
-	19	mean	0.323	-	-	0.246	-	3
-	5	mean	0.353	-	-	-	-	3
-	7	mean	0.244	-	-	-	-	3
-	6	mean	0.67	-	-	0.111	-	3
-	7	mean	0.3	-	-	0.096	-	3
-	2	mean	0.587	-	-	0.07	-	3
-	13	mean	0.546	-	-	0.219	-	3
-	10	mean	0.772	-	-	-	-	3
-	14	mean	0.505	-	-	-	-	3
-	20	mean	0.257	-	-	0.212	-	3
-	42	mean	0.441	-	-	-	-	3
-	15	mean	0.331	-	-	0.121	-	3
-	15	mean	0.713 (n = 28)	-	-	0.117	-	3
-	3	mean	0.258	-	-	0.373	-	3
-	45	mean	0.43	-	-	0.078	-	3
-	20	mean	0.703	-	-	0.122	-	3
-	2	mean	0.165	-	-	-	-	3
-	16	mean	0.17	-	-	0.099	-	3
-	3	mean	0.393	-	-	-	-	3
-	30	mean	0.283	-	-	-	-	3
-	4	mean	1.105	-	-	0.195	-	3
-	14	mean	0.256	-	-	0.161	-	3
-	18	mean	0.753	-	-	-	-	3
-	3	mean	0.191	-	-	0.44	-	3
-	15	mean	0.356 (n = 17)	-	-	0.251	-	3
-	10	mean	1.125 (n = 22)	-	-	0.257	-	3
-	20	mean	0.539	-	-	0.237	-	3
-	5	mean	0.327	-	-	-	-	3
-	34	mean	0.452	-	-	-	-	3
-	99	mean	0.485	-	-	-	-	3

ANNEX TABLE 2 Concentrations of organohalogen contaminants (ng/g wet weight) in freshwater invertebrates and fish from the Canadian Arctic

Species (common name)	Region	Location	Tissue	Year	Sex	% lipid	n	Statistic
Invertebrates								
Plankton	Northwest Territories	Great Slave Lake, West basin		1993–95	-	1.4 ± 0.6	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm		1993–95	-	26.2 ± 12.6	-	mean ± SD
Mysids	Northwest Territories	Great Slave Lake, West basin		1993–95	-	33.6 ± 4.6	-	mean ± SD
Amphipods	Northwest Territories	Great Slave Lake, West basin		1993–95	-	2.2 ± 0.6	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm		1993–95	-	9.9 ± 8.2	-	mean ± SD
Freshwater fish								
Lota lota (Burbot)	Northwest Territories	Great Slave Lake, West basin	liver	1993	-	22.9 ± 8.9	-	mean ± SD
	Northwest Territories	Great Slave Lake, West basin	liver	1995	-	21.2 ± 13.7	-	mean ± SD
	Northwest Territories	Great Slave Lake, West basin	liver	1996	-	43.3 ± 9.2	-	mean ± SD
	Northwest Territories	Great Slave Lake, West basin	liver	1999	-	-	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	liver	1993	-	30.0 ± 7.9	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	liver	1999	-	-	-	mean ± SD
	Northwest Territories	Fort Good Hope (Mackenzie River)	liver	1988	m, f	30.2 ± 13.5	10	mean ± SD
	Northwest Territories	Fort Good Hope (Mackenzie River)	liver	1994	m, f	30.6 ± 11.6	9	mean ± SD
	Northwest Territories	Fort Good Hope (Mackenzie River)	liver	1999	m, f	42.1 ± 13.3	21	mean ± SD
	Northwest Territories	Fort Good Hope (Mackenzie River)	liver	2000	m, f	36.2 ± 15.2	20	mean ± SD
	Yukon	Fox Lake	liver	1998	m, f	29.1 ± 7.57	9	mean ± SD
	Yukon	Lake Laberge	liver	1997	f	54.5	1	-
	Yukon	Lake Laberge	liver	1999	m, f	53.8 ± 6.56	11	mean ± SD
	Yukon	Lake Laberge	liver	1999	m, f	47.9 ± 2.47	7	mean ± SE
	Yukon	Kusawa Lake	liver	1999	m, f	23.8 ± 8.46	11	mean ± SD
	Yukon	Kusawa Lake	liver	1999	m, f	25.6 ± 2.67	9	mean ± SE
	Yukon	Quite Lake	liver	1997	-	30.0 ± 4.66	7	mean ± SE
	Yukon	Quite Lake	liver	1997–99	m, f	25.6 ± 12.3	9	mean ± SD
	Yukon	Klukshu Lake	liver	1996	-	36.0	1	-
Salvelinus alpinus (Arctic char)	Cornwallis Isl., Nunavut	Resolute Lake (landlocked)	muscle + skin	1997	-	5.17 ± 2.79	10	mean ± SD
	Cornwallis Isl., Nunavut	Resolute Lake (landlocked)	muscle + skin	1999	-	3.85 ± 1.28	10	mean ± SD
	Cornwallis Isl., Nunavut	Resolute Lake (landlocked)	muscle + skin	2000	-	5.45 ± 1.52	8	mean ± SD
	Cornwallis Isl., Nunavut	Resolute Lake (landlocked)	muscle + skin	2001	-	-	10	mean ± SD
	Cornwallis Isl., Nunavut	Char Lake (landlocked)	muscle + skin	1993	-	4.15 ± 2.12	5	mean ± SD
	Cornwallis Isl., Nunavut	Char Lake (landlocked)	muscle + skin	1999	-	2.86 ± 1.77	4	mean ± SD
	Cornwallis Isl., Nunavut	Char Lake (landlocked)	muscle + skin	2000	-	2.76 ± 0.40	3	mean ± SD
	Cornwallis Isl., Nunavut	Amituk Lake (landlocked)	muscle + skin	1993	-	4.40 ± 2.15	12	mean ± SD
	Cornwallis Isl., Nunavut	Amituk Lake (landlocked)	muscle + skin	2001	-	6.38 ± 1.54	2	mean ± SD
Esox lucius (Northern pike)	Northwest Territories	Great Slave Lake, West Basin	muscle	1996	-	2.2 ± 0.6	-	mean ± SD
	Northwest Territories	Great Slave Lake, West Basin	muscle	1999	-	0.9 ± 0.3	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1999	-	1.1 ± 0.8	-	mean ± SD
Coregonus sp. (Whitefish)	Yukon	Lake Laberge	muscle	1998	-	1.39 ± 0.14	5	mean ± SE
	Yukon	Watson Lake	muscle	1997	f	2.99 ± 1.55	2	mean
	Yukon	Watson Lake	muscle	1998	m, f	1.71 ± 0.12	12	mean ± SE
	Northwest Territories	Great Slave Lake, West Basin	muscle	1993–95	-	9.9 ± 7.0	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1993–95	-	5.9 ± 4.1	-	mean ± SD
Salvelinus namaycush (Lake trout)	Yukon	Atlin Lake	muscle	1998	m, f	1.76 ± 0.58	11	mean ± SD
	Yukon	Fox Lake	muscle	1998	m, f	1.07	2	mean
	Yukon	Kusawa Lake	muscle	1999	m, f	4.21 ± 0.62	3	mean ± SE
	Yukon	Kusawa Lake	muscle	1999	m, f	4.61 ± 2.98	14	mean ± SD
	Yukon	Lake Laberge	muscle	1993	m, f	8.22 ± 4.49	15	mean ± SD
	Yukon	Lake Laberge	muscle	1996	m, f	8.07 ± 4.17	5	mean ± SD
	Yukon	Lake Laberge	muscle	1998	m, f	8.98 ± 1.27	7	mean ± SD
	Yukon	Lake Laberge	muscle	1999	m	3.45	1	-
	Yukon	Quiet Lake	muscle	1999	m, f	2.40 ± 0.90	8	mean ± SD
	Yukon	Coal Lake	muscle	1999	m, f	3.97 ± 2.79	12	mean ± SD
	Yukon	Mandanna Lake	muscle	2000	f	1.83	2	mean
	Northwest Territories	Great Slave Lake, West basin	muscle	1993–95	-	12.8 ± 3.1	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1993–95	-	10.8 ± 6.2	-	mean ± SD
	Northwest Territories	Great Slave Lake, Lutsel K'e region	muscle	1995	-	16.0 ± 5.8	-	mean ± SD
	Northwest Territories	Great Slave Lake, Lutsel K'e region	liver	1995	-	9.6 ± 6.2	-	mean ± SD
	Northwest Territories	Great Slave Lake, Lutsel K'e region	stomach	1995	-	17.9 ± 12.8	-	mean ± SD
	Northwest Territories	Great Slave Lake, West basin	muscle	1993	-	12.8 ± 3.1	-	mean ± SD
	Northwest Territories	Great Slave Lake, West basin	muscle	1999	-	12.6 ± 6.8	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1993	-	7.6 ± 3.9	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1995	-	16.0 ± 5.8	-	mean ± SD
	Northwest Territories	Great Slave Lake, East arm	muscle	1999	-	4.9 ± 3.7	-	mean ± SD
Stizostedion vitreum (Walleye)	Northwest Territories	Great Slave Lake, West basin		1996	-	3.4 ± 1.3		mean ± SD
Stenodus leucichthys nelma (Inconnu)	Yukon	Peel River	muscle	1999	m, f	5.70 ± 1.22	10	mean ± SE
	Northwest Territories	Great Slave Lake, West basin		1996	-	20.5 ± 6.2		mean ± SD
Coregonus autumnalis (Arctic cisco)	Northwest Territories	Great Slave Lake, West basin		1995	-	8.1 ± 6.3		mean ± SD

"-" = no data available.

References:

1. Evans *et al.* (1996; 2001); ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDT = *o,p'* and *p,p'*-DDE, -DDD and -DDT; ΣPCB = 102 congeners; Toxaphene.
2. Stern *et al.* (2001); ΣCBZ = 1,2,3,4-, 1,2,3,5- & 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCH = α-,β-,γ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDT = *o,p'* and *p,p'*-DDE, -DDD and -DDT; ΣPCB = 102 congeners; Toxaphene.
3. Stern *et al.* (2000); ΣCBZ = 1,2,3,4-, 1,2,3,5- & 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCH = α-,β-,γ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDT = *o,p'* and *p,p'*-DDE, -DDD and -DDT; ΣPCB = 102 congeners.

