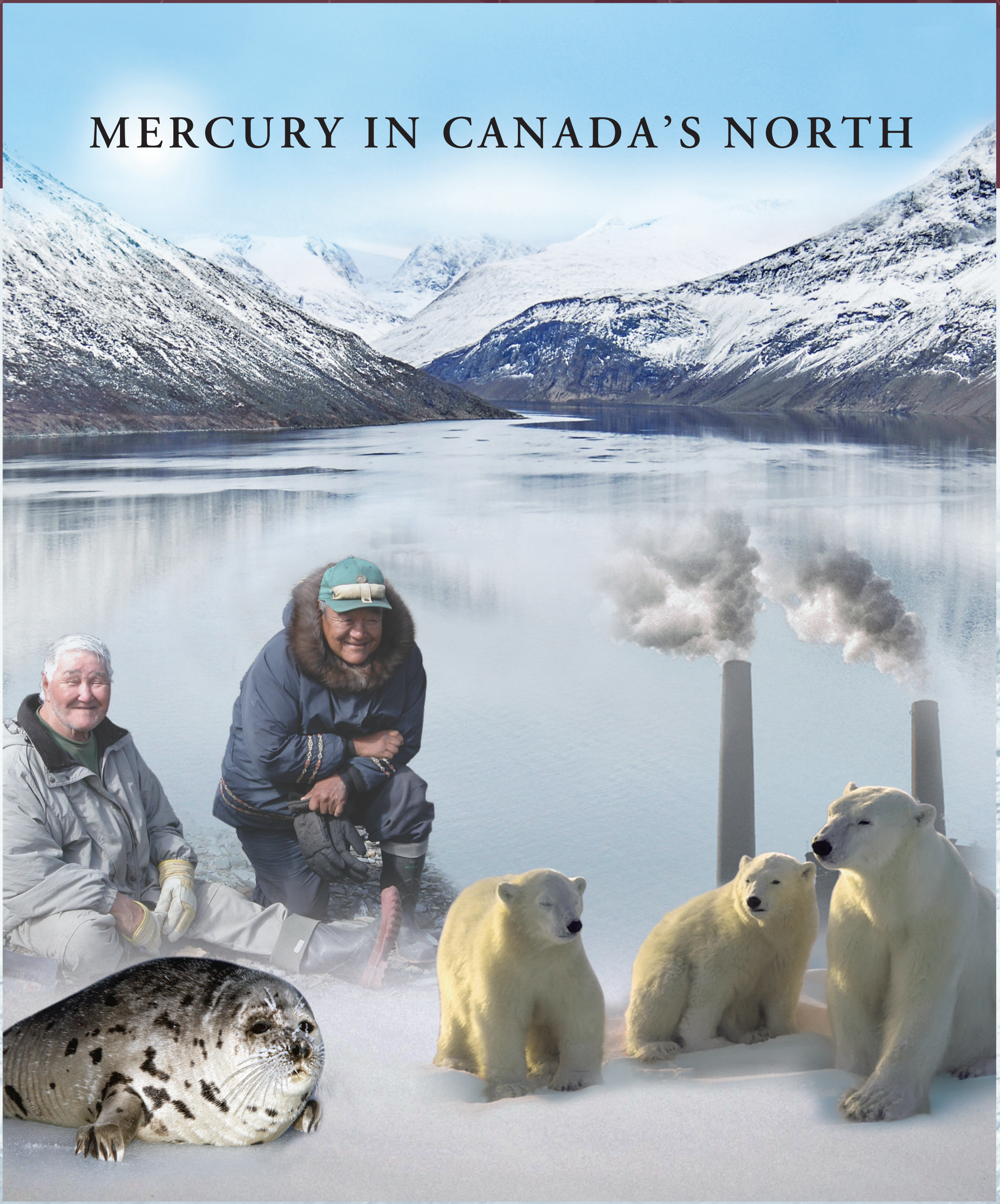
NORTHERN CONTAMINANTS PROGRAM

CANADIAN ARCTIC CONTAMINANTS ASSESSMENT REPORT III
2012

MERCURY IN CANADA'S NORTH



Aboriginal Affairs and
Northern Development Canada

Affaires autochtones et
Développement du Nord Canada

Canada


Chukchi Sea
Arctic Ocean
Beaufort Sea
Sachs Harbour (Ikaahuk)
Old Crow
Tuktoyaktuk
Aklavik (Ak∏arvik)
Inuvik (Inuuvik)
Fort McPherson (Teet∏'it Zheh)
Tsiigehtchic (Tsıigehtshık)
Holman (Uluqsaqtuuq)
Paulatuk (Paulatuuq)
Colville Lake (K'áhbamį́túé)
Fort Good Hope (Rádeyı̨lı̨kǫ́é)
Dawson
Keno Hill
Beaver Creek
Mayo
Pelly Crossing
Norman Wells (T∏egǫ́htı̨)
Kugluktuk (Coppermine)
Iqaluktuutiaq (Cambridge Bay)
Umingmaktuuq
Faro
Ross River
Carmacks
Déline (Délı̨ne)
Tulita (Tulít'a)
Qinguaq (Bathurst Inlet)
Haines Junction
Whitehorse
Wrigley (Pedzéh Kį́)
Rae Lakes (Gametì)
Snare Lakes (Wekweètì)
Carcross
Teslin
Wha Ti (Wha Tı)
Rae-Edzo (Behchokǫ̀-Edzo)
Watson Lake
Fort Simpson (¸íídlı¸Kų́ę́)
Jean Marie River (Tthek'éhdélı̨)
Yellowknife (Sǫ̀mbak'è)
Nahanni Butte (Tthenáágó)
Fort Providence (Zhahtı Kų́ę́)
Lutselk'e (¸útsëlk'é)
Fort Liard
Hay River (Xát∏'odehchee)
Fort Resolution (Denínu Kų́ę́)
Fort Smith (Tthebacha)


Alert
Ausuittuq
(Grise Fiord)
Resolute
(Qausuittuq)
Ikpiarjuk/Tununirusiq
(Arctic Bay)
Mittimatalik
(Pond Inlet)
Kangiqtugaapik
(Clyde River)
Baffin Bay
Qikiqtarjuaq
(Broughton Island)
Taloyoak
Igloolik
Sanirajak
(Hall Beach)
Uqsuqtuuq
(Gjoa Haven)
Kugaaruk
(Pelly Bay)
Pangnirtung
Naujaat
(Repulse Bay)
Iqaluit
Kinngait
(Cape Dorset)
Qamanittuaq (Baker Lake)
Salliq
(Coral Harbour)
Kimmirut
(Lake Harbour)
Igluligaarjuk (Chesterfield Inlet)
Ivujivik
Salluit
Kangiqsujuaq
Kangiqliniq (Rankin Inlet)
Tikirarjuaq (Whale Cove)
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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Executive Summary

Mercury is a naturally occurring element that is found globally in the environment. Mercury is released into the environment through natural processes and through human activities. Mercury transport and fate in the environment are complicated, and exposure to mercury poses significant health risks to both humans and biota. In order to effectively mitigate the risks posed by mercury exposure, the processes governing mercury transport, fate and effects must be understood.

Levels of mercury in the Canadian Arctic environment have increased several-fold since the advent of the Industrial Era. This rising trend in mercury levels continues in some biota and in some areas of the Arctic despite recognition of the issue and actions by many countries to mitigate mercury releases. Progress in reducing mercury levels in the environment is hindered by some key countries that are increasing the release of mercury through industrial emissions. These mercury emissions are then distributed around the globe, including to the Arctic, through long-range atmospheric transport.

For the last two decades, Canada's Northern Contaminants Program (NCP), which is administered by Aboriginal Affairs and Northern Development Canada, has supported research and monitoring of mercury transport, fate and effects in the Canadian Arctic. This program 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 Arctic Aboriginal people. The NCP focuses on: 1) monitoring of contaminant levels in Arctic biota that are most relevant to human contaminant exposure and is in keeping with Canada's monitoring obligations under international agreements; 2) research on contaminant cycling in the Arctic environment; and, 3) education and communication efforts to enable the provision of sound consumption advice. In the last several years, work supported by the NCP has been augmented by other research programs, most notably, ArcticNet and International Polar Year. These programs have supported several projects related to mercury transport and fate in Arctic ecosystems.

The purpose of the *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North* is to report on the scientific progress made under these programs and to evaluate our current understanding of the environmental fate of mercury in the Canadian Arctic. This report is part of a collection of companion reports on persistent organic pollutants (POPs) and human health as well as a highlights report which synthesizes all of the findings from these companion reports. This synthesis of scientific findings is intended to inform policy makers, managers, researchers, northerners and other relevant stakeholders. The present assessment also serves to take stock of progress made during the most recent phase of the NCP and to identify knowledge gaps and the most important scientific issues concerning mercury in Canada's northern environments. This report is the third NCP assessment of mercury in the Canadian Arctic in the last two decades and the first to focus exclusively on mercury.

Important scientific advances have been made in identifying the environmental fate of mercury and the processes that control its movement in the Canadian Arctic. The *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North* provides much new information on the sources and long-range transport of mercury to the Arctic, its cycling within marine, freshwater and terrestrial environments, and its bioaccumulation in and effects on the biota that live there. New information and data are also emerging on the complex ways in which climate change is affecting mercury cycling in the Arctic.

Mercury in the atmosphere

Research and monitoring of atmospheric mercury has improved our understanding of this key transport pathway to and within the Canadian Arctic. The chemical processes resulting in enhanced deposition of mercury during the spring-time phenomenon of atmospheric mercury depletion events (AMDEs) have been better characterized. More field measurements on the speciation of atmospheric mercury during AMDEs are now available, which indicate the oxidation of gaseous elemental mercury in the atmosphere is accompanied by large increases in reactive gaseous mercury and particulate mercury concentrations. The timing of AMDEs at the High Arctic monitoring station at Alert has changed over the last two decades, with the month of maximum AMDE activity shifting from May to April. The reason for the shift in timing of depletion events is not yet understood, but may be related to changes in air temperature. Air mercury concentrations declined between 2000 and 2009 at both Alert in the High Arctic and Kuujjuarapik in the sub-Arctic. The rate of decline at Kuujjuarapik was comparable to non-Arctic monitoring sites at lower latitudes while a slower rate of decline was observed at Alert. Model simulations suggest that long-range atmospheric transport from Asia likely contributes the most mercury (from anthropogenic, natural and re-emission sources) to the High Arctic and western Arctic, followed by contributions from North America, Russia and Europe.

Considerable uncertainty remains in the quantification of atmospheric mercury contributions to Arctic ecosystems. Direct measurements of wet and dry deposition in the Canadian Arctic are extremely limited, in part due to technical challenges, and more research using a variety of methodological approaches is essential to resolve this knowledge gap, especially since any changes in emission controls would most immediately affect the atmospheric mercury flux. Better characterization of geographic variation in atmospheric mercury is also warranted, particularly over the Arctic Ocean, to determine the influence of unique marine conditions on deposition.

Biogeochemical cycling of mercury

Some of the most significant advances in Arctic mercury science have been made in the area of biogeochemical cycling. Large amounts of new data are now available for water, snow, ice and sediment, providing more information on the concentrations, fluxes and transformations of inorganic and methylmercury in these abiotic matrices. This information was virtually absent for the Canadian Arctic a decade ago.

Much research effort has focused on mercury cycling in the cryosphere, particularly snow-air exchanges, transformations in the snowpack, and mercury transport by snowmelt. Snow mercury concentrations indicate that much of the mercury deposited during AMDEs is quickly reduced and emitted to the atmosphere. The level of evasion from the snowpack is dependant on proximity to the ocean because halogen compounds from the marine environment promote the stabilization and accumulation of mercury in snowpack. Areal loads of mercury in snow were estimated for Hudson Bay and the Arctic Archipelago, which suggest that snowmelt likely constitutes a small flux of mercury to Arctic marine waters during spring. In contrast, snowmelt can be an important source of mercury to freshwater ecosystems in the High Arctic.

The first measurements of methylation rates in Arctic seawater indicate that the water column of the Arctic Ocean is an important site for the methylation of inorganic mercury to form methylmercury. Production of dimethylmercury also occurs in the water column, and the breakdown of dimethylmercury in water or in the air (after evasion) may be another source of methylmercury available for biological uptake. Arctic marine waters were also found to be a substantial source of gaseous mercury to the atmosphere during the ice-free season. Microbial organisms and photochemical processes are key drivers of mercury cycling in the Arctic.

In spite of this progress, information remains limited on the methylation, oxidation and reduction of inorganic mercury in marine, freshwater and terrestrial environments in the Canadian Arctic. Further research is needed on mercury biogeochemical processes to develop a quantitative and mechanistic understanding of abiotic and microbially-driven mercury fluxes between air and various surfaces (soil, water, snow and vegetation), and to better understand the processes regulating methylmercury entry into Arctic food webs.

Monitoring of Arctic biota

Over the last two to four decades, mercury concentrations have increased in some marine and freshwater animals, specifically in several species of Arctic-breeding seabirds, some polar bear subpopulations, and some freshwater fish from the Mackenzie River Basin. Other monitored populations, including caribou, beluga, ringed seal, and freshwater fish from the Yukon and Nunavut showed no change or a slight decline in mercury concentrations. More frequent (annual) monitoring of key Arctic biota, which was initiated by the NCP in 2005, has resulted in more powerful datasets and a better ability to detect changes in biotic mercury levels. The different mercury trends reported for Arctic biota indicate that the drivers of temporal change may be regional or habitat-specific.

Marine biota exhibit considerable geographic variation in mercury concentrations across the Canadian Arctic. The Beaufort Sea remains an area of higher mercury contamination for beluga, ringed seals and polar bears compared to other Arctic regions. Habitat use and food web structure factor into mercury levels in Beaufort Sea beluga, which vary within the population according to size and sex segregation. Elevated concentrations in polar bears from the Beaufort Sea relative to Hudson Bay are related to the higher trophic level of bears in the Beaufort Sea area and higher water concentrations of methylmercury available for entry into the food web. Latitudinal trends have also been observed for ringed seals and seabirds, with greater concentrations in the High Arctic than at sub-Arctic sites.

While strong evidence exists for variable patterns of mercury bioaccumulation both regionally and temporally within the Arctic, the drivers of the observed trends in fish and wildlife remain unclear. Further research is required to identify the underlying processes leading to temporal changes in biotic mercury concentrations at sites where increases have been observed. Multiple factors may be implicated and need to be assessed, including the roles of mercury delivery processes (e.g., atmospheric deposition, mercury methylation), shifts in food web structure, and climate change. Geographic patterns of mercury bioaccumulation in marine biota deserve further investigation because certain factors may increase the vulnerability of biota to mercury exposure in some Arctic regions.

Climate change

The Arctic environment is undergoing profound change and at a rapid rate. Emerging evidence indicates that this environmental change is altering the cycling and bioaccumulation of mercury through effects on mercury fluxes and food web structure. Recent increases in algal primary production and catchment inputs may be enhancing mercury fluxes to Arctic lakes. Climate warming may be mobilizing mercury and leading to increased exposure of fish in the Mackenzie River, where a nearly two-fold increase in mercury concentration has occurred in burbot since 1985. Climate change also appears to be affecting the structure of food webs in certain regions resulting in dietary shifts and changes in mercury bioaccumulation for some marine biota such as thick-billed murres and ringed seals. However, large uncertainties remain in this emerging and complex science. A myriad of interconnected environmental changes are occurring in the Arctic, and detailed, process-focused investigations are needed to more precisely identify how these changes will alter the fate of mercury. Many aspects of the mercury cycle may be impacted including how mercury is delivered to Arctic ecosystems, its biogeochemical transformations and its trophic transfer in food webs.