ΣCBz	HCB	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Mirex	PBDE	Reference
-	-	-	0.2 ± 0.1	0.3 ± 0.3	3.4 ± 2.4	1.2 ± 0.2	0.1 ± 0.1	-	-	1
-	-	-	2.2 ± 3.6	0.5 ± 0.2	3.9 ± 1.9	9.2 ± 2.2	0.7 ± 1.1	-	-	1
-	-	-	0.89 ± 0.6	0.3 ± 0.1	2.2 ± 0.7	6.5 ± 2.2	0.2 ± 0.2	-	-	1
-	-	-	0.7 ± 0.4	0.4 ± 0.2	2.4 ± 0.8	6.1 ± 1.1	0.2 ± 0.1	-	-	1
-	-	-	1.4 ± 1.2	0.8 ± 0.6	2.6 ± 1.2	7.1 ± 3.4	0.3 ± 0.2	-	-	1
-	-	-	63.1 ± 16.0	26.9 ± 5.1	76.7 ± 16.9	263 ± 100	-	-	-	1
-	-	-	92.3 ± 29.0	50.0 ± 16.9	158 ± 21.6	424 ± 199	-	-	-	1
-	-	-	74.7 ± 16.7	27.7 ± 2.2	96.4 ± 9.5	348 ± 114	-	-	-	1
-	-	-	71.7 ± 19.3	32.0 ± 9.1	114 ± 47.3	277 ± 48.4	-	-	-	1
-	-	-	93.5 ± 34.7	51.2 ± 25.7	138 ± 52.2	762 ± 298	-	-	-	1
-	-	-	45.7 ± 23.0	23.0 ± 12.1	79.3 ± 50.4	190 ± 60.6	-	-	-	1
13.6 ± 4.21	13.7 ± 4.06	5.53 ± 1.71	23.8 ± 7.37	16.2 ± 5.25	58.1 ± 18.4	60.8 ± 19.3	2.38 ± 0.74	0.57 ± 0.17	-	2
8.63 ± 2.63	8.17 ± 2.48	4.31 ± 1.27	17.3 ± 6.14	19.0 ± 8.28	50.0 ± 17.5	46.8 ± 14.5	2.02 ± 0.62	0.41 ± 0.14	-	2
10.0 ± 3.81	5.43 ± 2.17	3.78 ± 1.38	21.0 ± 8.04	22.8 ± 8.59	62.8 ± 22.3	54.0 ± 20.4	2.38 ± 0.93	0.59 ± 0.24	1.59 ± 1.15 (n = 4)	2
8.72 ± 5.24	4.78 ± 2.89	3.29 ± 1.98	19.0 ± 12.5	21.2 ± 14.9	54.6 ± 36.2	47.0 ± 29.0	2.21 ± 1.57	0.58 ± 0.55	-	2
0.49 ± 0.18	4.45 ± 1.61	9.26 ± 3.42	16.8 ± 6.17	41.3 ± 13.4	32.2 ± 10.3	40.8 ± 13.9	3.11 ± 1.34	0.35 ± 0.14	-	3
5.20	-	19.4	5.20	1620	915	1950	-	-	-	4
41.6 ± 17.6	-	50.3 ± 23.7	248 ± 122	2850 ± 1200	1630 ± 727	3050 ± 1210	-	-	-	3
3.45 ± 0.27	-	17.6 ± 1.80	3.45 ± 0.27	2380 ± 497	985 ± 148	2450 ± 463	-	-	-	4
5.28 ± 2.10	-	5.55 ± 2.15	35.6 ± 16.8	56.2 ± 31.1	130 ± 51.6	208 ± 69.0	-	-	-	3
0.90 ± 0.13	-	5.11 ± 0.62	0.90 ± 0.13	46.1 ± 7.45	116 ± 15.3	618 ± 85.5	-	-	-	4
1.06 ± 0.14	-	14.7 ± 1.77	1.06 ± 0.14	92.8 ± 13.9	48.3 ± 7.16	74.4 ± 11.7	-	-	-	4
7.26 ± 1.72	-	13.3 ± 4.97	27.8 ± 11.6	87.9 ± 32.3	121 ± 114	99.6 ± 46.1	-	-	-	3
1.94	-	3.07	1.94	45.6	54.3	53.0	-	-	-	4
1.70 ± 1.64	1.55 ± 1.52	0.67 ± 0.58	6.80 ± 8.89	9.23 ± 13.0	160 ± 208	-	1.63 ± 1.60	0.09 ± 0.13	-	5
1.47 ± 0.41	1.34 ± 0.37	0.70 ± 0.28	4.67 ± 2.26	6.69 ± 3.34	96.5 ± 56.7	-	1.47 ± 0.47	0.08 ± 0.06	-	5
2.08 ± 0.56	1.90 ± 0.52	0.70 ± 0.18	1.73 ± 0.66	8.79 ± 4.83	133 ± 68.7	-	1.95 ± 0.56	0.10 ± 0.08	-	5
2.70 ± 0.80	2.51 ± 0.79	1.02 ± 0.21	8.10 ± 6.50	10.2 ± 7.8	150 ± 104		2.56 ± 0.72	0.08 ± 0.06		5
3.01 ± 1.46	2.52 ± 1.18	2.64 ± 1.40	17.8 ± 7.85	114 ± 53.9	290 ± 117	41.3 ± 21.1	2.87 ± 1.29	4.35 ± 2.33		5
1.14 ± 0.55	1.06 ± 0.50	0.33 ± 0.17	1.03 ± 0.40	32.3 ± 8.29	125 ± 48.1	-	1.85 ± 1.32	0.48 ± 0.26	-	5
1.00 ± 0.05	0.87 ± 0.04	0.46 ± 0.07	2.13 ± 0.75	14.8 ± 9.27	64.9 ± 31.4	-	1.24 ± 0.14	0.14 ± 0.06	-	5
6.72 ± 3.28	4.35 ± 2.47	2.05 ± 1.12	47.3 ± 23.5	32.2 ± 15.2	72.5 ± 40.4	203 ± 118	10.1 ± 5.35	0.74 ± 0.40		5
3.33 ± 0.04	3.17 ± 0.07	0.78 ± 0.17	62.9 ± 27.3	43.8 ± 18.4	108 ± 49.4		9.86 ± 1.58	0.94 ± 0.51		5
-	-	-	2.3 ± 0.7	2.7 ± 0.8	5.3 ± 1.5	22.2 ± 6.6	-	-	-	1
-	-	-	1.9 ± 1.1	2.3 ± 1.1	4.8 ± 2.5	21.5 ± 7.3	-	-	-	1
-	-	-	0.8 ± 0.2	1.3 ± 0.4	2.5 ± 0.9	12.1 ± 2.5	-	-	-	1
0.004 ± 0.002	-	0.32 ± 0.06	2.32 ± 0.25	24.6 ± 2.36	4.58 ± 1.00	42.7 ± 7.68	-	-	-	4
0.02	-	2.66	9.02	6.43	24.7	20.3 ± 3.13	-	-	-	4
0.26 ± 0.04	-	0.89 ± 0.15	1.66 ± 0.23	6.54 ± 0.79	7.92 ± 1.03	7.26 ± 0.98	-	-	-	4
-	-	-	4.2 ± 3.9	1.9 ± 1.2	4.7 ± 4.2	21.7 ± 16.6	0.7 ± 0.3	-	-	1
-	-	-	2.9 ± 2.3	2.8 ± 1.1	6.6 ± 2.8	25.5 ± 14.3	0.5 ± 0.2	-	-	1
2.24 ± 0.84	-	1.73 ± 0.60	4.71 ± 2.76	4.39 ± 1.58	17.2 ± 7.16	12.7 ± 4.97	-	-	-	3
0.05	-	0.31	0.52	1.46	2.37	5.19	-	-	-	4
0.32 ± 0.03	-	2.22 ± 0.65	20.8 ± 2.72	197 ± 41.3	125 ± 21.2	307 ± 58.0	-	-	-	4
1.52 ± 0.73	-	1.68 ± 0.84	27.4 ± 10.2	139 ± 73.8	91.1 ± 44.3	149 ± 110	-	-	-	3
4.09 ± 2.77	-	5.10 ± 3.53	42.9 ± 27.4	250 ± 98.4	183 ± 91.1	229 ± 66.9	-	-	-	3
2.24 ± 0.90	-	2.16 ± 0.82	22.3 ± 13.2	102 ± 51.1	74.7 ± 35.1	125 ± 35.1	-	-	-	3
2.06 ± 0.56	-	1.92 ± 0.51	13.1 ± 3.62	52.7 ± 14.6	42.6 ± 10.5	70.9 ± 38.2	-	-	-	3
0.15	-	0.20	2.73	19.6	22.8	8.06	-	-	-	4
0.93 ± 0.30	-	0.25 ± 0.12	3.01 ± 0.72	1.62 ± 0.79	7.24 ± 2.27	5.99 ± 1.88	-	-	-	3
0.59 ± 0.44	-	0.73 ± 0.53	4.67 ± 5.43	11.9 ± 9.70	25.2 ± 20.1	6.78 ± 4.85	-	-	-	3
0.15	-	0.45	0.65	3.40	8.18	15.2	-	-	-	4
-	-	-	9.0 ± 5.6	5.8 ± 3.4	13.9 ± 7.2	48.5 ± 23.4	0.7 ± 0.4	-	-	1
-	-	-	14.6 ± 8.5	8.9 ± 5.8	23.2 ± 5.3	122 ± 88.1	1.1 ± 0.5	-	-	1
-	-	-	-	8.0 ± 3.0	20.5 ± 9.0	77.1 ± 28.1	-	-	-	1
-	-	-	-	13.9 ± 11.4	89.4 ± 65.5	164 ± 121	-	-	-	1
-	-	-	-	6.1 ± 3.5	42.7 ± 27.8	331 ± 234	-	-	-	1
-	-	-	9.0 ± 5.6	5.1 ± 3.7	12.3 ± 8.2	43.5 ± 27.2	-	-	-	1
-	-	-	12.6 ± 6.8	6.7 ± 3.4	30.2 ± 16.7	80.3 ± 28.9	-	-	-	1
-	-	-	16.8 ± 9.9	9.6 ± 7.2	24.9 ± 18.5	151 ± 102	-	-	-	1
-	-	-	11.2 ± 4.5	8.0 ± 3.0	20.6 ± 9.0	77.1 ± 28.1	-	-	-	1
-	-	-	6.3 ± 2.7	6.4 ± 2.5	14.4 ± 6.8	52.4 ± 21.1	-	-	-	1
-	-	-	1.3 ± 0.6	1.6 ± 1.0	4.5 ± 1.5	14.7 ± 9.8	0.1 ± 0.02	-	-	1
0.22 ± 0.04	-	0.09 ± 0.02	1.19 ± 0.24	1.28 ± 0.17	3.44 ± 0.41	1.46 ± 0.33	-	-	-	4
-	-	-	6.8 ± 1.9	4.4 ± 1.1	11.5 ± 2.0	31.3 ± 5.5	0.9 ± 0.3	-	-	1
-	-	-	0.6 ± 0.4	1.5 ± 0.4	3.4 ± 3.3	1.7 ± 0.8	0.1 ± 0.1	-	-	1

4. Palmer and Roach (2001); ΣCBZ = 1,2,3,4-, 1,2,3,5- & 1,2,4,5-tetrachlorobenzene, hexachlorobenzene; ΣHCH = α-, β-, γ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane; ΣDDT = *o,p'* and *p,p'*-DDE, -DDD and -DDT; ΣPCB = 102 congeners.

5. Muir *et al.* (2001a); ΣCBz = sum of 1234-tetra, penta- and hexachlorobenzene; ΣHCH = sum of α-, β- and γ-HCH; ΣCHL = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor; ΣDDT = *o,p'*-DDT, *p,p'*-DDE, -DDD and -DDT; ΣPCB = sum of 103 congeners (1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33,20,53,51,22,45,46,52,49,43,48/47, 44,59,42,64/41/71,40,100,63,74,70/76/98,66,95,91,55,56/60,92,84,101,99,119,83,97,87/81,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146,153,132,105,141,179,137,176,130, 138/163,158,129,178,175,187/182,183,128,167,185,174,177,202/171,156,173,157/201,172,197,180,193,191,200,170/190,198,199,203/196,189,208/195,207,194,205,206,209).

ANNEX TABLE 3 Mercury, cadmium, arsenic, selenium and lead in beaver, muskrat, caribou and waterfowl from the Canadian Arctic

Species (common name)	Region	Location or herd	Tissue	Year	Age	Sex
Castor canadensis (Beaver)	NWT	Gwich'in Settlement Area	muscle	1998 & 2001	-	-
			liver		-	-
			kidney		-	-
Ondatra zibethicus (Muskrat)	NWT	Gwich'in Settlement Area	muscle	1998 & 2001	-	-
			liver		-	-
			kidney		-	-
Rangifer tarandus (Caribou)	Nunavut	Beverly herd	kidney	2000	-	-
			liver		-	-
	Nunavut	Bluenose herd	kidney	1998	-	-
			liver		-	-
	Nunavut	South Baffin population	kidney	1999	5.4 ± 2.8	-
			liver		5.4 ± 2.8	-
	Nunavut	Porcupine herd	kidney	1996	-	-
			kidney	1997	4.0 ± 1.9	-
			kidney	1998	3.2 ± 2.9	-
	Nunavut	Tay herd	kidney	1998	3.2 ± 2.2	-
Somateria mollissima (Common eiders)	Southampton Island, NT	East Bay Migratory Bird Sanctuary	liver	1997	adult	f
		East Bay Migratory Bird Sanctuary	kidney	1997	adult	f
	Victoria Island, NWT	Holman	liver	1997	adult	f
		Holman	kidney	1997	adult	f
	S. Hudson Bay	Belcher Islands	liver	1997	adult	f
		Belcher Islands	kidney	1997	adult	f
Somateria spectabilis (King eiders)	Southampton Island, NT	East Bay Migratory Bird Sanctuary	liver	1997	adult	m, f
		East Bay Migratory Bird Sanctuary	kidney	1997	adult	m, f
	Victoria Island, NWT	Holman	liver	1997	adult	f
		Holman	kidney	1997	adult	f

"-" = no data available. *References:* 1. Culhane, M. (2001). 2. Macdonald *et al.* (2002). 3. Wayland (1999; 2000).

n	Statistic	Hg (μg/g, dw)	Cd (μg/g, dw)	As (μg/g, dw)	Se (μg/g, dw)	Pb (μg/g, dw)	Reference
10	mean ± sd	-	< 0.1	0.83	-	< 0.10	1
10	mean ± sd	-	10.3 ± 5.88	0.78 ± 0.31	-	0.33 ± 0.17	1
10	mean ± sd	-	55.5 ± 33.8	0.86 ± 0.49	-	0.35	1
9	mean ± sd	-	< 0.10	0.53	-	< 0.10	1
10	mean ± sd	-	0.13	0.62	-	0.11	1
9	mean ± sd	-	0.26	0.64	-	< 0.10	1
20	geo mean	6.15	45.6	-	-	0.55	2
	range	5.64–8.16	24.9–83.4			0.34–0.89	
20	geo mean	0.80	5.20	-	-	1.08	2
	range	0.46–1.38	3.35–8.06			0.60–1.95	
11	geo mean	1.920	15.3	-	-	0.25	2
	range	1.03–3.59	8.41–27.8			0.14–0.45	
12	geo mean	0.460	3.12	-	-	0.39	2
	range	0.31–0.69	1.99–4.88			0.27–0.55	
19	geo mean	3.13	22.5	-	-	1.84	2
	range	2.28–4.30	11.7–43.4			1.46–2.31	
19	geo mean	0.75	3.47	-	-	7.8	2
	range	0.54–1.06	2.23–5.39			5.32–11.3	
42	geo mean	2.12	47.0	-	-	0.18	2
	range	1.58–2.84	24.6–89.8			0.13–0.24	
53	geo mean	2.03	32.3	-	-	0.18	2
	range	1.49–2.76	15.3–68.1			0.10–0.31	
56	geo mean	1.39	17.5	-	-	0.22	2
	range	0.42–4.57	1.55–197			0.12–0.40	
4	geo mean	0.87	47.3	-	-	0.18	2
13	geo mean	0.51	3.3	-	6.2	-	3
	range	0.28–1.26	2.1–5.6	-	3.2–9.9	-	3
13	geo mean	-	17.9	-	-	-	3
	range	-	11.9–31.2	-	-	-	3
10	geo mean	0.47	5.0	-	9.3	-	3
	range	0.28–0.79	2.9–8.1	-	5.0–16.5	-	3
10	geo mean	-	30.0	-	-	-	3
	range	-	17.5–44.7	-	-	-	3
10	geo mean	0.40	4.4	-	3.2	-	3
	range	0.23–0.84	1.5–12.0	-	2.3–5.1	-	3
10	geo mean	-	17.2	-	-	-	3
	range	-	17.5–44.7	-	-	-	3
10	geo mean	0.84	9.4	-	6.1	-	3
	range	0.44–1.42	5.0–13.0	-	4.6–11.9	-	3
10	geo mean	-	40.8	-	-	-	3
	range	-	31.4–56.7	-	-	-	3
10	geo mean	0.56	7.8	-	12.0	-	3
	range	0.33–0.87	4.4–14.0	-	7.8–21.7	-	3
10	geo mean	-	31.7	-	-	-	3
	range	-	14.4–68.1	-	-	-	3
	range	0.36–2.11				0.10–0.34	