Biological effects

Recent investigations on effects of methylmercury exposure specifically for Arctic wildlife have focused on adverse reproductive effects in seabirds, neurotoxicity in marine mammals, immunotoxicity in beluga, and the role of selenium in mercury detoxification. A laboratory egg injection study revealed that developing embryos of seabirds show species-specific sensitivity to methylmercury exposure at environmentally-relevant levels. On-going research suggests that methylmercury exposure at current environmental levels may affect brain neurochemical receptors in some Arctic top predators, although further verification is needed. Selenium is known to play an important role in sequestering and detoxifying mercury, and the formation of relatively inert mercury-selenium compounds may reduce the risk of mercury toxicity. The amount of selenium present relative to mercury in tissues of polar bears and beluga whales is variable, which suggests the protective effect of selenium against methylmercury toxicity may differ among species. Preliminary research on effects of mercury exposure on gene expression and immunotoxicity holds promise for providing new tools to better understand the toxicological effects of mercury in Arctic beluga. In general, Arctic biota have mercury concentrations that are below threshold levels of potential concern (derived from laboratory and field studies for non-Arctic species), although concentrations above threshold levels have been observed for some freshwater fish populations, Greenland shark, and some seabird species.

There is currently insufficient information to adequately assess effects of mercury exposure specifically for Arctic wildlife species, and further research should determine if and to what extent mercury exposure, combined with other stressors such as climate change, is affecting populations of Arctic fish, seabirds and marine mammals that have elevated mercury levels.

Future directions

Over the last two decades, the NCP has played a pivotal role in supporting research and monitoring of mercury in the Canadian Arctic environment. Earlier phases of the NCP provided important new knowledge on the Arctic mercury cycle, which in turn, resulted in the development and refinement of program priorities related to this contaminant. This assessment documents the progress made in our understanding of the behaviour of mercury in the Arctic environment, and reaffirms the importance of continuing research and monitoring of mercury pollution in the Arctic.

While the advances in scientific research described in this report are significant, the complex nature of the mercury cycle continues to provide challenges in characterizing and quantifying the relationships between mercury sources, transport processes, and levels as well as effects of mercury exposure on biota. These gaps in our knowledge of how global anthropogenic emissions of mercury are delivered to and accumulate in Arctic environments have implications for policy development related to risk management of mercury. Of particular concern are large uncertainties in our understanding of the processes that are contributing to increasing mercury concentrations in some Arctic fish and wildlife.

Current levels of mercury affecting Arctic ecosystems are a legacy of anthropogenic emissions that began with the onset of the industrial era. With continued global economic development dependent on combustion of coal, it is predicted that mercury deposition to the Arctic will continue to rise. If, however, emissions are reduced through implementation of the United Nations Environment Programme (UNEP) global treaty on mercury, deposition to the Arctic could be reduced significantly, which in the future would have a positive impact on Arctic ecosystems and human health. In order to better support national and global actions, and manage current mercury related risks, this assessment makes several science recommendations to improve our understanding of mercury in the Arctic. The key science recommendations of the *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North* are:

- Continue research and monitoring of atmospheric mercury, with an enhanced focus on deposition measurements to facilitate quantification of atmospheric contributions of mercury to Arctic ecosystems.
- Continue temporal trend monitoring of mercury in Arctic biota, and identify the processes that are changing mercury concentrations in some species.
- Further characterize the key processes acting on mercury after atmospheric deposition and their effects on the fate of mercury in the Arctic environment.
- Better characterize the processes that link climate change with mercury transport, cycling and bioaccumulation.
- Increase efforts to determine the biological effects of methylmercury exposure on Arctic fish and wildlife.

Résumé

Le mercure est un élément d'origine naturelle que l'on retrouve partout dans l'environnement. Les rejets de mercure résultent de processus naturels et d'activités humaines. Le transport et le devenir du mercure dans l'environnement sont complexes, et l'exposition au mercure présente des risques significatifs à la fois pour la santé humaine et pour le biote. Pour atténuer ces risques de manière efficace, il est nécessaire de comprendre les processus qui régissent le transport, le devenir et les effets du mercure.

Les concentrations de mercure dans l'Arctique canadien sont plusieurs fois supérieures à ce qu'elles étaient avant l'avènement de l'industrialisation. Cette tendance à la hausse des concentrations se maintient chez certaines espèces et dans certaines régions de l'Arctique, malgré la reconnaissance du problème et les mesures adoptées par de nombreux pays pour réduire les rejets de mercure. Les progrès réalisés en vue de la réduction des concentrations de mercure dans l'environnement sont entravés par certains pays clés, dont les rejets de mercure dus aux émissions industrielles sont en hausse. Ces rejets se répandent subséquemment dans le monde, et notamment dans l'Arctique, en raison du transport atmosphérique à grande distance des polluants.

Depuis deux décennies, le Programme de lutte contre les contaminants dans le Nord (PLCN), administré par Affaires autochtones et Développement du Nord Canada, appuie la recherche et les activités de surveillance sur le transport, le devenir et les effets du mercure dans l'Arctique canadien. Ce programme a été créé en 1991 en réaction aux préoccupations concernant l'exposition des humains à des concentrations élevées de contaminants présents dans le poisson et les espèces sauvages qui constituent une partie importante du régime alimentaire traditionnel des Autochtones de l'Arctique. Le PLCN est axé sur : 1) la surveillance des concentrations de contaminants chez les espèces sauvages de l'Arctique auxquelles les humains sont le plus exposés, compte tenu des obligations de surveillance contractées par le Canada dans le cadre d'accords internationaux; 2) la recherche sur le cycle des contaminants dans l'environnement arctique et 3) les activités d'éducation et de communication permettant d'offrir des avis de consommation appropriés. Au cours des dernières années, d'autres programmes de recherche, dont ArcticNet et l'Année polaire internationale, sont venus enrichir le travail réalisé dans le cadre du PLCN. Ces programmes ont permis la réalisation de plusieurs projets sur le transport et le devenir du mercure dans les écosystèmes de l'Arctique.

L'objet du rapport d'évaluation intitulé *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North* est de rendre compte des progrès scientifiques réalisés dans le cadre de ces programmes et d'évaluer notre compréhension actuelle du devenir du mercure dans l'Arctique canadien. Ce rapport fait partie d'une collection de rapports complémentaires sur les polluants organiques persistants (POP) et la santé humaine ainsi que d'un rapport des points saillants qui synthétise l'ensemble des résultats de ces rapports complémentaires. Cette synthèse de résultats scientifiques est destinée aux responsables des politiques, aux gestionnaires, aux chercheurs, aux résidents du Nord et aux autres parties intéressées. La présente évaluation vise également à dresser le bilan des progrès réalisés durant la plus récente phase du PLCN et à déterminer les lacunes en matière de connaissances ainsi que les enjeux scientifiques les plus importants liés au mercure présent dans le nord du Canada. Ce rapport constitue le troisième rapport d'évaluation publié par le PLCN au cours des deux dernières décennies afin de faire le point sur le mercure dans l'Arctique canadien, et est le premier rapport entièrement consacré au mercure.

D'importantes avancées ont été réalisées pour ce qui est de la détermination du devenir du mercure dans l'environnement et des processus qui régissent le déplacement de cette substance dans l'Arctique canadien. Le rapport intitulé *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North* fournit beaucoup de nouvelles données sur les sources et le transport à grande distance du mercure dans l'Arctique, le cycle du mercure dans les environnements marins, dulcicoles et terrestres, sa bioaccumulation dans le biote et les effets de cette bioaccumulation sur le biote. De nouvelles données viennent également éclairer les répercussions complexes du changement climatique sur le cycle du mercure dans l'Arctique.

Le mercure dans l'atmosphère

Les activités de recherche et de surveillance portant sur le mercure atmosphérique nous ont permis de mieux comprendre cette importante voie de transport vers l'Arctique canadien et à l'intérieur de cette région. Les processus chimiques favorisant le dépôt de mercure lors des épisodes d'appauvrissement en mercure de l'atmosphère (EAMA) qui se produisent au printemps ont été mieux caractérisés. Nous disposons maintenant de mesures plus nombreuses effectuées sur le terrain concernant la spéciation du mercure atmosphérique durant les EAMA. Ces mesures indiquent que l'oxydation du mercure élémentaire gazeux dans l'atmosphère s'accompagne d'importantes augmentations des concentrations de mercure gazeux réactif et de mercure particulaire. Le moment où surviennent les EAMA à la station de surveillance de l'Extrême-Arctique, à Alert, a changé au cours des deux dernières décennies, le mois d'activité maximale étant passé de mai à avril. La cause de ce changement n'est pas encore connue, mais pourrait être liée à des variations de la température de l'air. Les concentrations de mercure dans l'air ont diminué entre 2000 et 2009, tant à Alert, dans l'Extrême-Arctique, qu'à Kuujjuarapik, en région subarctique. Le taux de déclin à Kuujjuarapik était comparable aux taux enregistrés à des stations de surveillance situées à l'extérieur de l'Arctique, à des latitudes inférieures, alors qu'un taux de déclin moins élevé a été observé à Alert. Les simulations des modèles donnent à croire que le transport atmosphérique à grande distance depuis l'Asie représente probablement la contribution la plus importante au mercure (source anthropique ou naturelle et mercure réémis) présent dans l'Extrême-Arctique et la région ouest de l'Arctique, suivie de la contribution de l'Amérique du Nord, de la Russie et de l'Europe.

La quantification de la contribution du mercure atmosphérique dans les écosystèmes arctiques demeure en grande partie incertaine. Les mesures directes des dépôts de mercure humides et secs dans l'Arctique canadien sont extrêmement limitées, notamment à cause des difficultés techniques rencontrées. Il sera essentiel de mener d'autres travaux de recherche suivant différentes méthodes afin de combler cette lacune, surtout compte tenu du fait que tout changement touchant les mesures de lutte antiémissions aurait un effet immédiat sur les flux de mercure atmosphérique. Une meilleure caractérisation de la variation géographique du mercure atmosphérique est également nécessaire, plus particulièrement au-dessus de l'océan Arctique, pour déterminer l'incidence de conditions marines précises sur les dépôts.

Cycle biogéochimique du mercure

Certaines des avancées les plus significatives réalisées sur le mercure dans l'Arctique concernent le cycle biogéochimique du mercure. La grande quantité de données nouvelles obtenues sur l'eau, la neige, la glace et les sédiments fournit davantage d'information sur les concentrations, les flux et la transformation du mercure inorganique et du méthylmercure dans ces matrices abiotiques. Ce genre d'information sur l'Arctique canadien était pratiquement inexistant il y a dix ans.

Une grande partie des travaux de recherche a porté sur le cycle du mercure dans la cryosphère, et plus particulièrement sur les échanges air-neige, les transformations qui se produisent dans le stock nival et le transport du mercure résultant de la fonte des neiges. Les concentrations de mercure mesurées dans la neige indiquent qu'une grande partie du mercure déposé durant les EAMA est rapidement réduite et émise dans l'atmosphère. Le taux d'émissions passives provenant du stock nival dépend de la proximité de l'océan, car les composés halogénés du milieu marin favorisent la stabilisation et l'accumulation du mercure dans le stock nival. Selon les charges de mercure dans la neige qui ont été estimées pour la baie d'Hudson et l'archipel Arctique, la fonte des neiges serait à l'origine d'un petit flux de mercure vers les eaux marines de l'Arctique au printemps. Par ailleurs, la fonte des neiges peut constituer une source importante de mercure pour les écosystèmes d'eau douce dans l'Extrême-Arctique.

Selon les premières mesures des taux de méthylation effectuées dans les eaux marines de l'Arctique, la colonne d'eau de l'océan Arctique est un site important de méthylation du mercure inorganique, qui se transforme alors en méthylmercure. Du diméthylmercure est également produit dans la colonne d'eau, et la décomposition du diméthylmercure dans l'eau ou dans l'air (consécutive aux émissions passives) pourrait constituer une autre source de méthylmercure disponible pour l'absorption biologique. On a également constaté que les eaux marines de l'Arctique représentaient une source considérable de mercure gazeux pour l'atmosphère durant la saison libre de glace. Les microorganismes et les processus photochimiques jouent un rôle clé dans le cycle du mercure dans l'Arctique.

Malgré ces avancées, l'information demeure limitée en ce qui concerne la méthylation, l'oxydation et la réduction du mercure inorganique dans les environnements marins, dulcicoles et terrestres de l'Arctique canadien. D'autres recherches doivent être menées sur les processus biogéochimiques liés au mercure pour mieux comprendre, d'un point de vue quantitatif et mécaniste, les flux de mercure entre l'air et différentes surfaces (sol, eau, neige et végétation) qui reposent sur des facteurs abiotiques et biotiques, et pour approfondir notre compréhension des processus qui régissent l'introduction du méthylmercure dans les réseaux trophiques de l'Arctique.

Surveillance du biote de l'Arctique

Au cours des deux à quatre dernières décennies, les concentrations de mercure ont augmenté chez certains animaux des milieux marins et dulcicoles, notamment chez plusieurs espèces d'oiseaux marins nichant dans l'Arctique, certaines sous-populations d'ours blanc et certains poissons d'eau douce du bassin du fleuve Mackenzie. Les concentrations de mercure mesurées chez d'autres espèces étudiées, y compris le caribou, le béluga, le phoque annelé et des poissons d'eau douce du Yukon et du Nunavut, sont demeurées stables ou ont légèrement diminué. La surveillance plus fréquente (annuelle) de certaines espèces clés de l'Arctique, entreprise en 2005 dans le cadre du PLCN, a permis d'améliorer les ensembles de données et la capacité de détection des variations des concentrations de mercure dans le biote. Selon les différentes tendances observées au sujet des concentrations de mercure dans le biote arctique, les facteurs à l'origine des variations temporelles pourraient être liés à la région ou à l'habitat.

Les concentrations de mercure dans le biote marin présentent une variabilité géographique considérable à l'échelle de l'Arctique canadien. Dans la mer de Beaufort, on observe un taux de contamination par le mercure plus élevé chez le béluga, le phoque annelé et l'ours blanc que dans d'autres régions de l'Arctique. L'utilisation de l'habitat et la structure des réseaux trophiques ont une incidence sur les concentrations de mercure chez les bélugas de la mer de Beaufort; ces concentrations varient au sein de la population en fonction de la taille et du sexe des individus. Les concentrations élevées mesurées chez les ours blancs de la mer de Beaufort comparativement à ceux de la baie d'Hudson sont associées au niveau trophique supérieur des ours blancs de la mer de Beaufort et aux concentrations supérieures de méthylmercure dans l'eau pouvant s'introduire dans le réseau trophique. Des tendances ont également été observées en fonction de la latitude dans le cas des phoques annelés et des oiseaux marins, les concentrations étant plus élevées dans l'Extrême-Arctique que dans les régions subarctiques.

Bien que l'existence de profils variables de bioaccumulation du mercure dans l'Arctique ait été démontrée, tant sur le plan régional que temporel, les facteurs à l'origine des tendances observées chez certaines espèces de poisson et chez d'autres espèces sauvages demeurent incertains. Il faudra effectuer d'autres recherches pour déterminer les processus sous-jacents qui causent les variations temporelles des concentrations de mercure dans le biote aux sites où des augmentations ont été observées. De multiples facteurs pourraient être en cause et doivent être évalués, notamment le rôle des processus régissant l'apport de mercure (p. ex., dépôt atmosphérique, méthylation du mercure), l'évolution de la structure des réseaux trophiques et le changement climatique. Les profils géographiques de bioaccumulation du mercure dans le biote marin méritent qu'on s'y attarde, car certains facteurs peuvent accroître la vulnérabilité du biote à l'exposition au mercure dans certaines régions de l'Arctique.

Changement climatique

De profonds changements se produisent à un rythme rapide dans l'environnement arctique. Les nouvelles données indiquent que ces changements, qui se répercutent sur les flux de mercure et la structure des réseaux trophiques, ont une incidence sur le cycle et la bioaccumulation du mercure. La récente augmentation de la production primaire d'algues et des apports des bassins hydrographiques pourrait favoriser les flux de mercure vers les lacs de l'Arctique. Il est possible que le réchauffement climatique cause la mobilisation du mercure, ce qui entraînerait une augmentation de l'exposition du poisson dans le fleuve Mackenzie. La concentration de mercure chez la lotte a presque doublé dans ce fleuve depuis 1985. Le changement climatique semble également avoir des répercussions sur la structure des réseaux trophiques dans certaines régions, ce qui cause des modifications touchant les régimes alimentaires et la bioaccumulation du mercure chez certaines espèces marines, comme le Guillemot de Brünnich et le phoque annelé. Cette science nouvelle et complexe comporte toutefois d'importantes incertitudes. Il se produit dans l'Arctique une myriade de changements environnementaux qui sont interreliés : des études détaillées, fondées sur des processus, sont nécessaires pour bien comprendre l'incidence de ces changements sur le devenir du mercure dans l'environnement. De nombreux aspects du cycle du mercure pourraient être touchés, y compris le mode d'apport du mercure dans les écosystèmes arctiques, sa transformation biogéochimique et son transfert dans les réseaux trophiques.

Effets sur le plan biologique

Les études récentes portant sur les effets de l'exposition au méthylmercure observés chez les espèces sauvages de l'Arctique se sont attardées aux effets nocifs sur la reproduction des oiseaux marins, à la neurotoxicité chez les mammifères marins, à l'immunotoxicité chez le béluga et au rôle du sélénium sur la détoxification du mercure. Une expérience d'injection réalisée en laboratoire a révélé que les embryons d'oiseaux marins présentaient une sensibilité propre à l'espèce pour ce qui est de l'exposition au méthylmercure à des concentrations observées dans l'environnement. Les recherches en cours laissent entendre que l'exposition au méthylmercure selon les concentrations actuellement observées dans l'environnement pourrait affecter les récepteurs neurochimiques du cerveau chez certains prédateurs de niveau trophique supérieur de l'Arctique. D'autres vérifications doivent cependant être faites à ce sujet. On sait que le sélénium joue un rôle important dans la séquestration et la détoxification du mercure, et la formation de composés mercure-sélénium relativement inertes pourrait réduire le risque de toxicité associé au mercure. La quantité de sélénium présente par rapport à la quantité de mercure dans les tissus des ours blancs et des bélugas est variable, ce qui pourrait signifier que l'effet protecteur du sélénium contre la toxicité du méthylmercure diffère d'une espèce à l'autre. Les recherches préliminaires réalisées sur les effets de l'exposition au mercure sur l'expression des gènes et l'immunotoxicité promettent de fournir de nouveaux outils qui faciliteront la compréhension des effets toxiques du mercure sur le béluga dans l'Arctique. De façon générale, le biote de l'Arctique présente des concentrations de mercure inférieures aux seuils potentiellement préoccupants (établis d'après des études réalisées en laboratoire et sur le terrain sur des espèces présentes ailleurs que dans l'Arctique), bien que des concentrations supérieures à ces seuils aient été observées chez certaines populations de poissons d'eau douce, la laimargue atlantique et certaines espèces d'oiseaux marins.

Compte tenu de l'insuffisance des données actuelles, on ne peut évaluer adéquatement les effets de l'exposition au mercure sur les espèces sauvages de l'Arctique. La réalisation d'autres travaux de recherche devrait permettre de déterminer dans quelle mesure (le cas échéant) l'exposition au mercure, combinée à d'autres facteurs de stress comme le changement climatique, affecte les populations de poissons, d'oiseaux marins et de mammifères marins de l'Arctique qui présentent des concentrations élevées de cette substance.

Orientations futures

Au cours des deux dernières décennies, le PLCN a joué un rôle crucial dans le soutien des activités de recherche et de surveillance sur le mercure dans l'Arctique canadien. Les phases antérieures du PLCN ont favorisé l'acquisition de connaissances importantes sur le cycle du mercure dans l'Arctique; ces connaissances ont, à leur tour, permis d'élaborer et de raffiner les priorités du programme relativement à ce contaminant. Le présent rapport d'évaluation rend compte des progrès réalisés dans la compréhension du comportement du mercure dans l'environnement arctique, et confirme l'importance de poursuivre les activités de recherche et de surveillance sur la pollution par le mercure dans l'Arctique.

Bien que les progrès de la recherche scientifique mentionnés dans le présent rapport soient significatifs, la nature complexe du cycle du mercure rend difficiles la caractérisation et la quantification des relations entre les sources, les processus de transport et les concentrations de mercure ainsi que les effets de l'exposition au mercure sur le biote.

Ces lacunes dans notre compréhension du mode d'apport et d'accumulation des émissions anthropiques mondiales de mercure dans les milieux arctiques ont des répercussions sur l'établissement des politiques sur la gestion des risques liés au mercure. L'incertitude considérable entourant les processus qui contribuent à l'accroissement des concentrations de mercure chez certaines espèces de poisson et chez d'autres espèces sauvages de l'Arctique est particulièrement préoccupante.

Les niveaux actuels de mercure qui affectent les écosystèmes arctiques sont un héritage des émissions anthropiques qui ont commencé avec le début de l'ère industrielle. Avec la poursuite du développement économique mondial dépend de la combustion du charbon, il est prévu que le dépôt de mercure dans l'Arctique continuera d'augmenter. Si, toutefois, les émissions sont réduites grâce à la mise en œuvre du Programme des Nations Unies pour l'environnement (PNUE) traité mondial sur le mercure, le dépôt de l'Arctique pourrait être réduite de manière significative, ce qui à l'avenir pourrait avoir un impact positif sur les écosystèmes arctiques et la santé humaine. Afin de mieux soutenir les actions nationale et mondiale, et gérer les risques actuels de mercure liées, cette évaluation fait des recommandations scientifiques plusieurs mesures pour améliorer notre compréhension du mercure dans l'Arctique. Les recommandations clés de la science des contaminants dans l'*Arctique canadien d'évaluation Rapport III: Le mercure dans le Nord canadien* sont les suivants:

- Poursuivre la recherche et la surveillance sur le mercure atmosphérique, en mettant l'accent sur la mesure des dépôts afin de faciliter la quantification de la contribution du mercure atmosphérique dans les écosystèmes arctiques.
- Continuer de surveiller les tendances temporelles des concentrations de mercure dans le biote de l'Arctique et déterminer les processus qui entraînent une variation des concentrations de mercure chez certaines espèces.
- Continuer de caractériser les principaux processus agissant sur les dépôts atmosphériques de mercure ainsi que leurs effets sur le devenir du mercure dans l'environnement arctique.
- Améliorer la caractérisation des processus liant le changement climatique au transport, au cycle et à la bioaccumulation du mercure.
- Consacrer plus d'efforts à la détermination des effets biologiques de l'exposition au méthylmercure sur les poissons et les autres espèces sauvages de l'Arctique.

Acronyms and Abbreviations

AANDC
Aboriginal Affairs and Northern Development Canada

AMAP
Arctic Monitoring and Assessment Programme

AMDE
atmospheric mercury depletion event

ASGM
artisanal and small-scale gold mining

Br
bromine

Br_2
bromine molecule

BrO
bromine monoxide

C
Celsius

^{14}C
radiocarbon

$\delta^{13}C$
carbon stable isotope ratio

CACAR
Canadian Arctic Contaminants Assessment Report

ChE
acetylcholinesterase

Cl
chlorine

Cl_2
molecular chlorine

ClO
chlorine monoxide

cm
centimetre

^{137}Cs
cesium-137 isotope

D2
dopamine

DEHM
Danish Eulerian Hemispheric Model

DGM
dissolved gaseous mercury

DOC
dissolved organic carbon

dw
dry weight

EXEC
Extended Emissions Control scenario

fM
femtomole

F_{MeHg}
flux rate of methylmercury

F_{THg}
flux rate of total mercury

g
gram

GABA
gamma-aminobutyric acid

GEM
gaseous elemental mercury

GEOS-Chem
a global 3-D atmospheric composition model driven by data from the Goddard Earth Observing System

GLEMOS
Global EMEP Multi-media Modelling System

GOC
Government of Canada

GRAHM
Global/Regional Atmospheric Heavy Metals Model

GSC
Geological Survey of Canada

GSH
glutathione (reduced)

GSSG
glutathione (oxidized)

ha
hectare

Hg
mercury

Hg(0)
elemental mercury

Hg(I)
monovalent inorganic mercury

Hg(II)
divalent inorganic mercury

$HgBr_2$
mercuric bromide

$HgCl_2$
mercuric chloride

$HgSO_3$
mercuric sulphite

$\delta^{xxx}Hg$
mercury stable isotope ratio

$\Delta^{xxx}Hg$
mass-independent fractionation of a specific mercury stable isotope

ΔHgF
anthropogenic mercury flux

ΔHgF_{adj}
anthropogenic mercury flux adjusted for post-industrial changes in sedimentation rate

HO_2
hydroperoxyl radical

HOCl
hypochlorous acid

ID_{50}
median inhibition concentration

INAC
Indian and Northern Affairs Canada

INC
Intergovernmental Negotiation Committee

IPCC
Intergovernmental Panel on Climate Change

IPY
International Polar Year

kg
kilogram

km
kilometre

k_{pd}
rate constant of photodemethylation

L
litre

LC_{50}
median lethal concentration

LOAEL
lowest observable adverse effects level

LOI
loss on ignition

LRTAP
Long-range Transboundary Air Pollution

m
metre

M
mole

mACh
muscarinic (receptor)

MAO
monoaminoxidase

MDF
mass-dependent fractionation

MDN
Mercury Deposition Network

MeHg	monomethylmercury
ΔMeHg	net accumulation of methylmercury
MeHgCl	methylmercury chloride
Me_2Hg	dimethylmercury
merA	mercuric reductase enzyme
merB	organomercurial lyase (enzyme)
MFTR	*Maximum Feasible Technological Reduction* scenario
mg	milligram
MIF	mass-independent fractionation
mm	millimetre
MSCE-Hg	EMEP Meteorological Synthesizing Centre East model for mercury
MT	metallothionein
$\delta^{15}N$	nitrogen stable isotope ratio
N	north
NCP	Northern Contaminants Program
NEEM	Northern Greenland Eemian deep ice-coring project
ng	nanogram
NGR	National Geochemical Reconnaissance database
NMDA	N-methyl-D-aspartic acid
O_3	ozone
OCl^-	hyperchlorite ion
ODE	ozone depletion event
OH	hydroxyl radical
%	per cent
‰	per mil
PAR	photosynthetically active radiation
^{210}Pb	lead-210 isotope
PHg	particulate mercury
POC	particulate organic carbon
ppb	parts per billion
QA/QC	quality assurance and quality control
Redox	reduction-oxidation
RCC	Regional Contaminants Committee
RGM	reactive gaseous mercury
RHg	reactive inorganic mercury
S2	kerogen-derived carbon from algae
Se	selenium

SQ
Status quo scenario

t
tonne

THg
total mercury

TMF
trophic magnification factor

UNECE LRTAP
United Nations Economic Commission for Europe's *Convention on Long-range Transboundary Air Pollution*

UNEP
United Nation's Environment Programme

UV
ultraviolet

W
west

ww
wet weight

y
year

λ
net snow accumulation rate

µg
microgram

µM
micromole

µm
micrometre

Species Glossary

Common name	Latin name
American kestrel	*Falco sparverius*
Arctic char	*Salvelinus alpinus*
Arctic cisco	*Coregonus autumnalis*
Arctic cod	*Boreogadus saida*
Arctic fox	*Alopex lagopus*
Arctic hare	*Lepus arctica*
Arctic skate	*Amblyraja hyperborea*
Arctic tern	*Sterna paradisaea*
bearded seal	*Erignathus barbatus*
beluga whale	*Delphinapterus leucas*
black guillemot	*Cepphus grylle*
black-legged kittiwake	*Rissa tridactyla*
burbot	*Lota lota*
Canada goose	*Branta canadensis*
capelin	*Mallotus villosus*
caribou	*Rangifer tarandus*
caribou (barren-ground)	*Rangifer tarandus groenlandicus*
caribou (woodland)	*Rangifer tarandus tarandus*
Caspian tern	*Sterna caspia*
clam	*Mya truncata*
common eider	*Somateria mollissima*
common loon	*Gavia immer*
common tern	*Sterna hirundo*
crinkled snow lichen	*Flavocetraria nivalis*
curled snow lichen	*Flavocetraria cucullata*
Dolly Varden char	*Salvelinus malma malma*
double-crested cormorant	*Phalacrocorax auritus*
ermine	*Mustela erminea*
fourhorn sculpin	*Myoxocephlaus quadricornis*
flounder	*Platichthys stellatus, Pleuronectes glacialis*
glaucous gull	*Larus hyperboreus*
Greenland shark	*Somniosus microcephalus*
harbour seal	*Phoca vitulina*

Common name	Latin name
harp seal	*Phoca groenlandica*
herring gull	*Larus argentatus*
hooded merganser	*Lophodytes cucullatus*
ivory gull	*Pagophilus eburnea*
lake trout	*Salvelinus namayush*
lake whitefish	*Coregonus clupeaformis*
laughing gull	*Larus atricilla*
least cisco	*Coregonus sardinella*
long-tailed duck	*Clangula hyemalis*
mink	*Mustela vison*
moose	*Alces alces*
muskrat	*Ondatra zibethicus*
narwhal	*Monodon monoceros*
ninespine stickleback	*Pungitius pungitius*
northern fulmar	*Fulmarus glacialis*
northern pike	*Exos lucius*
osprey	*Pandion haliaetus*
Pacific herring	*Clupea palasii*
Pacific salmon	*Onchorynchus sp.*
polar bear	*Ursus maritimus*
polar cod	*Arctogadus glacialis*
rainbow smelt	*Osmerus mordax*
red-backed vole	*Clethrionomys rutilus*
ringed seal	*Phoca hispida*
round whitefish	*Prosopium cylindraceum*
royal tern	*Sterna maxima*
saffron cod	*Eleginus gracilis*
sandhill crane	*Grus canadensis*
sandlance	*Ammodytes sp.*
short-horn sculpin	*Myoxocephalus scorpius*
snow bunting	*Plectrophenax nivalis*
snowy egret	*Egretta thula*
thick-billed murre	*Uria lomvia*
tri-colored heron	*Egretta tricolor*
walrus	*Odobenus rosmarus rosmarus*
willow	*Salix sp.*
willow ptarmigan	*Lagopus lagopus*
wolf	*Canis lupus*

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Paul Vecsei

CHAPTER 1

Introduction

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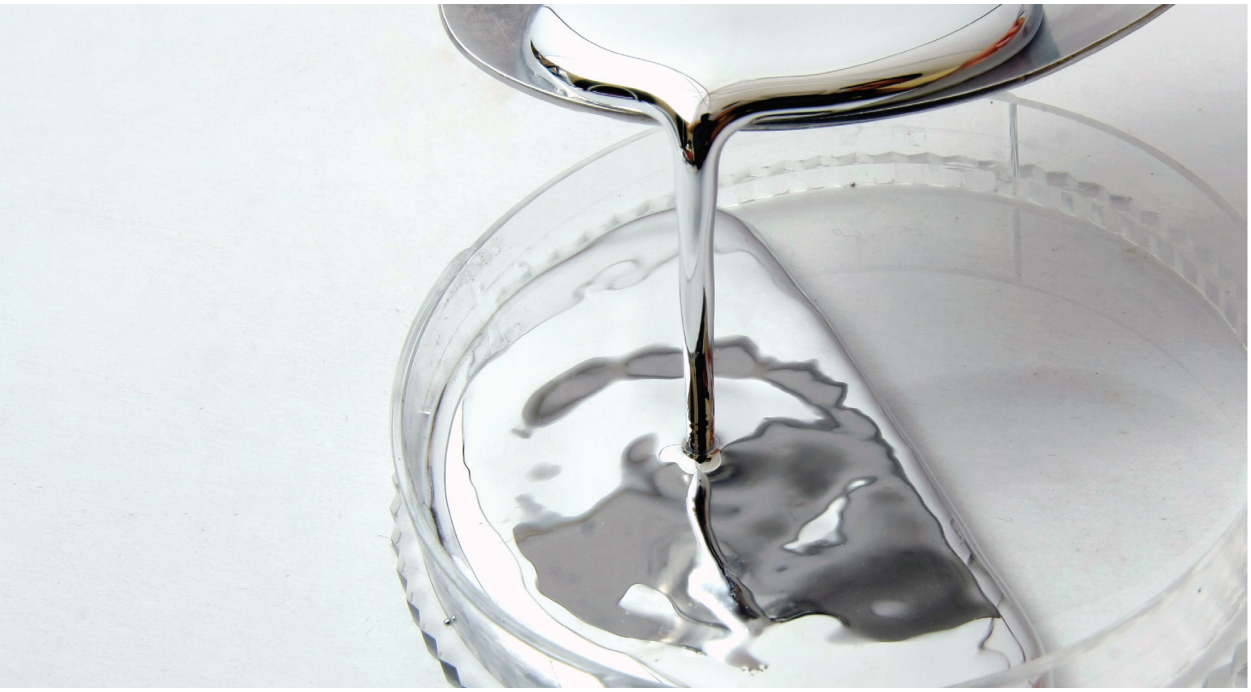
1.1 Introduction

Environmental levels of mercury (Hg) in the Canadian Arctic have increased several-fold since the advent of the Industrial Era. This trend in Hg levels continues in some biota and areas of the Arctic despite recognition of the issue and actions by many countries to mitigate Hg releases (Rigét et al. 2011). Mercury transport and its fate in the environment are complicated, and exposure to Hg poses significant health risks to humans and biota. In order to effectively mitigate the risks posed by Hg exposure, the processes governing Hg transport, its fate, and its effects must be understood.