ANNEX TABLE 4 Concentrations of organochlorine contaminants (ng/g wet weight) in terrestrial mammals from the Canadian Arctic

Species (common name)	Location	Tissue	Year	Age	Sex	% lipid	n	Statistic
Castor canadensis (Beaver)	Gwich'in Settlement, W. side of Mackenzie River, NWT	liver	1998 & 2001	-	f	-	?	mean
Castor canadensis (Beaver)	Gwich'in Settlement, W. side of Mackenzie River, NWT	liver	1998 & 2001	-	m	-	?	mean
Castor canadensis (Beaver)	Gwich'in Settlement, E. side of Mackenzie River, NWT	liver	1998 & 2001	-	f	-	?	mean
Castor canadensis (Beaver)	Gwich'in Settlement, E. side of Mackenzie River, NWT	liver	1998 & 2001	-	m	-	?	mean
Ondatra zibethicus (Muskrat)	Gwich'in Settlement, W. side of Mackenzie River, NWT	liver	1998 & 2001	-	unknown	-	?	mean
Ondatra zibethicus (Muskrat)	Gwich'in Settlement, E. side of Mackenzie River, NWT	liver	1998 & 2001	-	f	-	?	mean
Ondatra zibethicus (Muskrat)	Gwich'in Settlement, E. side of Mackenzie River, NWT	liver	1998 & 2001	-	m	-	?	mean
Canis lupus (Wolves)	Watson Lake & Haines Junction, Yukon	liver	1993–94	< 18 mo	m	4.1	3, pooled	-
Canis lupus (Wolves)	Watson Lake & Haines Junction, Yukon	liver	1993–94	19–36 mo	m	5.3	2, pooled	-
Canis lupus (Wolves)	Watson Lake & Haines Junction, Yukon	liver	1993–94	19–36 mo	f	3.7	7, pooled	-
Canis lupus (Wolves)	Watson Lake & Haines Junction, Yukon	liver	1993–94	> 36 mo	m	4.9	5, pooled	-
Canis lupus (Wolves)	Watson Lake & Haines Junction, Yukon	liver	1993–94	> 36 mo	f	4.4	3, pooled	-
Gulo gulo (Wolverine)	Qurluqtuuq area, NWT	liver	1999–2000	-	-	3.84 ± 1.74	12	mean ± SD

"-" = no data available. nd = not detected.

References:

1. Culhane (2001).
2. Gamberg and Braune (1999); ΣCBz = sum of di-, tri-, tetra-, penta- and hexa-chlorobenzene; ΣHCH = sum of α-, β-, γ-HCH; ΣCHL = sum of oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor and heptachlor epoxide; ΣDDT = sum of *o,p'*-and *p,p'*-DDE, -DDD, -DDT; ΣPCB = sum of 101 congeners (8/5,15,16/32,17,18,19,22,24/27,25,26,31/28,33,40,41/71/64,42,44,45,46,47/48,49,52,56/60,66, 70/76,74,83,84/89,85,87,90/101,91,95,97,99,105,107,110,114,118,128,129,130,131,134,136,137,138/163/164,141,144/135,146,149,151,153,156,157,158,170/190,171,172,174,175,176,177,178, 179,180,183,185,187/182,189,191,193,194,195,196/203,197,198,199,201,205,206,207,208,209).

ANNEX TABLE 5 Mercury, cadmium, arsenic, selenium and lead in marine or anadromous fish and marine invertebrates from the Canadian Arctic

Species (common name)	Region	Location	Tissue	Year	Age	Weight (kg)
Marine fish						
Salvelinus alpinus (Arctic char)	Labrador	Hopedale	muscle	1998	7 ± 1	1.78 ± 0.40
	Labrador	Hopedale	muscle	1999	6.7 ± 1.9	0.94 ± 0.42
	Labrador	Makkovik	muscle	1999	6.57 ± 1.60	1.34 ± 0.63
	Labrador	Nain	muscle	1997	-	-
	Labrador	Nain	muscle	1998–99	7.8 ± 1.5	1.38 ± 0.32
	N. Quebec	Kangirsuk	muscle	1998	7.9 ± 1.5	1.70 ± 1.98
	N. Quebec	Tasuijaq	muscle	1998	8.5 ± 1.5	1.35 ± 0.40
	N. Quebec	Quaqtaq	muscle	1998	8.9 ± 2.4	1.30 ± 0.58
	N. Quebec	Povungnituk	muscle	1998	8.1 ± 0.94	1.47 ± 0.43
	N. Quebec	Kaniqsujuaq	muscle	1998–99	11.3 ± 1.53	1.59 ± 0.62
Somniosus microcephalus (Greenland sharks)	Baffin Island	Cumberland Sound	liver	1999–2000	(female)	-
		Cumberland Sound	liver	1999–2000	(male)	-
Bivalves						
Mytilus edulis (Blue mussel)	N. Labrador	Makkovik	whole body	1998–99	-	-
	N. Labrador	Nain	whole body	1998–99	-	-
	Nunavik (Qc)	Kangiqsualujjuaq	whole body	1998–99	-	-
	Nunavik (Qc)	Quaqtaq	whole body	1998–99	-	-
	Nunavik (Qc)	Kangiqsujuaq	whole body	1998–99	-	-
	Nunavik (Qc)	Kuujjuaq	whole body	1998–99	-	-
	Nunavik (Qc)	Deception Bay	whole body	1998–99	-	-
Placopecten magellanicus (Scallops)	Labrador	Nain	muscle	1999	19	-
					9–30	
	Labrador	Nain	gut	1999	19	-
					9–30	
	Labrador	Nain	gonad	1999	19	-
					9–30	

"-" = no data available. *References:* 1. Muir *et al.* (1999a). 2. Muir *et al.* (2000a). 3. Fisk (2002a).

ΣCBz	α-HCH	β-HCH	γ-HCH (lindane)	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Mirex	Reference
< 0.01	-	-	-	< 0.01	1.12	8.82	< 0.01	< 0.01	-	< 0.01	1
< 0.01	-	-	-	< 0.01	2.78	17.3	< 0.01	< 0.01	-	< 0.01	1
< 0.01	-	-	-	8.14	0.79	10.7	20.3	< 0.01	-	< 0.01	1
< 0.01	-	-	-	< 0.01	1.93	13.2	6.09	< 0.01	-	< 0.01	1
< 0.01	-	-	-	< 0.01	4.39	18.9	1.14	< 0.01	-	< 0.01	1
< 0.01	-	-	-	0.47	< 0.01	4.22	< 0.01	< 0.01	-	< 0.01	1
< 0.01	-	-	-	< 0.01	2.14	23.8	< 0.01	< 0.01	-	< 0.01	1
4.2	-	-	-	nd	0.50	nd	0.7	-	nd	nd	2
2.0	-	-	-	nd	4.9	nd	8.1	-	0.3	nd	2
2.6	-	-	-	nd	1.5	nd	9.5	-	1.5	nd	2
7.2	-	-	-	nd	2.4	nd	5.2	-	0.4	nd	2
3.2	-	-	-	nd	2.8	nd	15.1	-	0.8	nd	2
1.48 ± 1.19				1.15 ± 2.13	14.3 ± 27.2	7.91 ± 17.4	75.4 ± 80.2	-	1.93 ± 4.40	1.04 ± 1.55	3

3. Braune *et al.* (2001b); ΣCBz = 1,3-,1,4-,1,2-diCBz,1,3,5-,1,2,4-,1,2,3-triCBz,1,2,3,4-tetraCBz and HCBz; ΣHCH = α-, β-, γ-HCH; ΣCHL = *cis*- and *trans*-chlordane, *cis*-and *trans*-nonachlor, oxychlordane, heptachlor and heptachlor epoxide; ΣDDT = *o,p'* and *p,p'*-DDE, -DDD and -DDT; ΣPCB = sum of 106 congeners (1,3,4/10,7/9,6,8/5,19,30,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33/20,53,51, 22,45,46,52,43,49,47/48,44,59,42,71/41/64,40,100,63,74,70/76/98,66,95,91,55,56/60,92,84,101,99,119,83,97,81/87,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146,153,132, 105,141,179,137,176,130,163/138,158,129,178,175,182/187,183,128,167,185,174,177,202/171,156,173,157/200,204,172,197,180,193,191,199,170/190,198,201,203/196,189,208/195,207,194, 205,206,209).

n	Statistic	Hg (μg/g, dw)	Cd (μg/g, dw)	As (μg/g, dw)	Se (μg/g, dw)	Pb (μg/g, dw)	Reference
13	mean ± SD	0.033 ± 0.005	-	-	-	-	1
7	mean	0.03 ± 0.01	0.001	0.20 ± 0.06	0.37 ± 0.04	0.002	2
22	mean	0.03 ± 0.01	0.002 (n = 9)	0.28 ± 0.13	0.49 ± 0.13	0.005 (n = 9)	2
6	mean ± SD	0.027 ± 0.007	-	0.20 ± 0.05	0.38 ± 0.05	-	2
24	mean ± SD	0.03 ± 0.01	-	0.211 ± 0.064	0.379 ± 0.040	-	1
15	mean ± SD	0.032 ± 0.026	-	0.256 ± 0.062	0.235 ± 0.069	-	1
15	mean ± SD	0.040 ± 0.011	-	0.414 ± 0.147	0.144 ± 0.023	-	1
14	mean ± SD	0.072 ± 0.035	-	-	-	-	1
11	mean ± SD	0.044 ± 0.009	-	1.015 ± 0.182	-	-	1
3	mean ± SD	0.04 ± 0.00	-	0.43 ± 0.07	0.27 ± 0.02	-	1
10	mean ± SD	0.52 ± 0.11	3.35 ± 0.51	9.64 ± 0.86	0.57 ± 0.04	-	3
14	mean ± SD	0.47 ± 0.06	4.31 ± 0.65	9.94 ± 1.06	0.48 ± 0.04	-	3
-	-	0.020	0.27	1.52	0.60	0.09	2
-	-	0.010	0.33	1.89	0.59	0.12	2
-	-	0.030	0.23	2.06	0.41	0.12	2
-	-	0.010	1.10	1.94	0.69	0.21	2
-	-	0.010	0.35	2.13	0.48	0.15	2
-	-	0.020	0.20	2.33	0.50	0.16	2
-	-	0.010	0.39	2.23	0.68	0.28	2
18	mean	0.03	0.95	0.36	0.19	< 0.001	2
	range	0.01–0.05	0.32–2.45	0.25–0.48	0.13–0.26	< 0.001–0.002	
18	mean	0.05	6.15	2.61	2.83	0.246	2
	range	0.03–0.07	3.41–10.6	1.98–3.12	1.63–5.50	0.151–0.350	
18	mean	0.02	1.68	1.38	0.81	-	2
	range	0.00–0.06	0.11–4.16	0.48–2.78	0.52–1.18		

ANNEX TABLE 6 Concentrations of organochlorine contaminants (ng/g wet weight) in marine invertebrates and marine and anadromous fish from the Canadian Arctic