For the last two decades, the Northern Contaminants Program (NCP) of Aboriginal Affairs and Northern Development Canada (previously Indian and Northern Affairs Canada) has supported research and monitoring of Hg transport, its fate, and its effects in the Canadian Arctic. This program was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in fish and wildlife species important to the traditional diets of Arctic Aboriginal people. The NCP focuses on: 1) monitoring of contaminant levels in Arctic biota that are most relevant to human contaminant exposure and is in keeping with Canada's monitoring obligations under international agreements; 2) research on contaminant cycling in the Arctic environment; and, 3) education and communication efforts to enable the provision of sound consumption advice. In the last several years, work supported by the NCP has been augmented by other research programs, most notably, ArcticNet and International Polar Year (IPY). These programs have supported several projects related to Hg transport and fate in Arctic ecosystems.

The purpose of the *Canadian Arctic Contaminants Assessment Report III: Mercury in Canada's North (CACAR III Mercury)* is to report on the scientific progress made under these programs and to evaluate our current understanding of the environmental fate of Hg in the Canadian Arctic. This assessment will



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inform policy and regulatory development and lay the foundation for building future scientific programs. The *CACAR III Mercury* is the third NCP assessment of Hg in the Canadian Arctic in the last two decades and the first to focus exclusively on Hg.

1.2 Why is Hg a concern in the Arctic?

Although Hg is a natural element, human activities—particularly the widespread combustion of coal—have released large quantities of Hg into the atmosphere and increased levels of Hg deposition around the globe. Air currents transport Hg over long distances and thereby connect emission sources at lower latitudes to the Arctic. The increase in anthropogenic Hg emissions due to industrialization over the last century and the transport of these emissions northward via air currents have resulted in levels of Hg in the Arctic environment that are significantly above historical levels (Fitzgerald et al. 2005, Muir et al. 2009, Outridge et al. 2009, Hare et al. 2010). Once Hg is deposited onto terrestrial and aquatic ecosystems it can undergo chemical reactions that convert it into methylmercury (MeHg). This form of Hg can accumulate to high levels in top predators which, in turn, can result in adverse health risks for wildlife and the humans that consume them.

Arctic Aboriginal people have a unique connection to the biota that share their environment. Activities associated with country food, including hunting, fishing, consumption, and community sharing, contribute to the cultural, social, and economic well-being of northern communities (NCP 2009). Country food in northern communities consists of fish and marine mammals such as seals, walruses, whales, and polar bears, as well as terrestrial animals such as caribou, ptarmigan, geese, eggs, and a selection of berries. High levels of Hg are found in some of these foods in certain areas of the Arctic. As a result, there are places in the Canadian Arctic where some of the human population is at risk because the dietary intake of Hg is greater than the level known to be safe (NCP 2009). Because of the sensitivity of the developing child to Hg, the unborn babies of pregnant women and young children are especially vulnerable to adverse health effects from exposure. For example, a recent study showed that nearly 60% of pre-school children in Nunavut exceeded the provisional tolerable weekly intake level for MeHg set by the World Health Organization (Tian et al. 2011). The main sources of Hg identified in that study were country foods, primarily whale muktuk, ringed seal liver, fish, and caribou meat.

Eric Loring

Although reports of recent declines in Hg exposure for Aboriginal people in the Canadian Arctic (NCP 2009) are encouraging, a reduction in country food consumption, which is the probable cause for this decline, is not a positive development. After balancing the risks and benefits, local health officials generally still promote the consumption of country foods low in Hg because they are rich in vitamins, nutrients, and fatty acids, and may protect against harmful effects of contaminants (NCP 2009). However, country foods low in Hg, such as Arctic char and caribou, may not always be accessible in particular areas or at certain times of the year.

The avoidance or reduction in country food consumption due to fear of health effects from Hg and other contaminants such as persistent organic pollutants can exacerbate the negative outcomes of a nutrition transition occurring among Aboriginal peoples and add to food insecurity experienced already in northern communities (Duhaime and Bernard 2008, Egeland et al. 2010, Tian et al. 2011). These concerns, in addition to climate change—which may impact the accessibility of traditional foods—combine to intensify the complex and cumulative challenges that Arctic Aboriginal people are experiencing. Hence, efforts to reduce Hg levels in all Arctic biota will not only reduce human health risks but also enhance food security by increasing the diversity of fish and wildlife that are safe to eat.

Elucidating the relationship between anthropogenic releases of Hg, levels of Hg in biota, and the effects of Hg exposure on humans and biota has challenged scientists because of the complicated chemistry of Hg in the environment in general, and in the Arctic in particular. Although these relationships are complex, studies assessing historical to present-day levels of Hg in animal hair, feather, and teeth show that most of the Hg in Arctic biota today is of anthropogenic origin (Dietz et al. 2009). Hence, actions to reduce anthropogenic releases of Hg are required to protect the Arctic and its people.

1.3 Risk management of Hg

In recognition of the contribution of anthropogenic emissions to Hg burdens in biota and humans, many governments are taking actions to reduce Hg releases.

Mercury is a heavy metal that has been identified as a toxic substance under the *Canadian Environmental Protection Act, 1999*. In October 2010, the Government of Canada (GOC) released the *Risk Management Strategy for Mercury*, which describes Canada's progress to date in managing the risks associated with Hg; ongoing and planned actions to further reduce Hg releases; and, monitoring programs to address ongoing risks associated with Hg (GOC 2010). The objective of the GOC's risk management strategy is "to protect the health of Canadians and their environment from the risks associated with mercury, by minimizing, and where feasible, eliminating anthropogenic mercury releases" (GOC 2010). Toward this objective, the GOC has implemented a wide range of regulatory and non-regulatory initiatives in collaboration with provincial and territorial governments, industry, and other stakeholders. As a result of these actions, domestic Hg emissions have been reduced by approximately 90% since the 1970s (GOC 2010).

In areas most affected by domestic emissions, measurable decreases in environmental Hg levels have been observed and attributed to emission reductions (Temme et al. 2007, Sunderland et al. 2008). However, Hg can be transported long distances from the source of emission. Hence, Canada is just one of many contributors to the anthropogenic Hg burden that is deposited on the Canadian landscape. It has been estimated that foreign anthropogenic emissions contribute more than 96% of the anthropogenic Hg deposited on Canada (GOC 2010). Hence, global action is also needed to address the Hg pollution problem in Canada.

In recognition of the global nature of Hg pollution, Canada is actively engaged in a number of regional, bi-lateral, and international efforts to address this issue. These efforts include the Arctic Council, a high-level intergovernmental forum among Arctic States and communities that addresses issues such as sustainable development and environmental protection. Canada is a Party to the *Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal*, which aims to protect human health and the environment from the adverse effects resulting from the generation, management, storage, transboundary movement and disposal of hazardous wastes, including Hg. Canada is also a Party to the *Protocol on Heavy Metals* under the United Nations Economic Commission for Europe's *Convention on Long-range Transboundary Air Pollution* (UNECE LRTAP). This Protocol aims to reduce emissions of mercury, lead, and cadmium from industrial sources, combustion processes, and waste incineration. The Protocol also introduces measures to lower emissions of Hg from products such as batteries. In addition, Canada participates in the Global Mercury Programme and the Global Mercury Partnership under the United Nations Environment Programme (UNEP). The Partnership Program aims to initiate actions in a number of key sectors such as coal combustion, and artisanal and small-scale gold mining to reduce Hg releases.

In February 2009, the UNEP Governing Council agreed to launch negotiations on a global, legally-binding agreement that may include voluntary approaches to reduce Hg emissions. A total of five intergovernmental negotiation committee (INC) meetings were planned, starting with INC-1 June 2010 in Stockholm, Sweden and ending in 2013 with the ministerial signing in Japan. The current negotiations form an important backdrop to Canada's risk management strategy and are a venue to communicate to the world the adverse effects of global anthropogenic Hg releases on the Arctic and its people.

1.4 Mercury research in the Canadian Arctic during the last two decades

1.4.1 The NCP

During Phase 1 of the NCP from 1991 to 1996, intensive research and monitoring focused on obtaining baseline information on Hg levels in the Canadian North, identifying transport pathways, and determining

Hg fate. It was clear from the *Benchmark Report*—published as a special issue of a science journal—that the impacts of long-range atmospheric transport of Hg were cause for serious concern in the Arctic although there were major knowledge gaps (Barrie et al. 1992, Lockhart et al. 1992, Muir et al. 1992). Early investigations laid the foundation for many of the ongoing monitoring programs within the NCP including the first routine measurements of atmospheric Hg in the Canadian Arctic at Alert and Hg measurements in freshwater, terrestrial, and marine animals. Information generated during Phase I of the NCP was synthesized and published in the first *CACAR* in 1997 (NCP 1997).

The NCP began its second phase in 1998. Continuing until 2003, Phase II supported studies designed to answer questions about the impacts and risks to human health that may result from the levels of contamination in key Arctic food species. Research and monitoring of Hg in the Arctic environment led to several important advances, particularly for temporal trends and transport pathways and processes.

In the 1990s, Canadian scientists discovered atmospheric mercury depletion events (AMDEs) in the Arctic, a phenomenon wherein increased deposition of atmospheric Hg occurs following polar sunrise in spring (Schroeder et al. 1998). Enhanced Hg deposition through AMDEs was highlighted as a potentially important mechanism contributing to the high levels of Hg observed in some Arctic biota. However, at the end of Phase II, there was insufficient evidence to determine the significance of AMDEs to levels of Hg in Arctic ecosystems.

During Phase II, retrospective analyses were performed on archived samples of biological tissue to augment temporal trend datasets of Hg in Arctic biota. For most freshwater and marine datasets that spanned the last 20 to 30 years, no changes in Hg levels of biota were observed. However, in a few cases including seabird eggs in Lancaster Sound, beluga from the Beaufort Sea and western Hudson Bay, and Mackenzie River burbot, significant increases in Hg were found. Further investigation was recommended to understand those trends.

By the end of Phase II, it had become clear that the Arctic was experiencing major changes in climate and in the cryosphere (Macdonald et al. 2005). Although little information was available, these changes were anticipated to have significant consequences for Hg pathways. Climate change was highlighted as a potentially important driver of Hg cycling in the Arctic that required further research.

Findings on Hg from Phase II of the NCP and recommendations for future research were documented in the *CACAR II* in separate volumes on the physical environment (Bidleman et al. 2003) and the biological environment (Fisk et al. 2003). The findings and recommendations from these reports formed the basis of the *Blueprint for Environmental Monitoring and*

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Research; a document that outlined the research and monitoring priorities for Phase III of the NCP. An important focus of the *Blueprint* in Phase III was an intensive core program involving the annual temporal trend monitoring of contaminants, including Hg, in key indicator species at specified locations throughout the Arctic. The *Blueprint* identified strategies to reduce sources of natural variation in contaminant datasets and to increase the statistical power of time series. The ongoing core monitoring program of the NCP represents the most comprehensive program of its kind in the circumpolar Arctic.

1.4.2 Other Arctic research programs

ArcticNet is a Network of Centres of Excellence of Canada, supported by the Natural Sciences and Engineering Research Council, that partners with scientists, managers, and northern communities to study the impacts of climate change in the Canadian Arctic. The main objective of this research network is to develop and disseminate knowledge that contributes to strategies and policies that will enable Canadians to adapt to impacts and opportunities of climate change and globalization in the Arctic (ArcticNet 2010). Within the context of climate change, research was conducted on Hg in the Arctic Ocean and in food webs of Arctic lakes during both Phase 1 (2004 to 2008) and Phase 2 (2008 to 2011) of ArcticNet. Where available, outcomes of this research are described in this assessment.

In addition, the fourth IPY from 2007 to 2008 stimulated national and international cooperation on research in the Arctic. Several IPY projects funded by the GOC investigated Hg in the Canadian Arctic including the global atmospheric transport of Hg to the Arctic, processes of atmospheric deposition within the unique environment of the Arctic Ocean, and Hg accumulation in marine and freshwater food webs. These collaborative projects produced much new information on Hg cycling in the Arctic and, where available, contributed to the findings in this assessment.

1.5 Northern engagement in activities related to Hg in the Arctic

1.5.1 NCP research and monitoring

The NCP was developed in response to concerns about the potential environmental and human health risks associated with Arctic pollution. Arctic Aboriginal peoples continue to experience levels of contaminant exposure that are significantly higher than other Canadians and, in some cases, exceed Health Canada guidelines (NCP 2009). Northerners are the most important stakeholders in the work of the NCP and play a major role in defining the program's priorities through representation on the multi-agency Management Committee. Northern priorities related to contaminants are also defined at a regional level through Regional Contaminants Committees (RCCs) in each of the five major regions: Yukon, Northwest Territories, Nunavut, Nunavik, and Nunatsiavut (Fig. 1). Chaired by regional Aboriginal representatives, these RCCs are responsible for coordinating projects in their respective regions and ensuring that adequate consultation, communication, and community engagement are a core part of each project.

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Community experts, hunters, and elders participate to some extent in all NCP projects and, in many cases, play a major role in the field collection of tissues and data for the core monitoring program. Community

FIGURE 1.1

Geographic coverage of the NCP includes the Yukon, Northwest Territories, and Nunavut as well as the Inuit regions of Nunavik in Quebec and of Nunatsiavut in Newfoundland and Labrador.

members are increasingly playing a greater role in the execution of NCP projects, not only through sample collection, but also by contributing traditional knowledge and local observations to strengthen the interpretation of results. The development of scientific capacity in communities facilitates the communication of NCP findings and promotes science education among community youth. The crucial roles that communities play in NCP research and monitoring activities are highlighted in a series of text boxes throughout this report. A current and future priority for the NCP is enhancing community engagement and leadership in community-based monitoring.

1.5.2 International agreements

As demonstrated over a decade ago during the negotiation process for the *Stockholm Convention on Persistent Organic Pollutants* (Stockholm Convention), Canadian Aboriginal organizations, in partnership with the GOC, play a constructive and important role at UNEP forums and negotiations (Fenge 2003, Furgal et al. 2003). The success of this partnership is particularly visible by the explicit mention of the vulnerability of the Arctic and its people in the Preamble of the Stockholm Convention.

The NCP recognizes that support from, and partnership with, Aboriginal organizations can successfully contribute to international policy development on the global abatement of Hg by bringing attention to its impacts on Arctic Aboriginal people. For the UNEP Treaty on Mercury currently under negotiation, the NCP supports the participation of interested Canadian Aboriginal organizations and, at the invitation of the GOC, the Inuit Circumpolar Council has participated with the Canadian delegation in meetings that have taken place to date.