Species (common name)	Region	Location	Tissue	Year	Sex	% lipid	Length	n
Marine invertebrates								
Zooplankton								
Copepod								
Calanus spp.*	Beaufort Sea	Holman, NWT	whole body	1999	-	48.9 ± 6.6	5	-
Calanus hyperboreus	Baffin Bay	-	whole body	1998	-	6.3 ± 0.70	20	-
Metridia longa	Baffin Bay	-	whole body	1998	-	2.1 ± 0.12	3	-
Euchaeta glacialis	Baffin Bay	-	whole body	1998	-	5.4 ± 0.33	3	-
Euphausiid								
M. occulata	Baffin Bay	-	whole body	1998	-	5.0 ± 0.53	7	-
Pandalus sp.	Baffin Bay	-	whole body	1998	-	4.9	2	-
Amphipod								
T. libellula ("adult")	Baffin Bay	-	whole body	1998	-	2.2 ± 0.40	4	-
T. libellula ("juv")	Baffin Bay	-	whole body	1998	-	3.1	1	-
A. nugax	Baffin Bay	-	whole body	1998	-	2.5 ± 0.31	5	-
A. nugax	Cumberland Sound	Pangnirtung	whole body	1999	-	4.2	1	-
Miscellaneous								
Sagitta spp.	Northern Baffin Bay	-	whole body		-	2.1 ± 0.30	6	-
Benthic Invertebrates								
Echinodermata								
G. arcticus	Baffin Bay	-	soft tissue	1998	-	8.5 ± 1.2	3	-
	Cumberland Sound	Pangnirtung	soft tissue	1999	-	11.4 ± 1.3	3	-
C. crispatus	Baffin Bay	-	soft tissue	1998	-	1.4 ± 1.8	2	-
Bivalve								
Y. thraciaeformis	Baffin Bay	-	soft tissue	1998	-	2.3	1	-
Mytilus edulis (Blue mussel)	N. Quebec	Quaqtaq	soft tissue, 1kg pooled	1998	-	4.95	2	-
	N. Quebec	Kuujjuaq	soft tissue, 1kg pooled	1998	-	2.95	2	-
	N. Quebec	Kangipsujuaq (Wakeham)	soft tissue, 1kg pooled	1998	-	4.75	2	-
	N. Quebec	Kangiqsualujjuaq (George River)	soft tissue, 1kg pooled	1998	-	2.00	2	-
	Labrador	Makkovik	soft tissue, 1kg pooled	1998	-	2.16	3	-
	Labrador	Nain	soft tissue, 1kg pooled	1998	-	2.49	2	-
	N. Quebec	Deception Bay	soft tissue, 1kg pooled	1998	-	1.65	3	-
M. truncata		Iqualit	soft tissue	1993	-	1.5 ± 0.26	6	-
Marine fish								
Gadus ogac (Greenland cod)	Beaufort Sea	Holman, NWT	whole body	1999	-	2.61 ± 0.75	10	47.0 ± 2.2
Boreogadus saida (Arctic cod)	Baffin Bay	-	whole body	1998	m/f	1.21 ± 0.25	8	18.0 ± 0.5
Reinhardtius hippoglossoides								
(Turbot/Greenland halibut)	Cumberland Sound	Pangnirtung	muscle, with skin	1999	m/f	18.6 ± 2.44	4	60.8 ± 6.0
Somniosus microcephalus								
(Greenland sharks)	Cumberland Sound	Pangnirtung	liver	1999	m/f	39.7 ± 3.6	15	283.6 ± 5.7
	Davis Strait		liver	1997	m/f	55.9 ± 2.1	2	135 ± 0
Anandromous fish								
Salvelinus alpinus (Arctic char)	Eastern Canada	Kangirsuk	muscle, with skin	1998	-	8.53 ± 2.21	9	-
		Makkovik	muscle, with skin	1998	-	4.88 ± 1.23	2	-
		Nain	muscle, with skin	1998	-	6.86 ± 2.00	7	-
		Quaqtaq	muscle, with skin	1998	-	6.06 ± 6.12	7	-
		Wakeham	muscle, with skin	1998	-	3.9 ± 3.58	7	-

"-" = no data available. * = Concentrations are ng/g dry weight.

References:

1. Hoekstra *et al.* (2002b); ΣCBz = sum of 1,2 diClBz, 1,4-diClBz, 1,2,3-triClBz, 1,2,4-triClBz, 1,3,5-triClBz, 1,2,3,4-tetraClBz, 1,2,3,5-tetraClBz, pentaClBz and hexaClBz; ΣHCH = sum of α, β, γ, δ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, *cis*-heptachlor epoxide; ΣDDT = sum of *o,p'*- and *p,p'*-DDD, -DDE and -DDT; ΣPCB = sum of 101 congeners (4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16,32,54/29,26,25,50,31/28,33/21/53,51,22,45,46,52/49,43,47/48,44,59,42,64,41/71,40,100,63,74,76/98,70,95,66,91,55,56/60,92/84,101,99,119,83,97,87,81,85,136,110,82,151,135,144,107/147,149/133,118,114,143,141,145,153,132,105,141/179,137,176/130,163,138,158,129/178,175,187,182,183,128,167,185,174,177,171,156,202/173,172,197,180/193,191,199,170/190,198,201,176/203,189,206,195,207,194,205,208,209); Toxaphene = total toxaphene.
2. Fisk *et al.* (2002b); ΣCBz = sum of 1,2,4,5-tetraClBz, 1,2,3,4-tetraClBz, pentaClBz and hexaClBz.; ΣHCH = sum of α-, β- and γ-HCH.; ΣCHL = sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane; ΣDDT = sum of *p,p'*- and *o,p*-DDD, -DDE, and -DDT; ΣPCB = sum of 86 congeners (1,3,4/10,7,6,8/5,19,18,17,24/27,16/32,26,25,31,28,33,22,45,46,52,49,47,48,44,42,41/71,64,40,74,70/76,95/66,56/60,91,84/89,101,99,83,97,87,85,136,110,82,151,144/135,149,118,134,114,131,146,153,132,105,141,130/176,179,137,138,158,178/129,175,187,183,128,185,194,196/203,189,208,195,207,194,205,206,209).

Statistic	ΣCBz	HCB	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Mirex	Reference
mean ± SE	3.83 ± 0.58	-	6.21 ± 1.08	3.32 ± 0.85	5.33 ± 0.94	33.8 ± 6.1	38.4 ± 3.3	-	-	1
mean ± SE	0.43 ± 0.05	-	2.5 ± 0.79	0.85 ± 0.19	0.84 ± 0.15	5.1 ± 0.63	-	-	-	2
mean ± SE	0.32 ± 0.09	-	0.78 ± 0.11	1.3 ± 0.12	1.8 ± 0.27	7.6 ± 1.9	-	-	-	2
mean ± SE	0.59 ± 0.03	-	1.3 ± 0.11	2.8 ± 0.11	2.5 ± 0.12	6.0 ± 0.04	-	-	-	2
mean ± SE	0.35 ± 0.05	-	0.58 ± 0.18	0.73 ± 0.18	1.4 ± 0.11	5.8 ± 0.46	-	-	-	2
mean	0.65	-	1.5	1.7	2	29.9	-	-	-	2
mean ± SE	0.43 ± 0.04	-	1.7 ± 0.25	3.8 ± 0.45	3.1 ± 0.53	8.8 ± 1.3	-	-	-	2
mean	0.29	-	0.81	0.9	0.55	3.2	-	-	-	2
mean ± SE	2.2 ± 0.57	-	10.0 ± 4.7	5.0 ± 0.32	5.0 ± 0.29	16.5 ± 4.0	-	-	-	2
mean	5.7	-	26.2	20.5	19.8	54.6	-	-	-	2
mean ± SE	0.31 ± 0.16	-	0.23 ± 0.06	0.33 ± 0.09	0.36 ± 0.04	2.2 ± 0.25	-	-	-	2
mean ± SE	0.64 ± 0.12	-	3.7 ± 1.4	26.3 ± 6.9	25.5 ± 7.5	28.1 ± 9.6	-	-	-	2
mean ± SE	2.0 ± 0.30	-	13.5 ± 0.94	48.5 ± 5.8	27.1 ± 4.8	31.6 ± 4.1	-	-	-	2
mean ± SE	0.21 ± 0.12	-	0.04 ± 0.01	0.03 ± 0.02	0.08 ± 0.08	2.0 ± 2.2	-	-	-	2
mean ± SE	0.22	-	0.04	0.07	0.33	3.4	-	-	-	2
mean	0.57	-	2.79	1.30	1.29	29.0	-	1.20	0.0	2
mean	1.11	-	1.37	0.97	0.86	45.8	-	0.77	0.03	2
mean	0.63	-	2.94	1.83	1.48	13.8	-	1.25	0.01	2
mean	0.29	-	1.12	0.79	0.49	7.01	-	0.55	0.02	2
mean	2.52	-	1.85	9.29	1.89	5.83	-	1.25	0.24	2
mean	2.06	-	1.69	7.10	1.42	7.09	-	1.36	0.01	2
mean	1.20	-	0.61	2.22	0.24	3.68	-	0.39	0.07	2
mean ± SE	0.27 ± 0.04	-	0.49 ± 0.03	0.94 ± 0.19	0.64 ± 0.06	7.8 ± 0.89	-	-	-	2
mean ± SD	1.11 ± 0.48	1.03 ± 0.45	2.40 ± 0.97	2.44 ± 1.32	0.71 ± 0.42	0.89 ± 0.51	-	-	-	1
mean ± SE	1.16 ± 0.11	0.80 ± 0.10	0.93 ± 0.19	3.13 ± 0.34	2.61 ± 0.22	3.73 ± 0.52	-	0.24 ± 0.05	0.07 ± 0.01	3
mean ± SE	10.2 ± 1.56	-	14.8 ± 1.38	31.1 ± 7.25	15.0 ± 3.1	11.1 ± 3.97	-	-	-	4
mean ± SE	192 ± 30	-	29.1 ± 3.5	1050 ± 185	4158 ± 830	2000 ± 426	-	-	-	4
mean ± SE	53.5 ± 4.74	-	27.1 ± 10.5	262 ± 28.4	817 ± 39.5	590 ± 176	-	-	-	4
mean ± SD	1.11 ± 0.64	0.90 ± 0.56	1.14 ± 0.75	1.19 ± 0.63	1.66 ± 1.25	17.6 ± 14.9	-	0.94 ± 0.67	0.02 ± 0.05	5
mean ± SD	9.26 ± 23.3	9.13 ± 23.4	1.66 ± 0.68	1.22 ± 0.57	2.61 ± 2.7	29.8 ± 54.8	-	1.07 ± 0.79	0.06 ± 0.14	5
mean ± SD	1.22 ± 0.8	1.22 ± 0.63	2.64 ± 1.30	1.71 ± 0.63	3.38 ± 2.98	63.3 ± 77.4	-	1.47 ± 0.70	0.12 ± 0.22	5
mean ± SD	1.13 ± 0.96	1.09 ± 0.81	1.01 ± 1.02	1.13 ± 0.81	1.16 ± 0.96	14.2 ± 16.1	-	1.06 ± 0.86	0.02 ± 0.04	5
mean ± SD	1.03 ± 0.54	0.91 ± 0.45	0.47 ± 0.27	1.29 ± 0.69	1.25 ± 0.73	12.7 ± 8.80	-	0.99 ± 0.70	0.04 ± 0.03	5

3. Fisk (2002c); as in reference 2.
4. Fisk *et al.* (2002a); as in reference 2.
5. Muir *et al.* (2000a); ΣCBz = sum of 1234-tetra, penta- and hexachlorobenzene; ΣHCH = sum of α-,β-,γ-HCH; ΣCHL = sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor; ΣDDT = sum of *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT; ΣPCB = sum of 103 congeners (1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33,20,53,51, 22,45,46,52,49,43,48/47,44,59,42,64/41/71,40,100,63,74,70/76/98,66,95,91,55,56/60,92,84,101,99,119,83,97,87/81,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146, 153,132,105,141,179,137,176,130,138/163,158,129,178,175,187/182,183,128,167,185,174,177,202/171,156,173,157/201,172,197,180,193,191,200,170/190,198,199,203/196,189,208/195, 207,194205,206,209).

ANNEX TABLE 7 Mercury, cadmium, arsenic, selenium and lead in seabirds from the Canadian Arctic

Species (common name)	Region	Location	Tissue	Year	Age	Sex
Fulmarus glacialis (Northern fulmars)	Lancaster Sound	Prince Leopold Island	eggs	1998	-	-
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Rissa tridactyla (Black-legged kittiwakes)	Lancaster Sound	Prince Leopold Island	eggs	1998	-	-
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Uria lomvia (Thick-billed murres or	Lancaster Sound	Prince Leopold Island	eggs	1998	-	-
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Cepphus grylle (Black or Common guillemot)	Lancaster Sound	Prince Leopold Island	eggs	1998	-	-
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Alle alle (Dovekie or Little auk)	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Larus hyperboreus (Glaucous gull)	Lancaster Sound	Prince Leopold Island	eggs	1998	-	-
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	-
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	-
Pagophila eburnea (Ivory gull)	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	f
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m
Larus thayeri (Thayler gulls)	Northern Baffin Bay	Northwater polynya	liver	1998–99	-	m
	Northern Baffin Bay	Northwater polynya	muscle	1998–99	-	m

"-" = no data available. *References:* 1. Braune (2000; 2001a). 2. Fisk (2002c).

n	Statistic	Hg (μg/g, dw)	Cd (μg/g, dw)	As (μg/g, dw)	Se (μg/g, dw)	Pb (μg/g, dw)	Reference
-	5 pools of 3	mean ± sd	1.4 ± 0.1	-	-	3.3 ± 0.2	1
5	mean ± se	2.90 ± 0.84	17.35 ± 3.75	5.61 ± 3.37	9.49 ± 1.29	0.019 ± 0.009	2
5	mean ± se	3.92 ± 0.59	26.32 ± 7.38	8.27 ± 3.10	10.56 ± 1.01	0.008 ± .004	2
5	mean ± se	0.35 ± 0.070	0.73 ± 0.13	2.19 ± 0.59	3.93 ± 0.53	0.008 ± .003	2
5	mean ± se	0.44 ± 0.057	1.60 ± 0.70	3.25	4.26 ± 0.69	0.005 ± 0.002	2
7 pools of 3	mean ± sd	0.6 ± 0.1	-	-	2.4 ± 0.2	-	1
5	mean ± se	1.08 ± 0.25	9.37 ± 3.36	4.74 ± 1.12	10.22 ± 1.56	0.015 ± 0.003	2
5	mean ± se	1.02 ± 0.14	7.86 ± 1.68	12.93 ± 3.49	12.22 ± 2.48	0.020 ± 0.007	2
5	mean ± se	0.27 ± 0.03	0.41 ± 0.11	2.12 ± 0.31	5.97 ± 0.41	0.014 ± 0.003	2
5	mean ± se	0.33 ± 0.047	0.52 ± 0.11	3.93 ± 1.29	5.62 ± 0.90	0.018 ± 0.004	2
8 pools of 3	mean ± sd	1.2 ± 0.1	-	-	2.2 ± 0.2	-	1
3	mean ± se	0.99 ± 0.06	9.35 ± 0.31	14.97 ± 2.01	3.36 ± 0.70	0.03 ± 0.01	2
2	mean ± se	1.16 ± 0.15	11.37 ± 1.74	11.36 ± 3.85	2.16 ± 0.15	0.02 ± 0.005	2
4	mean ± se	0.33 ± 0.04	2.26 ± 1.73	2.80 ± 0.67	2.97 ± 1.50	0.12 ± 0.10	2
2	mean ± se	0.27 ± 0.03	0.64 ± 0.28	2.90 ± 0.44	1.28 ± 0.25	0.03 ± 0.01	2
9 pools of 3	mean ± sd	1.8 ± 0.1	-	-	1.7 ± 0.1	-	1
5	mean ± se	0.85 ± 0.067	7.04 ± 0.92	5.63 ± 0.41	4.91 ± 1.07	0.038 ± 0.011	2
5	mean ± se	1.49 ± 0.19	6.61 ± 0.61	7.66 ± 1.40	4.62 ± 0.54	0.029 ± 0.005	2
5	mean ± se	0.27 ± 0.036	0.37 ± 0.077	1.68 ± 0.18	1.36 ± 0.13	0.045 ± 0.01	2
5	mean ± se	0.41 ± 0.022	0.47 ± 0.057	1.46 ± 0.15	1.56 ± 0.088	0.033 ± 0.003	2
4	mean ± se	0.26 ± 0.018	5.94 ± 0.75	1.52 ± 0.25	3.75 ± 0.42	0.048 ± 0.018	2
5	mean ± se	0.28 ± 0.048	5.66 ± 0.46	1.50 ± 0.13	3.93 ± 0.41	0.45 ± 0.36	2
5	mean ± se	0.078 ± 0.009	0.50 ± 0.085	0.44 ± 0.061	1.97 ± 0.34	0.023 ± 0.006	2
5	mean ± se	0.076 ± 0.012	0.33 ± 0.071	0.51 ± 0.022	1.99 ± 0.22	0.16 ± 0.14	2
6 pools of 3	mean ± sd	3.4 ± 0.3	-	-	3.4 ± 0.1	-	1
5	mean ± se	0.79 ± 0.16	0.25 ± 0.071	2.27 ± 0.55	1.45 ± 0.13	0.026 ± 0.007	2
5	mean ± se	0.81 ± 0.087	0.17 ± 0.079	1.46 ± 0.52	1.57 ± 0.10	0.046 ± 0.031	2
2	mean ± se	0.20 ± 0.03	0.16 ± 0.06	3.07 ± 0.7	2.33 ± 0.04	0.25 ± 0.22	2
3	mean ± se	0.22 ± 0.045	0.21 ± 0.085	1.58 ± 0.89	2.10 ± 0.43	0.03 ± 0.006	2
1	-	1.94	1.77	19.6	4.74	0.065	2
1	-	0.48	0.082	5.37	1.79	< 0.002	2

ANNEX TABLE 8 Concentrations of organochlorine contaminants (ng/g wet weight) in seabirds from the Canadian Arctic

Species (common name)	Location	Tissue	Year	Sex	Age	% lipid	n (# pools)
Alle alle (Dovekie or Little auk)	Northern Baffin Bay	liver	1998	-	-	4.0 ± 0.2	7 (2)
	Northern Baffin Bay	fat	1998	-	-	63.7 ± 2.1	10
Uria lomvia (Thick-billed murres)	Prince Leopold Isl., Lancaster Sound	eggs	1998	-	-	12.9 ± 0.4	15 (5)
	Northern Baffin Bay	liver	1998	-	-	4.1 ± 0.5	10
	Northern Baffin Bay	fat	1998	-	-	60.0 ± 1.9	10
						11.8–13.7	
Cepphus grylle (Black or Common guillemot)	Saglek Bay (reference site), Labrador	liver	1999	m, f	22d	-	10
	Saglek Bay (Island sites), Labrador	liver	1999	m, f	22d	-	10
	Saglek Bay (beach site), Labrador	liver	1999	m, f	22d	-	11
	Northern Baffin Bay	liver	1998	-	-	3.4 ± 0.3	9
	Northern Baffin Bay	fat	1998	-	-	60.0 ± 6.0	7
Rissa tridactyla (Black-legged kittiwakes)	Prince Leopold Isl., Lancaster Sound	eggs	1998	-	-	9.0 ± 0.2	15 (5)
	Northern Baffin Bay	liver	1998	-	-	4.1 ± 0.5	10
	Northern Baffin Bay	fat	1998	-	-	72.4 ± 3.9	8
Pagophila eburnea (Ivory gull)	Northern Baffin Bay	liver	1998	-	-	3.4 ± 0.6	5
	Northern Baffin Bay	fat	1998	-	-	81.1 ± 5.0	4
Larus hyperboreus (Glaucous gull)	Northern Baffin Bay	liver	1998	-	-	5.6 ± 0.4	11
	Northern Baffin Bay	fat	1998	-	-	72.8 ± 4.2	11
Fulmarus glacialis (Northern fulmars)	Prince Leopold Isl., Lancaster Sound	eggs	1998	-	-	11.3 ± 0.4	15 (5)
	Northern Baffin Bay	liver	1998	-	-	3.8 ± 0.6	10
	Northern Baffin Bay	fat	1998	-	-	71.9 ± 4.0	10

"-" = no data available. nd = not detected.

1. Fisk *et al.* (2001c); ΣCBz = sum of 1,2,4,5-tetraClBz, 1,2,3,4-tetraClBz, pentaClBz and hexaClBz.; ΣHCH = sum of α-, β- and γ-HCH.; ΣCHL = sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane; ΣDDT = sum of *p,p'*- and *o,p*-DDD, -DDE, and -DDT; ΣPCB = sum of 86 congeners (1,3,4/10,7,6,8/5,19,18,17,24/27,16/32,26, 25,31,28,33,22,45,46,52,49,47,48,44,42,41/71,64,40,74,70/76,95/66,56/60,91,84/89,101,99,83,97,87,85,136,110,82,151,144/135,149,118,134,114,131,146,153,132,105,141,130/176, 179,137,138,158,178/129,175,187,183,128,185,194,196/203,189,208,195,207,194,205,206,209).
2. Buckman *et al.* (2002); OCs same as reference 1.

Statistic	ΣCBz	HCB	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Mirex	Reference
mean ± SE	2.9 ± 0.47	2.0 ± 0.33	3.5 ± 0.82	10.2 ± 1.9	9.4 ± 1.5	15.7 ± 3.2	-	-	-	1, 2
mean ± SE	102 ± 8.3	63.5 ± 5.4	149 ± 12.4	459 ± 32.8	364 ± 63.5	635 ± 63.5	-	175 ± 14.8	10.1 ± 1.1	1, 2
mean ± SE	53 ± 2.0	-	17 ± 1.0	30 ± 0.4	100 ± 7.0	130 ± 9.0	-	15 ± 1.0	3.0 ± 0.5	3
mean ± SE	11.7 ± 1.3	9.8 ± 1.2	2.6 ± 0.35	6.7 ± 0.72	33.4 ± 3.8	37.3 ± 5.1	-	-	-	
mean ± SE	178 ± 15.0	149 ± 13.6	50.6 ± 4.1	158 ± 14.9	658 ± 57.2	772 ± 62.2	-	76.4 ± 9.6	11.4 ± 1.1	1,2
range		43.9–58.9	1.66-5.79	5.59–13.6	117–166	123–274	17.8–40.8	4.29–14.2		
mean	-	2.3	-	0.39	1.8	25	-	1.2	-	4
range						18–34				
mean	-	3.4	-	0.64	4.1	73	-	1.2	-	4
range						46–117				
mean	-	8.1	-	nd	8.9	830	-	nd	-	4
range						340–2030				
mean ± SE	8.6 ± 1.4	7.5 ± 1.2	5.7 ± 0.71	24.9 ± 2.5	54.5 ± 11.0	59.6 ± 11.3	-	-	-	1, 2
mean ± SE	277 ± 32.4	222 ± 25.4	161 ± 20.6	681 ± 75.9	602 ± 74.6	1263 ± 177	-	135 ± 15	16.7 ± 2.3	1, 2
mean ± SE	29 ± 4.0	-	5.0 ± 0.8	58.0 ± 9.0	60.0 ± 8.0	281 ± 22.0	-	9.0 ± 0.2	10.0 ± 7.0	3
mean ± SE	13.2 ± 1.2	11.6 ± 1.1	2.5 ± 0.3	25.8 ± 3.0	58.9 ± 6.5	143 ± 13.4	-	-	-	1, 2
mean ± SE	223 ± 84.7	186 ± 19.1	32.8 ± 3.0	385 ± 33.9	921 ± 147	3343 ± 480	-	103 ± 10.7	64.2 ± 10.0	1, 2
mean ± SE	20.1 ± 2.8	18.3 ± 2.6	8.5 ± 2.3	89.7 ± 25.9	260 ± 50.6	325 ± 71.3	-	-	-	1, 2
mean ± SE	459 ± 129	396 ± 108	112 ± 21.1	1618 ± 297	5717 ± 463	11664 ± 796	-	292 ± 41.9	203 ± 15.9	1, 2
mean ± SE	30.3 ± 2.1	26.1 ± 1.8	23.9 ± 3.6	120 ± 12.0	382 ± 52	453 ± 75.2	-	-	-	1, 2
mean ± SE	508 ± 35.8	427 ± 31.9	327 ± 39.0	1985 ± 196	4336 ± 355	11719 ± 1891	-	389 ± 53.9	226 ± 54.5	1, 2
mean ± SE	35 ± 4.0	-	2.0 ± 0.2	96 ± 10	209 ± 22	268 ± 20	-	14 ± 1.0	13 ± 2.0	3
mean ± SE	18.7 ± 1.7	17.4 ± 1.6	1.8 ± 1.7	95.7 ± 6.9	174 ± 22.9	158 ± 18.5	-	-	-	1, 2
mean ± SE	460 ± 31.2	410 ± 29.9	45.2 ± 3.0	1195 ± 92.1	3502 ± 422	6832 ± 894	-	133 ± 17.0	162 ± 24.0	1, 2

3. Braune (2001; 2001a); ΣClBz (chlorobenzene) = sum of 1,2,4,5-tetraClBz, 1,2,3,4-tetraClBz, pentaClBz and hexaClBz.; ΣHCH (hexachlorocyclohexane) = sum of α-HCH, β-HCH and γ-HCH.; ΣCHLOR (chlordanes) = sum of heptachlor, heptachlor epoxide, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane.; ΣDDT = sum of *p,p'*-DDD, *o,p*-DDD, *p,p'*-DDE, *o,p*-DDE, *p,p'*-DDT and *o,p*-DDT.; ΣPCB = sum of congeners 1, 3, 4/10, 7, 6, 8/5, 19, 18, 17, 24/27, 16/32, 26, 25, 31, 28, 33, 22, 45, 46, 52, 49, 47, 48, 44, 42, 41/71, 64, 40, 74, 70/76, 95/66, 56/60, 91, 84/89, 101, 99, 83, 97, 87, 85, 136, 110, 82, 151, 144/135, 149, 118, 134, 114, 131, 146, 153, 132, 105, 141, 130/176, 179, 137, 138, 158, 178/129, 175, 187, 183, 128, 185, 194, 196/203, 189, 208, 195, 207, 194, 205, 206 and 209.

4. Kuzyk *et al.* (2002); ΣCHL = *cis*-nonachlor; ΣDDT = *p,p'*-DDE; ΣPCB = sum of 45 congeners; n = 1,2,1 for pesticide concentrations.