1.6 The CACAR III: Mercury in Canada's North

The objective of the *CACAR III Mercury* is to evaluate the current state of science on Hg in the Canadian Arctic environment. This report presents the latest information on sources and long-range transport of Hg to the Arctic, its movement and cycling within marine, freshwater, and terrestrial environments, and its bioaccumulation and effects on the biota that live there. This synthesis of scientific findings is intended to inform policy makers, managers, researchers, Northerners, and other relevant stakeholders of recent advances in our understanding of key processes that control the fate of Hg in the Arctic. The assessment also serves to take stock of progress made during Phase III of the NCP and to identify knowledge gaps and the most important scientific issues concerning Hg in Canada's northern environments. A separate *CACAR* on persistent organic pollutants in the Canadian Arctic parallels this assessment. The NCP also recently completed a human health assessment for exposure to environmental contaminants (including Hg), and its findings are presented in the *Canadian Arctic Contaminants and Health Assessment Report* (NCP 2009).

The *CACAR III Mercury* summarizes research and monitoring conducted under the NCP from 2002 to 2009, and it also includes relevant information from published and unpublished studies under other initiatives including the IPY and ArcticNet. As such, this assessment focuses on priority activities of the NCP, namely spatial monitoring of Hg levels in species that are used as traditional food, temporal trend monitoring of Hg levels in biota, monitoring of atmospheric Hg, and wildlife health. Since 2003, the *Environmental Monitoring and Research Blueprint* has increasingly emphasized the need for research on the pathways and processes that influence contaminant behaviour and fate in the Arctic, as well as potential effects on Arctic ecosystems and highly exposed species. Building on the findings of *CACAR II*, the influence of climate change on contaminant processes was highlighted as a priority research area under the *Phase III Blueprint*. Recent findings on levels and biogeochemical processing of Hg in the physical environment—water, snow, ice, and sediment—are therefore a focus of this assessment because of their key importance in controlling the fate of Hg in Arctic ecosystems.

The NCP maintains a Quality Assurance and Quality Control (QA/QC) Program to ensure the quality, reliability, and comparability of Hg measurements generated during research and monitoring activities. Projects funded by the NCP are required to participate in interlaboratory comparison studies on the analysis of test samples. Results of the QA/QC Program during Phase III of the NCP are provided in Annex I.

From the field collection of samples in remote areas to their analysis in research laboratories, the Arctic studies described in this assessment involved many participants from government departments, universities, northern communities, and Aboriginal organizations. This report, too, is a product of collaborative efforts from dozens of scientists who contributed their expertise and summarized Arctic Hg science from their research and other sources. Internationally, the NCP works very closely with the circumpolar Arctic Monitoring and Assessment Programme (AMAP), an expert working group of the Arctic Council that provides scientific information on contaminant issues in circumpolar countries. Preparation of the *CACAR III Mercury* involved collaboration with AMAP, which has concurrently produced a Hg assessment for the circumpolar Arctic (AMAP 2011). In addition, under Environment Canada's Clean Air Regulatory Agenda, a Canadian Mercury Science Assessment is scheduled for release in 2013 and will be the first comprehensive national description of Hg in the Canadian environment. Information from the *CACAR III Mercury* will contribute to the Arctic component of the national assessment.

The information in this report is organized by environment, for which data on relevant physical and biological components are presented together. **Chapter 2** of this assessment provides an overview of sources, processes, and pathways that affect the transport of Hg to Arctic environments. Background information is also provided on the complex and emerging science of how climate change may affect Hg cycling. **Chapter 3** examines Hg levels, speciation, and chemistry in the atmosphere and depositional fluxes to Arctic surfaces. **Chapters 4, 5, and 6** examine how Hg moves within terrestrial, freshwater, and marine environments with their unique physical, chemical, and ecological characteristics. **Chapter 7** presents recent information on biological effects of Hg in Arctic biota. **Chapter 8** provides an overall summary of the state of the science on Hg in the Canadian Arctic environment and identifies knowledge gaps and important directions for future research.

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CHAPTER 2

Sources, Processes and Pathways

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2.1 Introduction

Mercury (Hg) is a heavy metal that is ubiquitous in the environment. It has a complex cycle involving transformations into various chemical forms by biogeochemical processes, and it moves within and between ecosystems through multiple pathways. While Hg is an element that occurs naturally in the environment, human activities have altered its cycle and increased its levels in water, air, and soil (Mason and Sheu 2002, Sunderland and Mason 2007). In this chapter, an overview of Hg sources, processes, and pathways is presented as we currently understand them. Many aspects of the Arctic Hg cycle remain poorly resolved, and more detailed discussions of progress in our understanding of these processes are presented in subsequent chapters on specific environments.

2.2 Physical and chemical properties of Hg

2.2.1 Physical properties

Mercury is a unique element because, in its pure form, it is the only heavy metal that is liquid at room temperature and it has a high surface tension that results in the formation of liquid droplets. Elemental Hg [Hg(0)] is also highly volatile and accumulates as a gas in the atmosphere. The stability of gaseous Hg(0) allows it to be transported over long distances through the atmosphere and deposited onto landscape surfaces around the world, making it a truly global pollutant.

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2.2.2 Chemical speciation of Hg in the environment

Mercury occurs in the environment primarily as one of four chemical forms—referred to as species: Hg(0), divalent inorganic Hg [Hg(II)], and organic Hg in the form of monomethylmercury (MeHg) or dimethylmercury (Me_2Hg) (Box 2.1). Mercury is transformed from one species to another through a variety of biogeochemical processes including oxidation or reduction reactions, and methylation or demethylation reactions driven by abiotic and/or biological mechanisms (Box 2.1). These transformations alter the chemical properties of Hg and thereby affect its retention and movement through the atmosphere, and terrestrial, marine, and freshwater environments.

The pool of Hg in the atmosphere is predominantly as Hg(0), although gaseous Hg(II)—referred to as reactive gaseous mercury (RGM)—and particulate Hg(II) (PHg) may also be present with short residence times. In fresh waters, Hg(II) is the dominant species with MeHg and Hg(0) together often accounting for < 30% of the total Hg (THg) pool (Morel et al. 1998). In marine waters, Hg(II) is also often the dominant species although Hg(0) can represent a more considerable portion of the THg pool (Fitzgerald et al. 2007). Dimethylmercury can be found in low concentrations in marine waters and is thought to be absent from fresh waters (Morel et al. 1998). Methylmercury often accounts for only a small portion of all the Hg present in marine and fresh waters. However, it is more toxic than inorganic species and it biomagnifies in food webs, making it the dominant form that accumulates in top predator organisms.

Most Hg(II) and MeHg in water and sediment are associated with other ions or molecules (ligands) that may be inorganic (especially hydroxide, chloride, or sulphide) or organic (Morel et al. 1998). The dominant complexes that are formed depend on the relative concentrations of different ligands present and the chemical reduction potential and pH of water (Morel et al. 1998). The nature of these complexes affects the mobility and bioavailability of Hg in marine and fresh waters.

2.2.3 Measurements of Hg

A variety of analytical techniques exist to measure environmental concentrations of Hg, and many of these methods target an operationally defined component of the Hg pool rather than a particular species. The main analytical measurements of environmental Hg found in this assessment are summarized in Table 2.1. Total concentrations of Hg are commonly measured in water or tissue and typically include both inorganic and organic species. A specific phase of Hg within a matrix may be analyzed such as the particulate or dissolved fraction in water, or the particulate fraction in air.

BOX 2.1 Overview of key Hg species and biogeochemical transformations

Elemental Hg—Also referred to as metallic Hg, this form is a shiny silver-white metal that is liquid at room temperature. It is volatile and also accumulates as a gas in the atmosphere. Elemental Hg is abbreviated as Hg(0) in this report.

Divalent Inorganic Hg—The mercuric ion (Hg^{++}) is the most common oxidized state of Hg, with two electrons lost. It forms Hg compounds such as mercuric sulphide, also know as cinnabar (HgS). Divalent inorganic Hg is abbreviated as Hg(II) in this report.

Monomethylmercury—This organic form of Hg is composed of a methyl group (CH_3^-) bonded to a Hg atom and has the chemical formula of CH_3Hg^+. It is abbreviated as MeHg in this report.

Dimethylmercury—This organic form of Hg is composed of two methyl groups (CH_3^-) bonded to a Hg atom and has the chemical formula of $(CH_3)_2Hg$. It is abbreviated as Me_2Hg in this report.

Hg(0) Oxidation—This transformation, where Hg(0) is oxidized to Hg(II), is thought to result primarily from abiotic processes and can occur in various environmental media including air, snow, and water. An important example is the photochemical oxidation of atmospheric Hg(0) that leads to deposition of Hg(II) on marine and terrestrial surfaces.

Hg(II) Reduction—This transformation, where Hg(II) is reduced to Hg(0), mediates the evasion of Hg(0) to the atmosphere from water, ice, snow, and soil. It is thought to be driven largely by photoreduction, although microbial activity may also play an important role, particularly in the marine environment.

Methylation—This transformation of Hg(II) into a more toxic organic form, MeHg, is thought to occur primarily through the activity of anaerobic microorganisms under anoxic conditions in sediment and water. Dimethylmercury is also formed in marine environments through methylation processes.

Demethylation—The breakdown of MeHg to Hg(II) or Hg(0) may occur via photochemical or microbial processes in water and sediment. In marine environments, Me_2Hg can be demethylated to form MeHg or inorganic Hg.

TABLE 2.1 Common measures of Hg in environmental samples

Sample Matrix	Analytical Method (abbreviation)	Description
Air		
	Reactive Gaseous Hg (RGM)	An operationally defined species assumed to be primarily gaseous Hg(II)
	Particulate Hg (PHg)	Hg(II) in airborne particles that is retained on a filter
	Gaseous Elemental Hg (GEM)	Hg(0) in air, following filtration which removes PHg and is thought to also remove RGM
Water		
	Total Hg (THg)	All Hg in water including inorganic and organic species; measured in unfiltered water or the particulate and dissolved fractions
	Reactive Inorganic Hg (RHg)	Operationally defined Hg(II) that is easily reducible and thought to be highly reactive
	Monomethylmercury (MeHg)	Measured in unfiltered water or the particulate and dissolved fractions
	Dimethylmercury (Me_2Hg)	Measured in unfiltered water or the dissolved fraction
	Dissolved Gaseous Hg (DGM)	Volatile species of Hg in water, primarily Hg(0) but also Me_2Hg (in marine waters only)
Tissue		
	Total Hg (THg)	All Hg in tissue, primarily Hg(II) and MeHg
	Monomethylmercury (MeHg)	MeHg in tissue
	Organic Hg	All organic forms of Hg, considered to be mainly MeHg

2.3 Sources and emissions of Hg

Mercury occurs naturally in the environment although human activities since industrialization have enhanced its release and movement globally (Mason et al. 1994, Sunderland and Mason 2007). Little of the Hg present in the Arctic environment is derived from pollution sources within this region; most is transported from anthropogenic and natural sources outside of the Arctic (AMAP 2005). Mercury emitted to air from sources around the world may be transported to the Arctic because of its long residence time in the atmosphere. As a result, the Arctic is intimately and inextricably linked to the global Hg cycle and global anthropogenic emissions.

2.3.1 Global anthropogenic Hg emissions to air in 2005

The most recent global inventory of anthropogenic Hg emissions to air was compiled for the year 2005 under a joint AMAP/UNEP project (AMAP/UNEP 2008), with updated information included in the recent *AMAP Assessment 2011: Mercury in the Arctic* (AMAP 2011). The following briefly summarizes the global anthropogenic emissions inventory for Hg by source sector and geographic region. More details on the inventory estimates can be found in Munthe et al. (2011).

By source sector in 2005

Anthropogenic Hg emissions to the atmosphere are generated either as an unintentional by-product of (mainly) industrial activities or through intentional use of Hg in products, processing, and their associated waste streams. The main *by-product* sectors are stationary combustion of fossil fuels in power plants and for residential heating, pig iron and steel production, non-ferrous metal production, cement production, industrial-use Hg production, and large-scale gold production. Intentional-use sectors include artisanal and small-scale gold mining (ASGM), emissions from dental amalgam (during cremations), chlor-alkali production, and waste disposal (including waste incineration).

The largest anthropogenic emissions of Hg to the global atmosphere occur as a by product of the combustion of fossil fuels, mainly coal in power plants and industrial and residential boilers. Combustion of fossil fuels for energy and heat production represented 46% of the roughly 1920 tonnes (t) of emissions from all anthropogenic sources worldwide in 2005 (Fig. 2.1). Emissions of Hg from coal combustion were between one and two orders of magnitude higher than emissions from oil combustion, depending on the country. Various factors affect the emission of Hg to the atmosphere during combustion of fuels. The most important are the Hg content of the coal and the type and efficiency of control equipment that can remove Hg from exhaust gases—as well as, naturally, the amount of fuel combusted.

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Other sectors were also responsible for significant emissions of Hg to the atmosphere (Fig. 2.1). The non-ferrous and ferrous metal industries—excluding Hg and gold production—were estimated to contribute about 10% to total anthropogenic Hg emissions. Depending on the country, primary metal production using ores emitted between one and two orders of magnitude more Hg than secondary smelters—with scrap as the main raw material. The production of metallurgical coke was the major source of atmospheric Hg related to the iron and steel industry. In the cement industry, the fuel-firing kiln system and the clinker-cooling and handling system were responsible for about 10% of the total anthropogenic Hg emissions on a global scale. Artisanal and small-scale gold mining contributed about 17% to global Hg emissions while industrial (large-scale) gold production using Hg technology contributed another 6%. The use of the Hg cell process to produce caustic soda in the chlor-alkali industry has decreased significantly over the past 15 years worldwide. The chlor-alkali industry in 2005 accounted for about 2.5% of total anthropogenic Hg emissions worldwide. Mercury production for industrial uses contributed just over 0.5% to global Hg emissions.

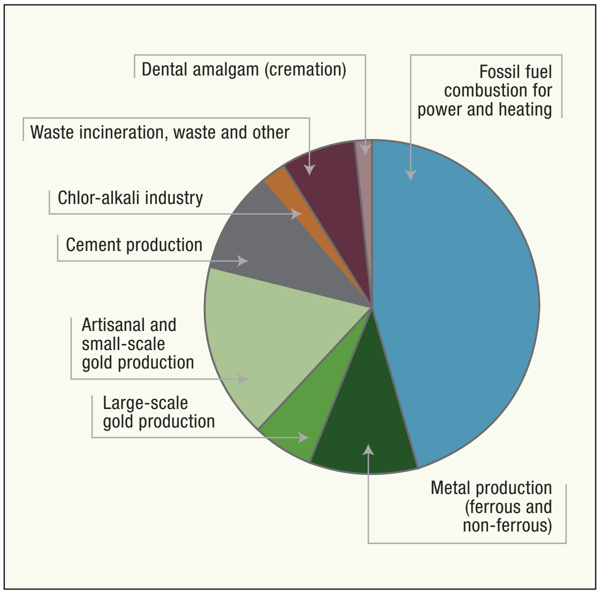

FIGURE 2.1

Proportion of global anthropogenic emissions of Hg to air in 2005 from various sectors (source: AMAP 2011).

The global product-related emissions of Hg—including all major uses of Hg in products—were estimated to be roughly 125 t (6.5% of total anthropogenic emissions) (Munthe et al. 2011). It is noteworthy that about 30% of the product-related emissions arose from waste incineration and another 52% from landfill waste.

By region in 2005

Asian countries contributed roughly 65% of global Hg emissions from anthropogenic sources in 2005, followed by North America and Europe (Fig. 2.2). Combustion of fuels to produce electricity and heat was the largest source of anthropogenic Hg emissions in Europe, North America, Asia, and Russia, and was responsible for about 35–50% of the anthropogenic emissions in Oceania and Africa. However, in South America, ASGM was responsible for the largest proportion of their emissions (about 60%). Relatively large Hg emissions from ASGM explained why some Asian and South American countries such as Indonesia, Brazil, and Colombia appeared in the top ten ranked Hg emitting countries (Fig. 2.3). If by-product emission sectors alone were considered, no South American countries were represented and all other countries listed had a high degree of industrial development.