ANNEX TABLE 9 Mercury, cadmium, arsenic, selenium and lead in marine mammals from the Canadian Arctic

Species (common name)	Region	Location	Tissue	Year	Age	n
Phoca hispida (Ringed seals)	Nunavut	Arctic Bay	liver	1998–2000	8.1	25
		Resolute	liver	1998–2000	5.8	23
		Pond Inlet	liver	1998–2000	3.3	25
		Pangnirtung	liver	1998–2000	5.3	24
		Arviat	liver	1998–2000	18.7	24
		Grise Fiord	liver	1998–2000	18.7	20
	Labrador	Nain & Makkovik	liver	1998–2000	5.9	28
	Nunavik, QC	Ungava Bay	liver	1998–2000	5.1	29
		Hudson Strait	liver	1998–2000	5.2	24
	Nunavut	Arctic Bay	kidney	1998–2000	8.1	25
		Resolute	kidney	1998–2000	6.0	25
		Pond Inlet	kidney	1998–2000	3.1	23
		Grise Fiord	kidney	1998–2000	18.7	20
	Labrador	Nain & Makkovik	kidney	1998–2000	5.9	28
	Nunavik, QC	Ungava Bay	kidney	1998–2000	4.9	31
		Hudson Strait	kidney	1998–2000	5.2	24
	Labrador	Nain & Makkovik	muscle	1998–2000	-	28
	Nunavik, QC	Ungava Bay	muscle	1998–2000	-	13
		Hudson Strait	muscle	1998–2000	-	22
Odobenus rosmarus (Walrus)	Nunavik, QC	Inukjuaq	muscle	1999	-	6
	Nunavik, QC	Inukjuaq	liver	1999	-	5
	Nunavik, QC	Inukjuaq	kidney	1999	-	7
Delphinapterus leucas (Beluga whales)	NWT	Mackenzie delta	liver	1981	12.2 ± 4.46	25
		Mackenzie delta	liver	1984	21.1 ± 9.36	8
		Mackenzie delta	liver	1993	22.2 ± 6.27	25
		Mackenzie delta	liver	1994	17.7 ± 6.21	41
		Mackenzie delta	liver	1995	15.7 ± 5.43	18
		Mackenzie delta	liver	1996	14.6 ± 5.68	10
		Mackenzie delta	liver	2001	15.5 ± 5.24	24
	Nunavut	Arviat	liver	1984	11.8 ± 6.29	21
		Arviat	liver	1999	11.1 ± 4.84	36
		Coral Harbour	liver	1993	16.1 ± 3.22	11
		Coral Harbour	liver	1997	13.1 ± 7.31	19
		Coral Harbour	liver	2000	8.92 ± 5.90	24
		Grise Fiord	liver	1984	5.65 ± 4.83	17
		Iqaluit	liver	1993	12.9 ± 3.48	23
		Iqaluit	liver	1994	12.9 ± 6.34	7
		Lake Harbour, Kimmirut	liver	1994	10.8 ± 6.78	19
		Nastapoka River	liver	1984	13.2 ± 7.67	14
		Pangnirtung	liver	1984	11.1 ± 3.81	11
		Pangnirtung	liver	1993	8.23 ± 3.77	11
		Pangnirtung	liver	1994	8.41 ± 6.32	27
		Pangnirtung	liver	1997	13.0 ± 4.19	24
		Paulatuk	liver	1993	14.3 ± 5.13	3
		Repulse Bay	liver	1993	8.00 ± 8.49	2
		Sanikiluaq	liver	1994	13.7 ± 5.52	30
		Sanikiluaq	liver	1998	13.0 ± 5.37	22
		Kangiqsujuaq & Puvirnituq	liver	1999	6–28	15

"-" = no data available. *References:* 1. Muir *et al.* (1999a). 2. Muir *et al.* (2001b). 3. Muir *et al.* (2000a). 4. Lockhart *et al.* (2001b). 5. Sang *et al.* (2000).

Statistic	Hg (μg/g, dw)	Cd (μg/g, dw)	As (μg/g, dw)	Se (μg/g, dw)	Pb (μg/g, dw)	Reference
mean ± 95% CI	7.83 ± 3.55	5.13 ± 1.28	0.49 ± 0.12	6.73 ± 1.58	0.014 ± 0.002	1
mean ± 95% CI	3.08 ± 2.50	5.98 ± 4.01	0.63 ± 0.17	3.39 ± 1.40	0.022 ± 0.008	1
mean ± 95% CI	4.00 ± 2.58	9.33 ± 4.05	0.87 ± 0.25	4.42 ± 1.48	0.010 ± 0.004	1
mean ± 95% CI	6.04 ± 4.34	8.91 ± 6.28	0.49 ± 0.09	5.33 ± 2.63	0.015 ± 0.006	1
mean ± 95% CI	14.0 ± 8.79	7.57 ± 4.25	0.28 ± 0.04	12.5 ± 5.04	0.005 ± 0.003	1
mean ± 95% CI	18.4 ± 11.0	5.75 ± 1.76	1.30 ± 0.35	12.1 ± 5.17	0.016 ± 0.006	1
mean ± 95% CI	6.31 ± 3.71	2.85 ± 0.76	0.26 ± 0.05	6.22 ± 1.19	0.004 ± 0.003	2
mean ± 95% CI	10.8 ± 6.29	4.72 ± 1.61	0.17 ± 0.02	7.80 ± 2.87	-	2
mean ± 95% CI	3.50 ± 3.24	2.74 ± 3.40	0.33 ± 0.08	4.57 ± 4.57	0.005 ± 0.003	2
mean ± 95% CI	1.56 ± 0.26	19.1 ± 5.02	0.31 ± 0.05	2.57 ± 0.17	0.020 ± 0.011	1
mean ± 95% CI	1.39 ± 0.27	24.3 ± 10.5	0.36 ± 0.06	1.95 ± 0.19	0.010 ± 0.003	1
mean ± 95% CI	1.23 ± 0.26	25.1 ± 7.91	0.45 ± 0.09	2.82 ± 0.25	0.009 ± 0.001	1
mean ± 95% CI	2.75 ± 0.59	18.8 ± 4.67	0.76 ± 0.10	2.70 ± 0.23	0.199	1
mean ± 95% CI	1.04 ± 0.13	5.71 ± 1.84	0.37 ± 0.12	2.22 ± 0.17	0.015 ± 0.003	2
mean ± 95% CI	1.08 ± 0.21	9.31 ± 2.85	0.17 ± 0.02	2.03 ± 0.17	0.014 ± 0.002	2
mean ± 95% CI	0.69 ± 0.14	5.51 ± 3.59	0.26 ± 0.05	2.00 ± 0.17	0.012 ± 0.001	2
mean ± 95% CI	0.328 ± 0.085	0.013 ± 0.006	0.089 ± 0.080	0.428 ± 0.039	0.001 ± 0.001	2
mean ± 95% CI	0.251 ± 0.078	-	-	-	-	2
mean ± 95% CI	0.182 ± 0.061	0.008 ± 0.012	4.335 ± 0.000	0.396 ± 0.024	-	2
mean	0.040	0.03	0.15	2.02	0.011	3
range	00.03–0.05	0.01–0.07	0.09–0.20	1.38–3.32	0.006–0.022	
mean	2.64	3.320	0.27	2.59	0.098	3
range	1.46–3.81	0.52–8.68	0.14–0.40	1.72–3.43	0.030–0.146	
mean	0.31	16.5	0.22	3.58	0.184	3
range	1.46–3.81	0.52–8.68	0.14–0.40	1.72–3.43	0.030–0.146	
mean ± SD	10.7 ± 10.7	-	-	-	-	4
mean ± SD	17.8 ± 16.5	-	-	-	-	4
mean ± SD	34.5 ± 27.4	-	-	-	-	4
mean ± SD	28.4 ± 29.3	-	-	-	-	4
mean ± SD	44.0 ± 35.1	-	-	-	-	4
mean ± SD	43.8 ± 31.3	-	-	-	-	4
mean ± SD	38.9 ± 41.7	-	-	-	-	4
mean ± SD	7.25 ± 6.94	-	-	-	-	4
mean ± SD	12.5 ± 10.0	-	-	-	-	4
mean ± SD	6.54 ± 2.97	-	-	-	-	4
mean ± SD	13.8 ± 29.3	-	-	-	-	4
mean ± SD	4.10 ± 2.40	-	-	-	-	4
mean ± SD	2.00 ± 1.72	-	-	-	-	4
mean ± SD	7.57 ± 4.86	-	-	-	-	4
mean ± SD	16.3 ± 8.93	-	-	-	-	4
mean ± SD	8.80 ± 6.16	-	-	-	-	4
mean ± SD	11.5 ± 13.9	-	-	-	-	4
mean ± SD	5.05 ± 4.43	-	-	-	-	4
mean ± SD	8.45 ± 7.00	-	-	-	-	4
mean ± SD	10.7 ± 13.4	-	-	-	-	4
mean ± SD	8.73 ± 4.67	-	-	-	-	4
mean ± SD	8.58 ± 10.0	-	-	-	-	4
mean ± SD	3.42 ± 3.09	-	-	-	-	4
mean ± SD	12.9 ± 9.53	-	-	-	-	4
mean ± SD	21.1 ± 25.3	-	-	-	-	4
(mean) & range	(11.6) 3.40–17.7	(7.48) 2.18–14.5	(6.83) 3.70–15.1	(0.018) < 0.006–0.04	(0.13) 0.10–0.14	5

ANNEX TABLE 10 Concentrations of organohalogen contaminants (ng/g wet weight) in marine mammals from the Canadian Arctic

Species (common name)	Location	Tissue	Year	Age, years	Sex	% lipid	n
Phoce hispida (Ringed seals)							
	Holman, NWT	blubber	2001	8–25	f	85.7 ± 10.7	5
	Holman, NWT	blubber	2001	2–14	m	83.0 ± 6.38	5
	Hudson Strait (Salluit+Quaqtaq)	blubber	1998	-	f	80.4 ± 10.4	10
	Hudson Strait (Salluit+Quaqtaq)	blubber	1998	-	m	77.0 ± 9.8	11
	Ungava Bay (George River, Kangnirsuk, Kuujjuaq)	blubber	1998	-	f	101 ± 16.5	16
	Ungava Bay (George River, Kangnirsuk, Kuujjuaq)	blubber	1998	-	m	92.4 ± 15.7	5
	Labrador (Nain+Makkovik)	blubber	1998	-	f	87.3 ± 5.6	6
	Labrador (Nain+Makkovik)	blubber	1998	-	m	88.2 ± 5.5	14
	Nain, Labrador	blubber	1998	1–24	m	-	4
	Saglek Bay, Labrador	blubber	1997 & 98	1–11	m	-	6
	Arctic Bay, Nunavut	blubber	2000	6.9 ± 3.5	f	94.1 ± 4.7	7
	Arviat, Nunavut	blubber	2001	17.1±6.3	f	81.8 ± 25.8	10
	Pangnirtung, Baffin Island	blubber	1998	-	f	-	3
	Pangnirtung, Baffin Island	blubber	1998	-	m	-	3
	Eureka, Ellesmere Island	blubber	1993	-	m, f	-	6
	Pangnirtung, Baffin Island	blubber	1994	-	m	-	3
	Pangnirtung, Baffin Island	blubber	1994	-	f	-	3
	Grise Fiord, Nunavut	blubber	1998	19 ± 3.9	f	90.4 ± 1.8	15
	Grise Fiord, Nunavut	blubber	1998	14.8 ± 3.0	m	91.2 ± 1.4	12
Odobenus rosmarus (Walrus)							
	E. Hudson Bay	blubber	1999	-	-	84.3 ± 3.96	6
Delphinapterus leucas (Beluga whales)							
	Hendrikson Island	brain	1992	-	m	8.88	3
						2.16–13.1	
	Hendrikson Island	liver	1992	-	m	2.82	3
						2.01–3.71	
	Hendrikson Island	muscle	1992	-	m	0.98	3
						0.61–1.31	
	Hendrikson Island	blubber	2001			92.2 ± 4.21	10
	Hendrikson Island	blubber	1995	15.3 ± 4.8	m		
	Mackenzie Bay	blubber	1995	-	m	-	-
	Naskapoka River, E. Hudson Bay	blubber	1999	-	f		2
	Naskapoka River, E. Hudson Bay	blubber	1999	-	m		8
	W. Hudson Bay	blubber	1993	-	f	35.3 ± 16.9	4
	W. Hudson Bay	blubber	1993	-	m	40.7 ± 19.0	6
	Pangnirtung, Baffin Island	blubber	1982	9.7 ± 5.4	m	90.8 ± 2.4	8
	Pangnirtung, Baffin Island	blubber	1986	5.6 ± 1.7	m	91.2 ± 2.3	17
	Pangnirtung, Baffin Island	blubber	1992	12.9 ± 4.9	m	90.9 ± 2.4	11
	Pangnirtung, Baffin Island	blubber	1996 & 97	13.5 ± 5.4	m	89.8 ± 4.1	17
	Kimmirut, NT	blubber	1994	21± 18	f	92.9 ± 1.7	3
	Kimmirut, NT	blubber	1994	6 ± 6	m	93.5 ± 1.9	3
Monodon monoceros (Narwhal)							
	Broughton Island, E. Baffin Island	blubber	1993–96	-	f	84.4 ± 4.0	18
	Broughton Island, E. Baffin Island	blubber	1993–97	-	m	86.6 ± 4.5	32
	Grise Fiord, Jones Sound	blubber	1993 & 95	-	f	82.5 ± 5.4	2
	Grise Fiord, Jones Sound	blubber	1995–99	-	m	87.6 ± 4.6	13
	Pangnirtung, SE Baffin Island	blubber	1996	-	m	91.2 ± 3.9	5
	Pond Inlet, NE Baffin Island	blubber	1994–99	-	f	92.1 ± 4.8	10
	Pond Inlet, NE Baffin Island	blubber	1992–99	-	m	92.6 ± 3.7	14

"-" = no data available. nd = not detected. SCCP = short chain chlorinated paraffin.