China was the largest single emitter of Hg worldwide, by a large margin (Fig. 2.3). Power plant emissions were an important part of the total combustion emissions of Hg in China. However, ongoing restructuring and improved emission control of air pollutants in the Chinese power sector may have reduced these emissions in recent years. Equally significant were emissions from combustion of poor-quality coal mixed with various kinds of wastes used in small residential units in rural areas to produce heat and cook food. China contributed about 40% (600 t) to the global by-product emissions of Hg, and this contribution may have been even higher if Hg emission factors for non-ferrous metal production in China were underestimated. China also had significant emissions from ASGM.

Together, China, India, and the United States were responsible for roughly 60% of the total estimated global anthropogenic emission inventory for 2005 (1,095 out of 1,920 t). Canada emitted an estimated 7 t of anthropogenic Hg to the atmosphere in 2005, representing < 1% of total global anthropogenic emissions (GOC 2010).

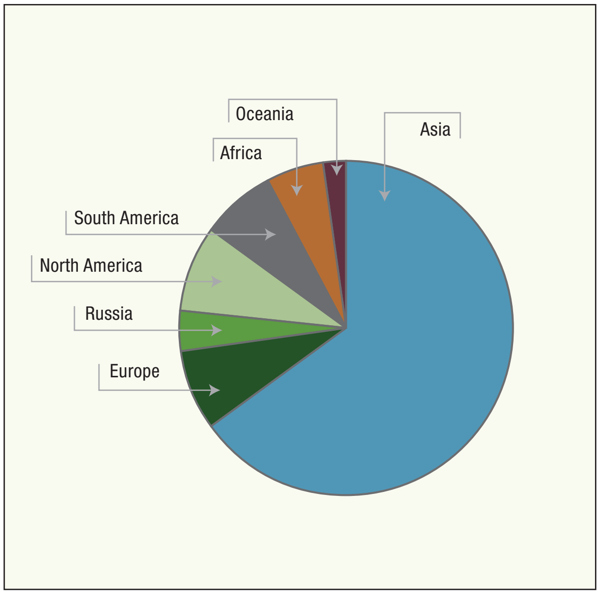

FIGURE 2.2

Proportion of global anthropogenic emissions of Hg to air in 2005 from different regions (source: AMAP 2011).

2.3.2 Local sources of Hg pollution in the Canadian Arctic

Environmental releases of Hg by local human activities are not considered important on a regional level within the Canadian Arctic. Although Hg is emitted to the air through fossil fuel combustion and waste incineration at Arctic communities and industrial sites, international sources likely dominate the atmospheric delivery of Hg to the Canadian Arctic (Durnford et al. 2010). There are also contaminated sites in northern Canada that have released Hg to aquatic ecosystems and terrestrial soils. A review of these contaminated sites, specifically Dew-line stations and abandoned mines, was conducted in the previous *Canadian Arctic Contaminants Assessment Report (CACAR II)*, and Hg was not identified as a main contaminant in the vast majority of cases (Bidleman et al. 2003). Mercury was identified as a local concern in only a few cases: in runoff from two abandoned mines in the Northwest Territories (Discovery Mine and Terra #1); in soil from the remediated Aishihik airstrip in Yukon; and, in groundwater at the remediated Rainy Hollow and Border Pump Station in northern British Columbia (Bidleman et al. 2003). Therefore, local sources of Hg pollution to the Arctic environment are not considered further in this assessment.

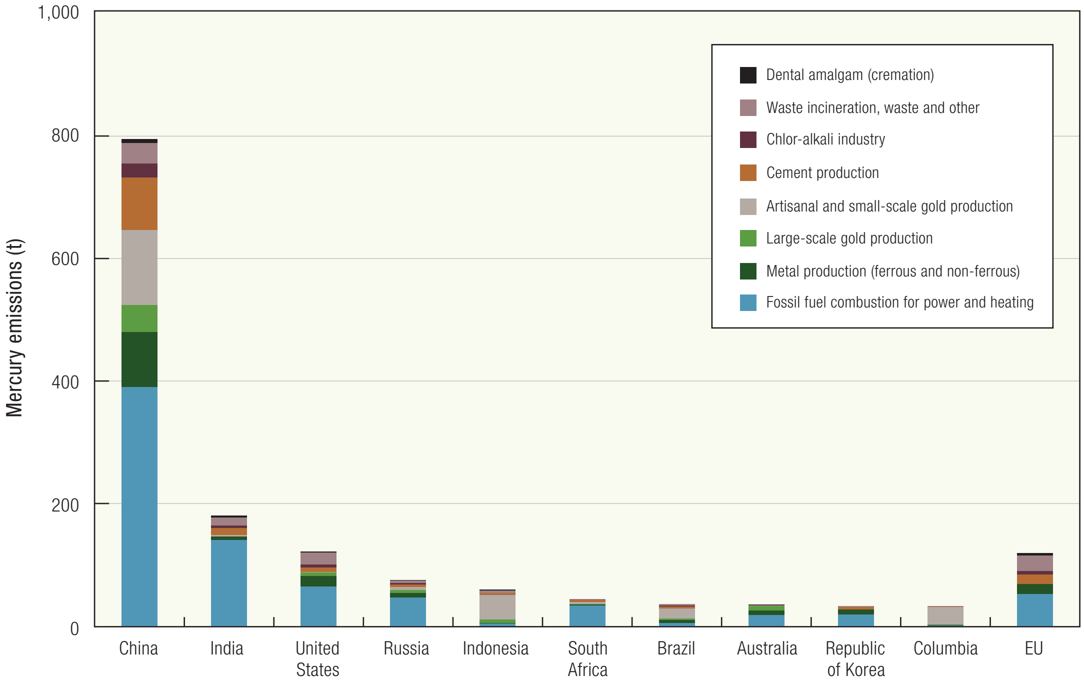

FIGURE 2.3

Emissions of Hg to air in 2005 from various anthropogenic sectors in the ten largest emitting countries (source: AMAP 2011).

2.3.3 Natural sources of Hg to the Arctic

Mercury occurs in the earth's crust, especially in the form of the mineral cinnabar whose chemical composition is Hg(II) sulphide. The metal is released naturally through the weathering of rocks and as a result of volcanic and geothermal activities. In pre-industrial times, natural Hg was globally dispersed via atmospheric transport, freshwater discharge, and oceanic currents (Sunderland and Mason 2007). Mercury emissions to the atmosphere occurred mainly from volcanic eruptions, photochemical or biological reduction of Hg(II) in water and soil, and from wildfires. Wildfires and intentional biomass burning can release back into the atmosphere significant amounts of Hg that was sequestered in forests and soils (Friedli et al. 2009). The natural release of Hg continues globally and is compounded by the Hg released by human activities over the past two centuries. Anthropogenic activities have enriched global atmospheric levels of Hg several-fold above pre-industrial conditions (Mason et al. 1994, Sunderland and Mason 2007).

FIGURE 2.4

Mercury levels in the Arctic environment are affected by global sources through ocean currents (blue arrows) and long-range atmospheric transport (white arrows).

2.4 Movement of Hg through the Arctic

2.4.1 How Hg is delivered to Arctic environments

On a global scale, the movement of Hg through the environment is constrained by the amount that accumulates in different reservoirs—namely, the atmosphere, ocean and terrestrial soil—and the processes and rates of exchange between those reservoirs. The world's surface soils are by far the largest Hg reservoir

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(Sunderland and Mason 2007). However, with the exception of soils present in the Arctic itself, Hg in global soil only interacts indirectly with the Arctic environment through the atmosphere and ocean. The most important pathways for transport of global Hg to the Arctic involve upper ocean layers and the atmosphere. Mercury is also removed from the Arctic by these same pathways. Sunderland and Mason (2007) estimated that approximately 134,000 t of Hg resides in the global upper oceans and about 5,600 t in the atmosphere. Relative to the pre-industrial period, these reservoirs include pollution-related enhancement of about 25% and 300–500%, respectively. There is geographic disparity in the anthropogenic increases in oceans, with the North Atlantic Ocean north of 55° N—a major source of seawater to the Arctic Ocean—exhibiting more than a 50% increase in Hg storage (Sunderland and Mason 2007).

Mercury levels in the Arctic environment are affected by global sources through ocean connectivity and atmospheric deposition (Fig. 2.4). Currents transport Hg to the Arctic Ocean via the Bering Strait from the Pacific Ocean and via the Fram Strait and Barents Sea from the Atlantic Ocean. Atmospheric Hg is deposited on Arctic terrestrial and marine surfaces through wet precipitation and dry deposition. The atmospheric transport of Hg to the Arctic is faster (within days) than via ocean currents, which may take decades. Residence times for Hg in the atmosphere are also much shorter than in the upper oceans and are estimated at 0.7 and 27 years, respectively (Sunderland and Mason 2007). These differences in residence time imply that changes in Hg storage in the global oceans and atmosphere will impact the Arctic environment on varying time scales. That is, relatively rapidly for the atmosphere and slowly for marine waters.

On a regional scale, Hg moves between terrestrial, freshwater, and marine environments within the Arctic via atmospheric deposition, oceanic transport, coastal erosion, and fluvial transport (Box 2.2). Mercury delivered by these pathways may originate from natural sources within the Arctic or from long-range transport of emissions from a combination of anthropogenic and natural sources outside the Arctic. The fate of Hg in the environment depends on its form and on the subsequent biogeochemical transformations that occur (Box 2.1). Atmospheric GEM that does not undergo oxidation and deposition onto Arctic surfaces is transported out of the Arctic by the movement of air masses. Oxidized Hg in terrestrial and aquatic environments may be reduced to Hg(0) and released back into the atmosphere. Inorganic Hg may be transformed into

BOX 2.2 Pathways of Hg delivery to Arctic ecosystems

Atmospheric Deposition—Atmospheric Hg is deposited onto terrestrial and marine surfaces through wet precipitation and dry deposition.

Fluvial Transport—Lakes receive Hg from their watersheds (accumulated in snow, soil, and wetlands) via runoff and stream inflows. Large rivers transport terrestrial Hg to marine environments. Mercury transported by fluvial processes originates from bedrock weathering and atmospheric deposition.

Oceanic Transport—Water currents transport Hg to the Arctic Ocean via the Bering Strait from the Pacific Ocean and via the Fram Strait and Barents Sea from the Atlantic Ocean. Currents also move Hg between water masses within the Arctic Ocean.

Coastal Erosion—Soil-bound Hg enters Arctic marine environments through coastal erosion driven by wave action and permafrost degradation.

Biotransport—A phenomenon whereby migrating animals that congregate at high densities release their body burden of Hg (via carcasses, feathers, excrement, etc.), which results in localized contamination of a site. Spawning sites for salmon and breeding colonies of birds are two such examples.

MeHg, enter food webs, and biomagnify to top predator animals. Mercury may also be captured in aquatic sediments, terrestrial soils, and glacier ice and this will result in long-term storage.

The biological transport of Hg can be a significant pathway for the localized contamination of ecosystems where fish and birds congregate at high densities in the Arctic (Blais et al. 2007). Earlier studies on sockeye salmon in Alaska demonstrated that carcasses represent a large, discrete flux of Hg to inland waters (Zhang et al. 2001). More recently, seabirds have been identified as biovectors of Hg to ponds adjacent to nesting colonies in the High Arctic (Blais et al. 2005, Choy et al. 2010, Michelutti et al. 2010). Birds transport and concentrate marine-derived Hg in freshwater ponds by feeding in the ocean and then releasing their guano at sites where they congregate for nesting. Seabirds that occupy a higher trophic position are more efficient biovectors of Hg (Michelutti et al. 2010). While biovector transport is not considered a major flux of Hg in the Canadian Arctic on a large scale (Outridge et al. 2008), it can increase Hg several-fold in sediment adjacent to seabird colonies (Blais et al. 2005, Michelutti et al. 2010).

2.4.2 Bioaccumulation of Hg in organisms and its biomagnification through food webs

Bioaccumulation

Mercury accumulates in the tissues of organisms over time—termed bioaccumulation. Fish and wildlife are mainly exposed to Hg through their diet because typically low concentrations in air and water result in minimal transfer through inhalation, gill or dermal exposure (Rodgers 1994, Hall et al. 1997, Duffy et al. 2001). Bioaccumulation occurs because Hg is efficiently assimilated into tissues following absorption in the gut but is slowly eliminated from the body. In freshwater fish, for example, the estimated time required to excrete half their Hg burden is nearly 500 days (Van Walleghem et al. 2007).

Bioenergetic models of contaminant bioaccumulation show that the concentration of Hg in an animal's body is related to processes of energy intake and metabolism (Trudel and Rasmussen 2001, Karasov et al. 2007). The uptake of Hg into the body over time is determined by the rate of food consumption, the Hg content of the food, and the efficiency of absorption in the gut. Mercury is eliminated from the body through several loss processes, primarily excretion, reproduction (e.g., transfer to offspring) and/or moulting (Wiener et al. 2003). Growth also influences tissue concentrations of Hg (but not the total burden) in an animal's body. Individuals with higher growth rates tend to have lower Hg concentrations because more biomass is produced per unit of energy (and Hg) consumed; this phenomenon is referred to as a *biodilution effect* (Kidd et al. 1999, Karimi et al. 2007). Therefore, the balance between rates of growth and Hg uptake and loss determine the concentration of Hg in fish and wildlife.

Methylmercury is the main species of Hg absorbed by fish and wildlife. Once MeHg is absorbed in the gut, it enters the blood stream and is distributed to tissues and organs in the body. In fish, MeHg first accumulates in the viscera (kidney, spleen, liver) and is only later redistributed to muscle and brain tissue (Oliveira et al. 1999). Mammals and birds are capable of detoxifying their MeHg burden through demethylation (Wagemann et al. 1998, Wiener et al. 2003). As a result, a large and variable portion of the Hg in liver and kidney is inor-

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ganic, and it may be rendered non-toxic through complexation with selenium (Wagemann et al. 1998, Wiener et al. 2003). The role of selenium in Hg detoxification is

discussed further in section 7.3. Mercury in skeletal muscle and brain is predominately in the form of MeHg (Scheuhammer et al. 1998, Wagemann et al. 1998, Basu et al. 2009), which is more toxic than Hg(II). In birds, moulting is an important detoxifying mechanism, and feathers can accumulate large amounts of organic Hg (Braune and Gaskin 1987, Kim et al. 1996).

Mercury levels often increase in older and larger individuals of an animal population. For example, tissue concentrations of Hg in beluga or Arctic char are positively correlated with their length and age (Loseto et al. 2008b, Gantner et al. 2010). Size and age may influence Hg bioaccumulation because older, larger individuals tend to eat larger types of prey that are at a higher trophic position and are more contaminated. Mercury bioaccumulation can also be related to age because if the rate of Hg elimination is slower than the rate of Hg uptake in the body, it will result in accumulation over time. In some cases, older and larger individuals can have higher metabolic costs that reduce growth dilution of Hg and result in higher concentrations (Trudel and Rasmussen 2006).

Biomagnification of Hg through food webs

In the Canadian Arctic, MeHg is found in marine and fresh waters at ultra-low levels—commonly less than 1 part per trillion—yet concentrations are orders of magnitude higher in predatory animals such as beluga and polar bears. This phenomenon occurs because MeHg is biomagnified through a food web; it first concentrates in microbes (algae and bacteria) and then is further amplified in dietary transfers between consumers (Fig. 2.5). The greatest biomagnification occurs at the base of food webs where MeHg is approximately 10^5 times more concentrated in microbes than in water (Watras et al. 1998). Biomagnification also occurs at each additional trophic level, by a factor of about two to seven times between a consumer and its diet in freshwater ecosystems (Mason and Sullivan 1997, Gorski et al. 2003), and by as much as 13–23 times in Arctic marine food webs (Loseto et al. 2008a).

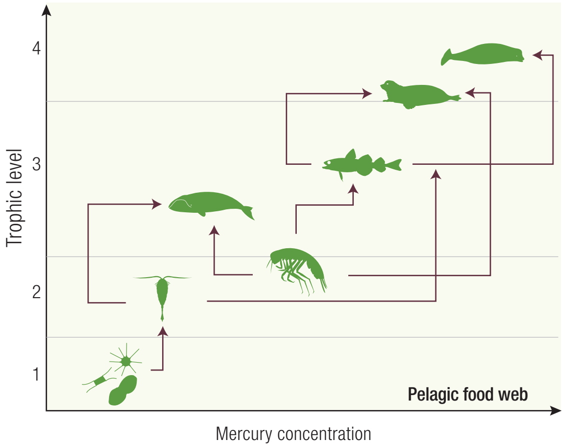

FIGURE 2.5

Schematic diagram of Hg biomagnification through a pelagic food web in the Arctic (source: AMAP 2011).