References:

1. Hoekstra (2002c); ΣCBz = sum of 1,2-diClBz, 1,4-diClBz, 1,2,3-triClBz, 1,2,4-triClBz, 1,3,5-triClBz, 1,2,3,4-tetraClBz, 1,2,3,5-tetraClBz, pentaClBz, hexaClBz; ΣCBz = sum of 1,2-diClBz, 1,4-diClBz, 1,2,3-triClBz, 1,2,4-triClBz, 1,3,5-triClBz, 1,2,3,4-tetraClBz, 1,2,3,5-tetraClBz, pentaClBz, hexaClBz; ΣHCH = sum of α-, β-, γ-, δ-HCH; ΣCHLOR = sum of *cis*- and *trans*-chlordane, oxychlordane, *cis*- and *trans*-nonachlor, heptachlor, *cis*-heptachlor epoxide; ΣDDT = sum of *o,p'*- and *p,p'*-DDD, -DDE and -DDT; ΣPCB = sum of 101 congeners (4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16,32,54/29,26,25,50, 31/28,33/21/53,51,22,45,46,52/49,43,47/48,44,59,42,64,41/71,40,100,63,74,76/98,70,95,66,91,55,56/60,92/84,101,99,119,83,97,87,81,85,136,110,82,151,135,144,107/147,149/133,118, 114,143,141,145,153,132,105,141/179,137,176/130,163,138,158,129/178,175,187,182,183,128,167,185,174,177,171,156,202/173,172,197,180/193,191,199,170/190,198,201,176/203, 189,206,195,207,194,205,208,209).
2. Muir *et al.* (1999a); ΣHCH = sum of α-, β-, γ-HCH, ΣCHL = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, ΣDDT = sum *o,p'*- and *o,p'*-DDE, -DDD and -DDT, ΣPCB = sum 103 congeners (1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33,20,53,51,22,45,46,52,49,43,48/47,44,59,42,64/41/71,40,100,63,74,70/76/98,66,95,91, 55,56/60,92,84,101,99,119,83,97,87/81,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146,153,132,105,141,179,137,176,130,138/163,158,129,178,175,187/182,183,128, 167,185,174,177,202/171,156,173,157/201,172,197,180,193,191,200,170/190,198,199,203/196,189,208/195,207,194,205,206,209), Toxaphene = total toxaphene.
3. Environmental Sciences Group (ESG) (2002).
4. Muir *et al.* (2001b); ΣHCH = sum of α-, β- and γ-HCH, ΣCHL = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor, ΣDDT = sum *o,p'*- and *o,p'*-DDE, -DDD and -DDT, ΣPCB = sum of 86 peaks (103 congeners;1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33,20,53,51,22,45,46,52,49,43,48/47,44,59,42,64/41/71,40,100,63,74,70/76/98, 66,95,91,55,56/60,92,84,101,99,119,83,97,87/81,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146,153,132,105,141,179,137,176,130,138/163,158,129,178,175,187/182,183, 128,167,185,174,177,202/171,156,173,157/201,172,197,180,193,191,200,170/190,198,199,203/196,189,208/195,207,194,205,206,209); Toxaphene = total toxaphene.
5. Muir *et al.* (1999d); ΣDDT = *o,p'* and *p,p'*-DDD, DDT and DDE; ΣPCB = 102 congeners; Toxaphene = determined by GC-ECD, ΣPBDE = sum of mono,di, tri tetra, penta, hexa and hepta brominated diphenyl ethers.
6. Tomy *et al.* (2000).
7. Fisk *et al.* (2002c); ΣCBz = sum of 1,2,4,5-CBz, 1,2,3,4-triClBz, pentaCBz, hexaCBz; ΣHCH = sum of α-, β-, γ-HCH; ΣCHLOR = sum of heptachlor, heptachlor epoxide, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane. ΣDDT = sum of *p,p'*- and *o,p'*-DDD, -DDE and -DDT; ΣPCB = sum of 77 congeners (1,3,4/10,7,6,8/5,19,18,17,24/27,16/32,26,25,31,28,33,22,45,46,52,49,47,48,44,42, 41/71,64,40,74,70/76,95/66,56/60,91,84/89,101,99,83,97,87,85,136,110,82,151,144/135,149,118,134,114,131,146,153,132,105,141,130/176,179,137,138,158,178/129,175,187,183,128, 185,194,196/203,189,208,195,207,194,205,206,209).

Statistic	ΣCBz	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Dieldrin	Mirex	ΣPBDE	SCCPs	Reference
mean ± SD	64.0 ± 15.3	234 ± 137	367 ± 87.0	188 ± 37.6	335 ± 19.9	-	50.4 ± 4.49	11.1 ± 2.83	-		1
mean ± SD	64.6 ± 29.4	225 ± 131	514 ± 218	263 ± 134	487 ± 306	-	44.2 ± 6.93	7.59 ± 2.20	-		1
mean ± SD	5.9 ± 2.8	117 ± 38.4	190 ± 162	405 ± 403	608 ± 500	266 ± 250	43.7 ± 16.5	5.3 ± 4.9	-		2
mean ± SD	9.0 ± 13.3	140 ± 91.7	303 ± 493	884 ± 2130	1010 ± 1860	309 ± 218	70.6 ± 97.3	10.8 ± 27.4	-		2
mean ± SD	8.2 ± 2.8	155 ± 63.3	161 ± 78.6	259 ± 142	513 ± 225	87.7 ± 55.1	58.3 ± 39.0	4.1 ± 3.1	-		2
mean ± SD	11.4 ± 4.4	299 ± 171	81.1 ± 24.6	728 ± 526	1040 ± 670	117 ± 82.4	78.4 ± 24.6	6.5 ± 6.5	-		2
mean ± SD	5.1 ± 3.7	103 ± 41.8	109 ± 62.0	187 ± 107	730 ± 450	nd	32.1 ± 19.8	3.4 ± 3.0	-		2
mean ± SD	5.2 ± 0.2	90.8 ± 47.9	108 ± 45.1	198 ± 86.0	572 ± 290	nd	31.6 ± 21.2	2.3 ± 1.9	-		2
mean/range	-	-	-	228	344–625	-	-	-	-		3
range	-	-	-	? (higher than 228)	500–9400	-	-	-	-	3	
mean ± SD	10.4 ± 4.5	133 ± 87.0	172 ± 43.3	200 ± 108	358 ± 145	-	57.4 ± 17.0	4.0 ± 4.2	-		4
mean ± SD	16.8 ± 29.4	79.6 ± 39.8	241 ± 170	463 ± 533	728 ± 707	-	45.1 ± 35.4	11.7 ± 6.6			
mean ± SD	-	-	-	510 ± 190	585 ± 180	207 ± 33	-	-	0.50 ± 0.18		5
mean ± SD	-	-	-	1200 ± 1660	976 ± 1090	185 ± 121	-	-	0.97 (n = 2)		5
mean ± SD	-	-	-	-	-	-	-	-	-	527 ± 164	6
mean ± SD	-	-	-	-	-	-	-	-	-	100 ± 27	5
mean ± SD	-	-	-	-	-	-	-	-	-	89 ± 44	5
mean ± SE	42 ± 4	148 ± 12	338 ± 38	394 ± 89	483 ± 86	-	-	-	-		7
mean ± SE	47 ± 10	179 ± 38	854 ± 168	963 ± 271	1000 ± 206	-	-	-	-		7
mean ± SD	0.33 ± 0.06	104 ± 47.8	291 ± 167	53.9 ± 34.5	300 ± 157	-	-	-	-		8
mean	45.3	24.9	13.1	46	130	-	11.8	1.56	-		9
range	32.0–68.3	19.4–30.4	9.90–13.5	21.9–86.5	107–173		-	-			
mean	37.9	2.7	19.8	73.6	132	-	13.6	2.05	-		9
range	17.0–70.4	2.1–3.3	17.2–23	39.0–100	85.8–172		-	-			
mean	10.7	1.0	9	28.2	56.1	-	4.71	0.70	-		9
range	9.0–12.8	0.5–1.4	8.1–10.4	16.5–43.9	32.9–81.5		-	-			
mean ± SD	418 ± 265*	172 ± 97.4	929 ± 61	1120 ± 846	2790 ± 1060	2330 ± 925	-	-	-		10
								15500 ± 4400		10	
-	-	-	-	-	-	-	-	-	-	206 ± 96	6
mean	203*	120	904	1300	2220	-	-	-	-		11
range	88.4–318	97.1–142	784–1020	1240–1360	2140–2300						
mean ± SD	253 ± 97*	119 ± 50.9	1460 ± 768	2630 ± 1930	3550 ± 1950	-	-	-	-		11
range	97.2–414	62.2–205	578–2860	732–6480	1260–7010						
mean ± SD	256 ± 127*	171 ± 59.2	1240 ± 232	1070 ± 172	1490 ± 225	-	318 ± 124	15.4 ± 5.04	-		12
mean ± SD	340 ± 76.7*	216 ± 57.2	1590 ± 293	2100 ± 857	2200 ± 363	-	477 ± 134	20.2 ± 4.47	-		12
mean ± SD	455 ± 66*	267 ± 37	1650 ± 372	5340 ± 2550	4230 ± 1360	9650 ± 2390	616 ± 108	-	-		13
mean ± SD	379 ± 93*	261 ± 69	1420 ± 313	3280 ± 1050	2570 ± 609	7760 ± 1790	445 ± 102	-	-		13
mean ± SD	343 ± 182*	189 ± 48	2030 ± 519	5650 ± 1870	3990 ± 1140	9430 ± 2540	387 ± 119	-	-		13
mean ± SD	694 ± 226*	217 ± 57	2110 ± 552	5570 ± 1740	4570 ± 942	10600 ± 3850	568 ± 215	-	-		14
mean ± SD	-	-	-	3540 ± 2620	4390 ± 3120	7350 ± 4460	-	-	-	116 ± 52	5
mean ± SD	-	-	-	7110 ± 3360	7650 ± 2870	16300 ± 8890	-	-	-	168 ± 35	5
mean ± SD	440 ± 177*	112 ± 41	1470 ± 482	3610 ± 1340	3890 ± 1330	5150 ± 1410	-	-	-		10
mean ± SD	512 ± 191*	126 ± 48	1750 ± 610	4830 ± 2270	4820 ± 1830	7440 ± 4030	-	-	-		10
mean ± SD	332 ± 115*	81 ± 3.4	1420 ± 96	3800 ± 417	3500 ±440	8090 ± 210	-	-	-		10
mean ± SD	369 ± 130*	102 ± 32	1500 ± 450	4020 ± 1750	3440 ± 955	7950 ± 1960	-	-	-		10
mean ± SD	458 ±70*	117 ± 31	1850 ± 348	4950 ± 1990	4600 ± 1090	8760 ± 3670	-	-	-		10
mean ± SD	344 ± 84*	114 ± 32	1610 ± 525	4470 ± 945	2980 ± 721	5200 ± 1470	-	-	-		10
mean ± SD	672 ± 362*	165 ± 74	2600 ± 1060	7220 ± 2380	5820 ± 2020	11800 ± 4440	-	-	-		10

8. Muir and Kwan (2000); ΣHCH = sum of α-, β-, γ-HCH; ΣCHL = sum of oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, heptachlor, heptachlor epoxide; ΣDDT = sum of 2,4'- and 4,4'-DDE,-DDD,-DDT; ΣPCB = sum of 92 congeners (1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33/20,53,51,22,45,46,52,43,49,47/48,44,59,42,71/41/64,40,100,63, 74,70/76/98,66,95,91,55,56/60,92,84,101,99,119,83,97,81/87,85,136,110,82,151,135/144,147,137,176,130,163/138,158,129,178,175,182/187,183,128,167,185,174,177,202/171,156,173, 157/200,172,197,180,193,191,199,170/190,198,201,203/196,189,208/195,207,194,205,206,209).
9. Metcalfe *et al.* (1999); ΣHCH = sum of α-, β-, γ-, δ-HCH; ΣCHL = sum of *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor; ΣDDT = sum of *p,p*-DDT, -DDD, -DDE; ΣPCB = sum of 27 congeners (18,28,31,52,49,47,44,66,87,99,101,110,105,118,119,151,149,153,156,138,180,170,199,195,196,194,209).
10. Stern (2001); ΣPCB = sum of 86 peaks (103 congeners), ΣDDT = sum *o,p'*- and *o,p'*-DDE, -DDD and -DDT; Toxaphene = determined by GC-ECD.
11. Sang *et al.* (2000); ΣCBz = sum of 1234-tetra, penta- and hexachlorobenzene; ΣHCH = sum of α-, β-, γ-HCH; ΣCHL = sum of heptachlor, heptachlor epoxide, *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor; ΣDDT = sum *o,p'*- and *p,p'*-DDE, -DDD and -DDT; ΣPCB = sum of 103 congeners (1,3,4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16/32,54/29,26,25,31/28,50,33,20,53,51,22,45,46,52,49, 43,48/47,44,59,42,64/41/71,40,100,63,74,70/76/98,66,95,91,55,56/60,92,84,101,99,119,83,97,87/81,85,136,110,82,151,135/144,147,107,149,118,133,114,134/131,146,153,132,105,141,179, 137,176,130,138/163,158,129,178,175,187/182,183,128,167,185,174,177,202/171,156,173,157/201,172,197,180,193,191,200,170/190,198,199,203/196,189,208/195,207,194,205,206,209).
12. Hobbs *et al.* (2002); ΣCBz = sum of 1,2,4,5-TCB, 1,2,3,4-TCB penta-CBz,HCBz; ΣHCH = sum of α-, β-, γ-HCH; ΣCHL = sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, nonachlor, C, C3, C5, heptachlor, C1B/U6, C2/U5, heptachlor epoxide; ΣDDT = sum of *o,p'*- and *p,p'*-DDE,-DDD,-DDT; ΣPCB = sum of 85 congeners (1,3,4/10,7,6,8/5,19,18,17,24/27,16/32,26,25,31,28,33,22, 45,46,52,49,47/48,44,42,41/71/64,40,74,70/76,66,95,56/60,91,84/89,101,99,83,97,87,85,136,110,82,151,144/135,149,118,134/131,114,146,153,132,105,141,130/176,179,137,138,158,178/129, 175,187,183,128,185,174,177,171,156,201/157,172/197,180,193,191,200,170/190,198,199,196/203,189,208/195,207,194,205,206,209).
13. Stern (1999); ΣCBz = sum of 1,2,4,5-TCB, 1,2,3,4-TCB penta-CBz,HCBz; ΣHCH = α-, β, -γ-HCH; ΣCHL = *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane, heptachlor, heptachlor epoxide, nonachlor III; ΣDDT = sum of *o,p'* and *p,p'*-DDE,-DDD,-DDT; ΣPCB = sum of 90 PCB congeners; Toxaphene quantified using a single response factor based on 27 peaks in a technical mixture (primarily Parlar 26, 50).
14. Stern and Addison, 1999; as in reference 12.

ANNEX TABLE 11 Concentrations of organochlorine contaminants (ng/g wet weight) in Arctic fox and polar bear from the Canadian Arctic

Species (common name)	Location	Tissue	Year	Age, years	Sex	% lipid	n	Statistic
Alopex lagopus (Arctic fox)	Holman, NWT	muscle	1999–2000	0–2	m, f	5.6 ± 0.6	20	mean ± SD
	Holman, NWT	liver	1999–2000	0–2	m, f	9.2 ± 1.3	20	mean ± SD
Ursus maritimus (Polar bears)	Resolute	whole plasma	1997	subadults	-	-	6	mean ± SD
		whole plasma	1997	adult	m	-	13	mean ± SD
		whole plasma	1997	adult	f	-	14	mean ± SD

"-" = no data available.

References:

1. Hoekstra *et al.* (2002e); ΣCBz = sum of 1,2-diClBz, 1,4-diClBz, 1,2,3-triClBz, 1,2,4-triClBz, 1,3,5-triClBz, 1,2,3,4-tetraClBz, 1,2,3,5-tetraClBz, pentaClBz, and hexaClBz; ΣHCH = sum of α-, β-, γ-, δ-HCH; ΣCHLOR = sum of *cis*- & *trans*-chlordane, oxychlordane, *cis*- & *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide; ΣDDT = sum of *o,p'*- & *p,p'*-DDD, -DDE and -DDT; ΣPCB = sum of 101 congeners (4/10,7/9,6,8/5,19,12/13,18,15/17,24/27,16,32,54/29,26,25,50,31/28,33/21/53,51,22,45,46,52/49,43,47/48,44,59,42,64,41/71,40,100,63,74,76/98,70,95,66,91,55,56/60,92/84,101,99,119,83,97,87,81,85,136,110,82,151,135,144,107/147,149/133,118,114,143,141,145,153,132,105,141/179,137,176/130,163,138,158,129/178,175,187,182,183,128,167,185,174,177,171,156,202/173,172,197,180/193,191,199,170/190,198,201,176/203,189,206,195,207,194,205,208 & 209); Toxaphene = sum of 24 chloroborane congeners.
2. Sandau (2000).

ANNEX TABLE 12 Congener specific concentrations (ng/g lw) of PBDEs in marine and freshwater biota of the Canadian Arctic

Species (common name)	Region	Tissue	Year	Sex	% lipid	n	Statistic	BDE#15	BDE#52
Freshwater fish									
Lota lota (Burbot)	Fort Good Hope, NWT	liver	1988	m, f	30.2 ± 13.5	10	mean ± SD	-	-
	Fort Good Hope, NWT	liver	1999	m, f	35.0 ± 9.59	4	mean ± SD	-	-
	Fort Good Hope, NWT	liver	2000	m, f	33.3 ± 13.1	11	mean ± SD	-	-
Marine mammals									
Phoca hispida (Ringed seal)	Holman	blubber	1996	m	-	8	mean	-	-
	Holman	blubber	2000	m	-	8	mean	-	-
	Pangnirtung, Baffin Island	blubber	1993	f	94 ± 1.5	3	mean ± SD	-	-
	Pangnirtung, Baffin Island	blubber	1993	m	95	2	mean	-	-
Delphinapterus leucas (Beluga whales)	Kimmirut, Baffin Island	blubber	1994	f	92.9 ± 1.7	3	mean ± SD	-	-
	Kimmirut, Baffin Island	blubber	1994	m	93.5 ± 1.9	3	mean ± SD	-	-
	Hendrickson Island, W. Canada	blubber	1989	m	90.1 ± 3.7	12	mean ± SD	-	-
	Hendrickson Island, W. Canada	blubber	1995	m	92.6 ± 1.8	9	mean ± SD	-	-
	Hendrickson Island, W. Canada	blubber	2001	m	93.1 ± 3.5	11	mean ± SD	-	-

References: 1. Stern *et al.* (2001). 2. Ikonomou *et al.* (2002). 3. Muir *et al.* (1999d). 4. Stern and Ikonomou (2001).

ΣCBz	α-HCH	β-HCH	γ-HCH (lindane)	ΣHCH	ΣCHL	ΣDDT	ΣPCB	Toxaphene	Reference
4.43 ± 0.63	1.5 ± 0.39	3.9 ± 0.64	-	5.40 ± 0.91	34.0 ± 8.86	1.91 ± 0.47	48.0 ± 9.15	4.5 ± 1.2	1
4.16 ± 0.95	3.1 ± 0.94	7.7 ± 1.6	-	10.9 ± 2.12	219 ± 56	3.99 ± 0.82	124 ± 39.4	17 ± 4.5	1
7.5 ± 5.2	-	-	-	5.0 ± 2.7	56 ± 46	1.5 ± 1.1	79 ± 54	-	2
3.0 ± 0.8	-	-	-	3.6 ± 1.2	14 ± 5.8	0.7 ± 0.4	42 ± 16	-	2
3.2 ± 1.7	-	-	-	2.9 ± 1.2	29 ± 16	1.7 ± 3.2	33 ± 21	-	2

BDE#47	BDE#49	BDE#85	BDE#99	BDE#100	BDE#138	BDE#153	BDE#154	BDE#183	BDE#209	ΣPBDEs	Reference
226 ± 280		-	84.5 ± 131	35.2 ± 46.7	-	29.4 ± 44.7	20.5 ± 28.9	-	-	-	1
583 ± 522		-	370 ± 270	208 ± 155	-	161 ± 125	157 ± 116	-	-	-	1
620 ± 629		-	320 ± 274	180 ± 183	-	135 ± 134	81.3 ± 84.2	-	-	-	1
2.8	-	-	-	-	-	-	-	-	-	3.44	2
3.7	-	-	-	-	-	-	-	-	-	4.62	2
0.33 ± 0.15	-	-	0.11 ± 0.01	-	-	-	-	-	-	0.05 ± 0.18	3
0.76	-	-	0.09	-	-	-	-	-	-	0.97	3
1.09 ± 0.82	-	-	0.19 ± 0.06	-	-	-	-	-	-	1.44 ± 1.01	3
2.14 ± 0.66	-	-	0.32 ± 0.07	-	-	-	-	-	-	2.84 ± 0.90	3
5.81 ± 2.26	3.45 ± 1.53	-	0.59 ± 0.27	0.60 ± 0.23	-	0.03 ± 0.02	0.11 ± 0.04	-	-	11.7 ± 4.43	4
8.76 ± 2.45	3.12 ± 1.61	-	1.03 ± 0.28	1.13 ± 0.30	-	0.12 ± 0.04	0.38 ± 0.11	-	-	15.5 ± 4.40	4
8.77 ± 4.78	1.29 ± 0.88	-	1.71 ± 0.99	1.48 ± 0.90	-	0.36 ± 0.19	0.63 ± 0.40	-	-	15.5 ± 8.52	4

ANNEX TABLE 13 Concentrations and TEQs for PCDDs, PCDFs and non-*ortho* PCBs (nPCBs) in Canadian Arctic biota

Species (common name)	Region	Location	Tissue	Year	Age	Sex	% lipid	n
Freshwater invertebrates								
Plankton	Great Slave Lake, NWT	West basin	whole body	1994–95	-	-	-	-
	Great Slave Lake, NWT	East arm	whole body	1994–95	-	-	-	-
Freshwater fish								
Coregonus sp. (Whitefish)	Great Slave Lake, NWT	West basin		1994–95	-	-	-	-
	Great Slave Lake, NWT	East arm	muscle	1994–95	-	-	-	-
Salvelinus namaycush (Lake trout)	Great Slave Lake, NWT	West basin	muscle	1994–95	-	-	-	-
	Great Slave Lake, NWT	East arm	muscle	1994–95	-	-	-	-
Lota lota (Burbot)	Great Slave Lake, NWT	West basin	muscle	1994–95	-	-	-	-
	Great Slave Lake, NWT	East arm	liver	1994–95	-	-	-	-
			liver					
Seabirds								
Rissa tridactyla (Black-legged kittiwakes)	Lancaster Sound, Nunavut	Prince Leopold Island		1975	-	-	9.0	4 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	4.3	5 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	5.1	3, pooled
Fulmarus glacialis (Northern fulmars)	Lancaster Sound, Nunavut	Prince Leopold Island	egg	1975	-	-	2.6	5 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	4.4	5 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	8.9	3, pooled
Uria lomvia (Thick-billed murres)	Lancaster Sound, Nunavut	Prince Leopold Island	egg	1975	-	-	8.6	3 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	3.3	3 pools of 3
	Lancaster Sound, Nunavut	Prince Leopold Island	liver	1993	-	-	6.8	3, pooled
Cepphus grylle (Black or Common guillemot)	Labrador	Saglek Bay (beach site)	egg	1999	22	d	-	1
Marine mammals			liver					
Phoca hispida (Ringed seals)	Nunavut, Canada	Pangnirtung, Cumberland Sound		1993	9-July	m		3
		Pangnirtung, Cumberland Sound	blubber	1993	37846	f		3
	NWT, Canada	Holman Island	blubber	2000	0–35	m, f	91.3 ± 3.1	20
Delphinapterus leucas (Beluga whales)	Nunavut, Canada	Kimmirut	blubber	1994	37627	m		3
		Kimmirut	blubber	1994	6-January	f		3
Ursus maritimus (Polar bears)	Canada	Queen Maud Gulf	blubber	1984	-	-	-	9, pooled
			adipose	1990	-	-	-	10, pooled
		Barrow Strait	adipose	1984	-	-	-	7, pooled
			adipose	1990	-	-	-	9, pooled
		North Baffin Bay	adipose	1984	-	-	-	10, pooled
			adipose	1990	-	-	-	10, pooled
		Davis Strait	adipose	1984	-	-	-	7, pooled
			adipose	1990	-	-	-	10, pooled

References:

1. Evans *et al.* (1996).
2. Braune (2002); TEQ values calculated using World Health Organization (WHO) avian toxic equivalency factors (TEFs) for PCDDs, PCDFs and non-*ortho* PCBs as given in Van den Berg *et al.* (1998).
3. Kuzyk *et al.* (2002); combined sum of ΣPCDD & ΣPCDF TEQs provided.
4. Helm *et al.* (2002); ΣnPCB = sum of 8 congeners (77,81,105,126,118,114,156,169).
5. Ikonomou *et al.* (2002).
6. Norstrom (1997); ΣPCDD = sum of 2,3,7,8-TCDD, 1,2,3,7,8-PnCDD.

Units	Concentration			TEQ			∑TEQ		Reference
	∑PCDD	∑PCDF	∑nPCB	∑PCDD	∑PCDF	∑nPCB	ng/g lw	ng/g ww	
pg/g ww	45.3	22.8	-	-	-	-	-	-	1
pg/g ww	-	-	-	-	-	-	-	-	1
pg/g ww	0.05	0.9	-	-	-	-	-	-	1
pg/g ww	nd	0.6	-	-	-	-	-	-	1
pg/g ww	0.15	1.0	-	-	-	-	-	-	1
	nd	0.7	-	-	-	-	-	-	1
pg/g ww	1.05	2.7	-	-	-	-	-	-	1
	nd	5.7	-	-	-	-	-	-	1
ng/g lw	0.66	0.65	20.6	0.38	0.49	1.51	2.39	0.214	2
ng/g lw	0.20	0.65	7.44	0.12	0.50	0.50	1.12	0.047	2
ng/g lw	0.19	0.88	10.86	0.13	0.67	0.73	1.53	0.078	2
ng/g lw	8.52	27.4	5.38	2.71	23.9	0.39	27.0	0.688	2
ng/g lw	2.46	7.22	24.1	0.63	6.21	1.36	8.19	0.357	2
ng/g lw	0.24	0.65	5.47	0.12	0.45	0.37	0.94	0.083	2
ng/g lw	0.03	0.05	32.1	0.02	0.04	1.87	1.92	0.166	2
ng/g lw	0.17	0.43	5.07	0.09	0.29	0.34	0.72	0.024	2
ng/g lw	0.07	0.21	3.30	0.04	0.12	0.22	0.38	0.026	2
pg/g ww	8.30	4.50	-	4.5		20.0	-	-	3
pg/g lw	-	-	31000 ± 11200	-	-	0.72 ± 0.20	-	-	4
pg/g lw	-	-	17800 ± 1590	-	-	0.59 ± 0.12	-	-	4
pg/g lw	-	-	-	-	-	8.6 ± 7.3	-	-	5
pg/g lw	-	-	185000 ± 116000	-	-	1.84 ± 0.66	-	-	4
pg/g lw	-	-	68300 ± 47100	-	-	1.49 ± 1.05	-	-	4
pg/g lw	13.6	-	-	-	-	-	-	-	6
pg/g lw	3.78	-	-	-	-	-	-	-	6
pg/g lw	15.8	-	-	-	-	-	-	-	6
pg/g lw	3.40	-	-	-	-	-	-	-	6
pg/g lw	4.00	-	-	-	-	-	-	-	6
pg/g lw	2.90	-	-	-	-	-	-	-	6
pg/g lw	2.20	-	-	-	-	-	-	-	6
pg/g lw	1.20	-	-	-	-	-	-	-	6