Consumers that feed at a higher position in the food chain contain more Hg because of biomagnification during dietary transfer.

In aquatic food webs, a key factor in determining levels in top predator species is the concentration of MeHg that is bioavailable for uptake by microbes. Methylmercury may move across the membrane of microbial cells by passive diffusion or active transport (Mason et al. 1996, Moye et al. 2002). It forms complexes with dissolved organic compounds in water, and the nature of this complexation can either inhibit or enhance MeHg accumulation in cells (Lawson and Mason 1998, Pickhardt and Fisher 2007, Gorski et al. 2008). The standing pool of MeHg in water is determined by biogeochemical processes of MeHg production and decomposition, and by transport from other environments.

In terrestrial food webs of the Arctic, primary producers such as sedges, lichens, moss, and vascular plants are the main entry points for Hg. Lichens, which are an important food for many Arctic herbivores, have no root system and accumulate Hg mainly from the atmosphere (Estrade et al. 2010). Below the tree line, foliage of trees and shrubs also accumulates Hg mainly from atmospheric deposition directly onto leaves rather than through uptake by roots (Grigal 2002, Ericksen et al. 2003, Graydon et al. 2009b). Terrestrial food webs have been less studied in the Arctic relative to marine and fresh waters, and factors that control Hg uptake by these primary producers remain poorly understood.

2.4.3 Influence of climate change on Hg cycling in the Arctic

Climate change has the potential to affect the movement of Hg in Arctic ecosystems because of intimate links between the Hg cycle and temperature, the cryosphere, and organic matter cycling (Macdonald et al. 2005). Changes to food webs driven by climate warming are also anticipated to impact Hg biomagnification to top predator animals (Macdonald et al. 2005). The role of climate change in the Hg cycle is a complex issue because of a multitude of potential interactions as well as uncertainties in our understanding of both the movement of Hg and environmental change in the Arctic. Empirical evidence is beginning to accumulate on climate change impacts to the Hg cycle in the Canadian Arctic, and these findings are presented in chapters 3 to 6 of this report according to the environment where they were observed. Given the emerging nature of this issue, the purpose of this section is to provide a brief overview of potential connections between climate change and aspects of the Hg cycle, specifically: 1) the delivery of Hg to Arctic ecosystems; 2) biogeochemical transformations; and, 3) food web dynamics. A more in-depth discussion on this topic can be found in Chapter 4 of the recent *AMAP Assessment 2011* (Stern et al. 2011) and in Macdonald et al. (2005).

Recent climate change in the Canadian Arctic

The Arctic climate is changing at a rapid rate (ACIA 2005, Furgal and Prowse 2009). Natural archives such as lake and marine sediments, glacier ice, and tree rings, which provide a long-term historical perspective on climate change, show that the Northern Hemisphere has been warming more quickly in the last 150 years than in the previous two millennia (Mann and Jones 2003, Moberg et al. 2005). Instrument measurements recorded in the Canadian Arctic since the middle of the 20th Century indicate that temperatures are warming, precipitation is increasing, and greater melt of ice and snow is occurring (Box 2.3). Aboriginal peoples living in different Arctic regions of Canada have observed recent environmental changes such as more variable and less predictable weather, thinner sea ice conditions, and lower water levels (Huntington et al. 2005).

Although widespread change is occurring in the Arctic, it is not uniform across this vast region. The Canadian Arctic—broadly defined here as the area north of 60° latitude—covers about 4 million km^2 of land with diverse climate, landforms, vegetation and wildlife

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(Prowse et al. 2009a). Enormous marine environments extend approximately 80° in longitude across northern Canada including the Arctic Ocean, Beaufort Sea, Hudson Bay, and Baffin Bay. Regional variation must therefore be considered in the context of climate change (Prowse et al. 2009a). In the second half of the 20th Century, greater warming occurred in the western Canadian Arctic whereas precipitation increased more in the northeast (Box 2.3). Environmental responses to climate change have varied and will likely continue to vary among different Arctic regions. Recent maximum increases in shallow permafrost temperature were higher in the western Arctic than in the High Arctic, and glaciers in these areas are melting at variable rates (Box 2.3).

Potential effects on the delivery of Hg

Climate change could impact the delivery of Hg to Arctic environments by altering atmospheric deposition, hydrological transport, permafrost inputs, and local sources from human activities. Brief descriptions of these potential connections between Hg cycling and climate change are provided as follows.

Atmospheric deposition (section 3.2)—Atmospheric deposition of Hg during atmospheric mercury depletion events (AMDEs) is driven by the oxidation of Hg(0) with bromine atoms, a reaction that is likely temperature sensitive (Goodsite et al. 2004, Cole and Steffen 2010). Based on theoretical calculations, rising temperatures in the Arctic could slow down the oxidation and deposition of atmospheric Hg in marine environments. The supply of reactive bromine for Hg(0) oxidation may also be affected by changes to sea ice formation and by temperature effects on bromine speciation. Recent anomalies in atmospheric pressure fields in the Arctic could alter air currents which are the pathways of long-range Hg(0) transport from different emission source regions (Macdonald et al. 2005).

Hydrological transport (section 5.3)—Rivers are a significant source of Hg to marine environments

BOX 2.3 Some recent observations of a changing environment in the Canadian Arctic*

Air Temperature:	Mean annual air temperatures increased by 0.8–2.2°C from 1948 to 2005 in different Arctic regions, with the greatest warming occurring in the western Arctic (Yukon and Mackenzie District). Trends were strongest in the winter and spring when, for example, the average winter temperature in the Yukon warmed by 4.5°C.
Precipitation:	Total annual precipitation increased 16–25% from 1948 to 2005 in more northerly Arctic tundra and mountain regions, primarily in Nunavut. Trends were strongest in fall, winter, and spring. The western Arctic had small, insignificant increases (2–5%) during the same time period.
Glacier Cover:	Glaciers in the Canadian Arctic have retreated during the 20th Century. In the Arctic Archipelago, the rate of ice loss was 25 $km^3\ y^{-1}$ for the period of 1995 to 2000. Greater release of fresh water has occurred from glacier melting in the Yukon.
Snow Cover:	The duration of annual snow cover has decreased by about 20 days in both the eastern and western Canadian Arctic since 1950.
Lake and River Ice:	Freeze-up and break-up dates for lake and river ice are shifting towards a longer open-water season. Historical records from 1856 to 1995 indicate an earlier break-up by 6.5 days per century and a later freeze-up by 5.8 days per century for lakes and rivers in the Northern Hemisphere (Magnuson et al. 2000). Limited observations are available for the Canadian Arctic, but they are consistent with these trends. The Mackenzie River showed a later freeze-up by 6.1 days from the late 19th to the late 20th Century (Magnuson et al. 2000).
Sea Ice:	The extent of sea ice in the Arctic Ocean during late summer decreased by 11% per decade between 1979 and 2009 (NSIDC 2009).
Permafrost:	The active layer of permafrost warmed and deepened in recent decades, with considerable variation among regions. Shallow permafrost temperatures in the High Arctic increased by 0.3–0.5°C per decade near the end of the 20th Century, while those in the western Arctic increased from 0–1°C.

* Unless otherwise referenced, information was summarized from the Federal Government report *From Impacts to Adaptation: Canada in a Changing Climate 2007* (Lemmen et al. 2008).

such as Hudson Bay (Hare et al. 2008). Fluxes of Hg from rivers could be altered by long-term changes in hydrology because of the positive relationship between discharge and aqueous Hg concentrations (Leitch et al. 2007, Graydon et al. 2009a). Snowmelt is the dominant source of Hg to many Arctic lakes (Semkin et al. 2005), and changes to the amount of snow, the timing of snowmelt, or the importance of rain in the hydrological cycle (Prowse et al. 2006) could have implications for Hg delivery to these ecosystems. Higher annual precipitation could increase wet deposition rates of Hg to Arctic environments.

Permafrost thaw (section 5.4.3)—Melting of permafrost and deepening of the active layer in summer are expected to increase particulate and dissolved organic carbon transport to Arctic lakes and rivers (Prowse et al. 2006, Xu et al. 2009). Watershed export of Hg, which is associated with organic carbon transport, may consequently also change. Permafrost slumping and subsidence may result in the export of soil-bound Hg to fresh waters (Klaminder et al. 2008, Rydberg et al. 2010).

Local anthropogenic sources—Reductions in summer sea ice could stimulate commercial shipping through the Northwest Passage and economic development throughout the Canadian Arctic (Prowse et al. 2009b). Greater accessibility through new shipping routes and a longer open-water season may allow for more exploration activity and the extraction of oil, gas, and mineral resources during the 21st Century (Prowse et al. 2009b). These activities present potential risks to the Arctic environment including increased fossil fuel combustion and waste generation that could result in new local sources of Hg.

Potential effects on biogeochemical transformations of Hg

Climate change could impact biogeochemical transformations of Hg, in particular, methylation, photochemical reactions, and dynamic processes in the cryosphere. Brief descriptions of these potential connections between Hg cycling and climate change are provided as follows.

Methylation (sections 4.5.2, 5.5.1 and 6.7.1)—Mercury methylation results primarily from bacterial activity in sediment and in the water column where rates are affected by temperature, the influx of organic matter, pH, redox potential, Hg(II) complexation and concentration, and sulphate concentration (Ullrich et al. 2001). Production of MeHg in the Arctic may be affected by environmental change including temperature warming, a longer ice-free season, altered stratification and redox conditions, or increased watershed inputs of nutrients, inorganic Hg, and dissolved organic matter.

Photochemical transformations (sections 5.5.2 and 6.7)—Photochemical reactions play a key role in the demethylation of Hg (Sellers et al. 1996, Hammerschmidt and Fitzgerald 2006) and in the production of volatile Hg(0) in aquatic environments (Amyot et al. 1997, Tseng et al. 2004). Mercury fluxes driven by these key photochemical reactions may be affected through longer exposure to light during the ice-free season or changes to the aquatic light environment from watershed inputs of coloured dissolved organic matter.

Transformations in the cryosphere (sections 4.3, 4.5.2, 6.4 and 6.7)—Arctic snow and ice are important environmental surfaces where key biogeochemical transformations of Hg are thought to occur, particularly Hg(II) reduction to volatile Hg(0) and potentially also Hg methylation. In spring, sea ice is a highly heterogeneous environment where drainage channels, melt ponds, brine pockets, and films of underwater ice algae may be sites of Hg transformations and entry into the food web (Poulain et al. 2007, Macdonald and Loseto 2010, Chaulk et al. 2011). Changes to snow and sea ice conditions, including the length of the ice-free season and sea-ice thickness, may have consequences for Hg fluxes in the Arctic.

Potential effects on food web bioaccumulation

Climate warming may affect Hg levels in biota through changes in food web structure, growth rates, or ecosystem productivity. A change in the amount of Hg available for uptake in food webs (as previously described) may also affect bioaccumulation. Brief descriptions of these potential connections between Hg cycling and climate change are provided as follows.

Food web structure (sections 5.7–5.8 and 6.8–6.11)—Trophic position and feeding habitat are important aspects of food web structure that affect the bioaccumulation of Hg in biota (Atwell et al. 1998, Power et al. 2002, Loseto et al. 2008a). Environmental change driven by climate warming may impact food webs through changes in species composition, the availability of suitable habitat, or the abundance of different prey for top predators (ACIA 2005). Dietary changes could affect Hg bioaccumulation in predators if their trophic position shifts or if they feed in a new habitat (e.g., benthic versus pelagic zones or estuary versus offshore waters) because of variation in the level of Hg contamination.

Growth rates—Shifts in the thermal regime, ice conditions, or food availability in marine and freshwater environments may alter growth rates of Arctic fish and marine mammals (Reist et al. 2006, Chambellant 2010, Peacock et al. 2010). The resulting changes in growth may, in turn, affect their bioaccumulation of Hg. Organisms with higher growth rates tend to have lower Hg concentrations (section 2.4.2). The direct effect of warmer temperatures on Hg bioaccumulation will depend on whether a species is near its temperature optimum for growth. For example, cold-adapted fish species such as Arctic char and lake trout grow less efficiently in warmer waters (Reist et al. 2006), which could potentially lead to higher Hg concentrations.

Ecosystem productivity—Climate warming will likely increase the productivity of aquatic ecosystems which may, in turn, reduce Hg bioaccumulation in biota through growth dilution processes at lower trophic levels in the food web. As phytoplankton production increases, the concentration of Hg per algal cell decreases, resulting in less transfer to zooplankton (Pickhardt et al. 2002). A similar reduction in Hg concentration occurs in benthic algae that have higher growth rates (Hill and Larsen 2005). Zooplankton with higher growth rates also have lower Hg concentrations because more biomass is produced per unit of food (and Hg) consumed (Karimi et al. 2007). Bio-dilution effects have been observed in highly nutrient-enriched lakes at temperate latitudes (Chen and Folt 2005); it is unclear to what extent biodilution will occur with more modest increases of aquatic productivity in the Arctic.

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2.4.4 Source- and process-driven bioaccumulation of Hg in a changing climate

Concentrations of Hg have increased in Arctic marine mammals since the onset of the Industrial Era (section 6.11.1), and there is little doubt that theses changes are a result of increased Hg emissions to the atmosphere from global anthropogenic activities. What makes the phenomenon of Hg bioaccumulation in the Arctic puzzling, however, is an apparent lack of a dose-response relationship between Hg concentrations in the Arctic atmosphere and those in animals over recent decades. Since the 1970s, tropospheric Hg concentrations in the circumpolar Arctic have been constant or slightly declining (section 3.5; Munthe et al. 2011) due to emission controls in Europe and North America. During the same period, Hg concentrations in Arctic animals have shown a remarkable degree of spatial and temporal variation (this report; Rigét et al. 2011). This wide variation in animal Hg concentrations suggests that Hg bioaccumulation in Arctic ecosystems in recent decades may no longer be primarily driven by atmospheric concentrations. Factors affecting deposition rates from the atmosphere (Dastoor et al. 2008, Cole and Steffen 2010) and post-depositional biogeochemical processes of Hg (Wang et al. 2010) need to be considered more fully to understand the recent trends in biota. While biogeochemical processes are important for Hg bioaccumulation in any ecosystem, dramatic climate change in the Arctic in recent decades has potentially altered, and further change will likely alter, the cycling of Hg with consequences for its bioaccumulation.

2.5 Summary

Mercury is a heavy metal with a complex cycle in the environment because it originates from many sources (both natural and anthropogenic), it is transformed into various chemical species by biogeochemical processes, and it is transported to ecosystems via several pathways. Mercury is transformed from one species to another through oxidation or reduction reactions and methylation or demethylation reactions driven by abiotic and/or biological mechanisms. These transformations alter the chemical properties of Hg, thereby affecting its retention and movement through the environment and its accumulation in food webs.

Mercury occurs in the earth's crust, especially in the form of mineral cinnabar. This metal is released from natural sources through the weathering of rocks and as a result of volcanic and geothermal activities. Globally, the natural release of Hg continues today, and this provides a ubiquitous base amount of Hg in all environmental media to which has been added the Hg released by human activities over the past two centuries. Anthropogenic activities have enriched global atmospheric levels of Hg several fold above pre-industrial conditions.

The stability of gaseous Hg(0) allows it to be transported over long distances through the atmosphere and deposited onto terrestrial and marine surfaces around the world, making it a truly global pollutant. In 2005, the largest anthropogenic emissions of Hg to the global atmosphere occurred from the combustion of fossil fuels, mainly coal in power plants and industrial and residential boilers. Metal production (ferrous, non-ferrous and gold), cement production, and artisanal or small-scale gold mining were other important sources of global Hg emissions. Asian countries contributed the most to global Hg emissions from anthropogenic sources in 2005, followed by North America and Europe. Within the Canadian Arctic, environmental releases of Hg by local human activities are not considered significant on a regional level.

Mercury levels in the Arctic environment are affected by global sources through ocean connectivity and atmospheric deposition. Currents transport Hg to the Arctic Ocean from the Pacific and Atlantic oceans, while atmospheric Hg is deposited onto Arctic terrestrial and marine surfaces through precipitation and dry deposition. On a regional scale, Hg moves between terrestrial, freshwater and marine environments within the Arctic via atmospheric deposition, oceanic transport, coastal erosion, and fluvial transport. Mercury delivered by these pathways may originate from natural sources within the Arctic or from long-range transport of emissions from anthropogenic and natural sources outside the Arctic. The fate of Hg in the environment depends on its form and the subsequent biogeochemical transformations that occur.

Mercury bioaccumulates in the tissues of organisms over time. Fish and wildlife are mainly exposed to Hg through their diet because typically low concentrations in air and water result in minimal transfer through inhalation, gill, or dermal exposure. The concentration of Hg in an animal's body is related to processes of energy intake and metabolism; the balance between rates of growth, Hg uptake and Hg excretion determines the concentration of Hg in tissues.

In the Canadian Arctic, MeHg is found in marine and fresh waters at ultra-low levels yet concentrations are orders of magnitude higher in predatory animals. This phenomenon occurs because MeHg is biomagnified through a food web; it first concentrates in microbes (algae and bacteria) and then is further amplified in dietary transfers between consumers. In aquatic environments, the concentration of MeHg that is bioavailable for uptake by microbes is a key factor determining levels in top predator species. In terrestrial food webs of the Arctic, primary producers such as sedges, lichens, moss, and vascular plants are the main entry points for Hg. Lichens and foliage of trees and shrubs mainly accumulate Hg directly from the atmosphere.

The climate in northern Canada is rapidly changing, although these changes are not uniform across this vast and diverse region. Climate change has the potential to affect the movement of Hg in Arctic environments because of intimate links between the Hg cycle and temperature, the cryosphere, and organic matter cycling. Changes to food webs driven by climate warming are also anticipated to impact Hg biomagnification to top predator animals. The role of climate change in the Hg cycle is a complex and emerging issue because climate change may potentially affect the transport of Hg, biogeochemical transformations, and food web dynamics in Arctic environments.

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Atmospheric Mercury

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3.1 Introduction

The atmosphere plays a fundamental role in the mercury (Hg) cycle through long-range atmospheric transport to the Canadian Arctic and local deposition onto terrestrial, freshwater, and marine surfaces. Mercury is a global pollutant that impacts the Arctic because of its long residence time in the air (6–24 months) and its transport from distant source regions. Scientific efforts to characterize the atmospheric transport and delivery of Hg to the Arctic have been challenging because Hg emissions to the air occur from both natural sources and anthropogenic activities, and complex exchanges of Hg occur between the air and its interfaces with soil, water, and the cryosphere.

Canadian scientists have led research on atmospheric Hg in the Arctic since the first atmospheric measurements of Hg were made in the High Arctic in the early 1990s. Much progress has been made on understanding the levels, transport, and transformations of Hg in Arctic air and the role that the atmosphere plays in this region. In 1995, Canadian scientists made the hugely influential discovery of atmospheric mercury depletion events (AMDEs). During springtime AMDEs, gaseous elemental Hg is rapidly depleted from the lower atmosphere through oxidative processes and deposited on the ground or physically bound to aerosol surfaces. This transformation has sparked tremendous interest in atmospheric Hg and the role of AMDEs in the Arctic Hg cycle.

By the end of the Northern Contaminants Program (NCP) Phase II in 2003, important progress had been made in the field of atmospheric Hg research in the Canadian Arctic. Automated, long-term measurements of atmospheric Hg at Alert revealed seasonal patterns resulting from AMDEs and rapid removal of Hg from the air within a period of hours after polar sunrise. Sites

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where AMDEs predominately occur and the chemistry responsible for these events were better characterized. The first measurements of ionic or non-elemental atmospheric Hg species, namely reactive gas phase Hg (RGM) and particulate Hg (PHg), were conducted at Alert (Nunavut) and Kuujjuarapik (Quebec). This new understanding of Hg processes in the Arctic was noted as a key finding in the previous *Canadian Arctic Contaminants Assessment Report (CACAR II)* (Bidleman et al. 2003). Although considerable progress was made in the study of atmospheric Hg behaviour in the Arctic, the report also concluded that the significance of AMDEs to the Arctic environment was yet to be determined.

Advances have been made in atmospheric research and monitoring during Phase III of the NCP. The chemical reactions resulting in oxidation of atmospheric Hg in the Arctic and the critical role of halogens have been characterized. Considerable field measurements on Hg speciation—measurements of gaseous elemental Hg (GEM) and its oxidized forms—are now available for the Canadian Arctic, particularly at Alert, and changes in speciation that occur during springtime AMDEs have been documented. Automated measurements of air Hg concentrations at Alert (High Arctic) and Kuujjuarapik (sub-Arctic) over the last decade provide new insights into temporal trends at different time scales. The geographic coverage of air measurements has been somewhat expanded with continuous data now available for one new site in the Yukon. Extensive modelling of atmospheric processes has been conducted. This has resulted in new estimates of long-range atmospheric transport of Hg from various source regions around the world as well as the impacts on Hg deposition and Hg cycling in the Arctic. These new scientific insights support the hypothesis that the cycle of atmospheric Hg is unique in the Arctic compared to lower-latitude regions and results in significant deposition of Hg to the Arctic environment. With a focus on Canadian results, this chapter will report on the most recent research findings since the *CACAR II*, identify knowledge gaps, and recommend directions for future research on the most important scientific issues concerning atmospheric deposition of Hg in the Canadian Arctic.

3.2 Atmospheric processes of Hg

Three forms of Hg are considered in atmospheric Hg processes: 1) GEM or elemental Hg [Hg(0)]; 2) RGM, which consists of oxidized gaseous Hg species—gas phase of monovalent or divalent inorganic Hg; and, 3) oxidized Hg associated with particles (PHg), each of which has a different residence time in the atmosphere. Gaseous elemental Hg can remain in the air for long periods of time and, thus, is transported globally. In contrast, more reactive oxidized forms of Hg are generally scavenged or deposited locally near emission sources either directly or following atmospheric chemical conversions. Hence, GEM is the dominant form of Hg that is transported to the Arctic through the atmosphere. It is the conversion of GEM to PHg and RGM in the Arctic atmosphere and subsequent deposition and methylation of these Hg species in aquatic environments that potentially lead to its bioaccumulation in biota and humans.

3.2.1 Overview of atmospheric Hg depletion events

AMDEs are characterized as a series of photochemically-initiated reactions in the atmosphere which result in the conversion of GEM to a short-lived form of Hg that can be deposited on terrestrial and aquatic surfaces. In 2008, a synthesis of AMDE science was produced by experts in this field of research that summarizes the most recent understanding of these processes (Steffen et al. 2008). Research from this and other reviews has shown that the initiation of AMDEs requires Hg in the atmosphere, cold temperatures, a stable inversion layer, sunlight, and reactive halogens (Poissant et al. 2008, Dommergue et al. 2009, Nguyen et al. 2009). Sea salt in ice or snow may provide a large source of halogen species that, under certain conditions, can react to form radicals such as bromine (Br), bromine monoxide (BrO), chlorine (Cl), and chlorine monoxide (ClO) to feed the photochemical reactions. In these reactions, GEM is converted to RGM during AMDEs, and RGM either associates with aerosols (as PHg) and/or is deposited onto surfaces—primarily snow and ice during polar spring when these reactions occur. The fate of this deposited Hg is less clear. Studies have concluded that some of the deposited RGM is reduced to GEM and re-emitted into the atmosphere while some remains or is re-oxidized at the surface of, or within, the snowpack. Studies of the photochemistry of Hg within the snowpack have shown that reactions occur within the first few centimetres of the snow surface, and flux measurements indicate that there is net deposition of Hg to the snow in early spring followed by a net emission of Hg from the snowpack in the summer (Lalonde et al. 2002, Dommergue et al. 2003, Poulain et al. 2004, Ferrari et al. 2005, Constant et al. 2007, Dommergue et al. 2007). Global and regional models now incorporate AMDEs into their chemical mechanisms although there is still an incomplete understanding of the chemical processes that drive deposition and re-emission of Hg in this environment.

3.2.2 Chemical reactions of Hg in the Arctic atmosphere

At the end of NCP Phase II, only indirect evidence was available for the oxidation of Hg from long-lived Hg(0) to easily deposited Hg(I) and Hg(II) by halogen atoms, particularly bromine. This evidence was based on a strong correlation between AMDEs and ozone depletion events (ODEs), which were believed to be caused by catalytic loss of ozone through reaction with halogen atoms and showed a negative correlation with concentrations of bromine oxide—one of the components of the catalytic ozone loss cycle—in the boundary layer and total air column (Simpson et al. 2007).

Direct evidence for the oxidation of Hg in the gas phase has more recently been gathered through laboratory measurements (Ariya et al. 2002, Raofie and Ariya 2003, Pal and Ariya 2004, Donohoue et al. 2006) and theoretical calculations (Khalizov et al. 2003, Goodsite et al. 2004, Shepler et al. 2005) of reaction rates and reaction products. These findings are reviewed in more detail elsewhere (Ariya et al. 2008, Ariya et al. 2009) but a summary of selected findings is presented in Table 3.1. This table illustrates how the importance of each oxidation reaction depends on both the oxidant concentration and the rate coefficient for a given reaction. For example, the reaction of Hg(0) with Br is slower than the reaction with Cl by a factor of 2–25 but since the concentration of Br during *bromine explosions* can be approximately 100–2,000 times higher than that of Cl (Cavender et al. 2008), the reaction with Br is considered more important. In fact, models of atmospheric Hg chemistry suggest that oxidation by bromine radicals alone can account for the rapid oxidation of GEM that occurs during AMDEs when GEM concentrations can sometimes drop by a factor of ten in a matter of hours (Ariya et al. 2004, Goodsite et al. 2004, Skov et al. 2004, Holmes et al. 2006, Holmes et al. 2010). However, there is evidence to suggest that Cl concentrations may be significantly higher during some AMDEs and could significantly contribute to observed GEM oxidation (Stephens et al. 2012). It is also clear from Table 3.1 that there is a great deal of uncertainty associated with many of the rate coefficients. Some discrepancy between different experiments has been attributed to reactions on the walls of reaction chambers, suggesting not only that the gas phase reaction rates may be inaccurate but also that additional reactions on the surface of particles, snow, ice, and ocean water may contribute to GEM oxidation in the polar atmosphere (Sabir et al. 2011). Additionally, there are limited data for the temperature dependence of these reactions (Goodsite et al. 2004, Donohoue et al. 2005, Donohoue et al. 2006).

Regardless of their source, these large uncertainties in reaction rates must be narrowed in order to accurately model Hg oxidation during AMDEs and throughout the year.

TABLE 3.1 A summary of selected experimental and theoretical reaction rate coefficients for the oxidation of GEM, along with estimated GEM lifetime based on estimated oxidant concentrations (Ariya et al. 2008, Ariya et al. 2009). Lifetime corresponds to the time required to oxidize 67% of initial Hg(0) with each oxidant.

Oxidant	Estimated Concentration in Polar Spring (molecules cm^{-3})	Rate Coefficient for Reaction with GEM (cm^3 $molecules^{-1}$ s^{-1})	GEM Chemical Lifetime with This Oxidant
Br	10^7–10^8	4×10^{-13}–3×10^{-12}	< 1 hr to 3 days
Cl	10^4	5×10^{-13}–1×10^{-11}	115 days to 5.9 years
BrO	$< 5 \times 10^8$	10^{-15}–10^{-13}	> 5 hrs to > 23 days
OH	10^5–10^6	9×10^{-14}–3×10^{-13}	36 days to 3.6 years
O_3	$< 10^{12}$	10^{-20}–10^{-18}	11 days to 4 years

The reduction of Hg(I) and Hg(II) back to Hg(0) is another important component of the atmospheric Hg cycle in the Arctic. Reduction is generally assumed to take place in the aqueous phase, since Hg(I) and Hg(II) species are soluble, and reduction results in volatile Hg(0) that is easily released to the atmosphere. This process occurs in lakes and oceans, as discussed in chapters 5 and 6. However, Hg reduction also occurs in snowpack and potentially in cloud and rain droplets, and in particles with significant water content. Emission of GEM from snow has been observed following AMDEs (section 3.3) and appears to depend on exposure to ultraviolet (UV-B) radiation (Lalonde et al. 2003, Poulain et al. 2004). Little is known about the reactants involved in reduction reactions or their rates. For example, rate constants for the reduction of Hg(II) ions by hydroperoxyl (HO_2) radicals and for the decomposition of the mercuric sulphite ($HgSO_3$) complex to Hg(0)—both in water—have been measured but are still highly uncertain (Sabir et al. 2011, and references therein). Photoreduction of Hg on surfaces, rather than within the water itself, has not been studied (Sabir et al. 2012). It is a difficult research question to investigate because oxidation of Hg(0) back to Hg(I) or Hg(II) can occur at the same time and may be enhanced by the presence of water (Shepler et al. 2007) and Cl or Br ions in the water (Sheu and Mason 2004).

Given the uncertainty in both reduction and oxidation chemistry on environmental surfaces, heterogeneous (i.e. air-surface) reactions of Hg are a major knowledge gap that currently limits our understanding of factors that drive Hg fluxes in the Arctic environment. Finally, although Ariya et al. (2002) did identify mercuric chloride ($HgCl_2$) and mercuric bromide ($HgBr_2$) as products of the Hg+Cl and Hg+Br reactions in the laboratory, the actual products of Hg reactions in the ambient environment have not yet been characterized.

3.2.3 Temporal trends of atmospheric Hg depletion events at Alert

Patterns in Hg speciation data collected over several years provide insight into the atmospheric processes that occur during AMDEs. Datasets reporting the AMDE phenomenon have been gathered for varying periods of time at several Arctic stations, including Alert (Nunavut), Barrow (Alaska), Amderma (Russia), Station Nord (Greenland), and Ny-Ålesund (Svalbard) (Steffen et al. 2008) and in the sub-Arctic at Kuujjuarapik (Quebec) (Poissant and Pilote 2003, Gauchard et al. 2005, Steffen et al. 2005). Analysis of the Alert dataset from 1995 to 2007 revealed no significant time trend in the overall combined frequency and strength of depletion events over the AMDE season (Cole and Steffen 2010). However, it did reveal significant changes with respect to when these AMDEs occur. Over time, the month of maximum AMDE activity has shifted from May to April, with increased AMDE activity in March as well. This observation is illustrated in Fig. 3.1, which shows the average distribution of AMDE occurrence in each of the spring months for 1995 to 2002 and for 2003 to 2009. The reason for the shift in timing of depletion events is not yet understood, though it has been found that the frequency of depletion events and overall GEM concentrations exhibit complex relationships with local meteorology (Fig. 3.2). The top panel of Fig. 3.2 shows that the integrated frequency of depletion events—a quantity that combines the length, magnitude, and frequency of depletion events—is lower at higher temperatures within each month. This pattern is consistent with previous findings that the halogen chemistry initiating AMDEs and ODEs may be temperature-dependent (Koop et al. 2000, Adams et al. 2002, Tarasick and Bottenheim 2002, Sander et al. 2006). The frequency of AMDEs was not related to local wind speed but was found to have a relationship with wind direction (Fig. 3.2, bottom panel). The fact that air masses originating from over the Arctic Ocean in the spring are depleted of GEM more frequently than those originating elsewhere suggests that AMDEs occur over the ocean. Finally, it was found that March GEM concentrations at Alert correlated with the Polar/Eurasian Teleconnection and North Atlantic Oscillation indices. This association suggests that variability in transport of Hg from lower latitudes during the winter explained some of the interannual variability in AMDE activity in the early spring. However, there was no temporal trend in that index, nor did the correlation extend into April and May (the primary AMDE season).

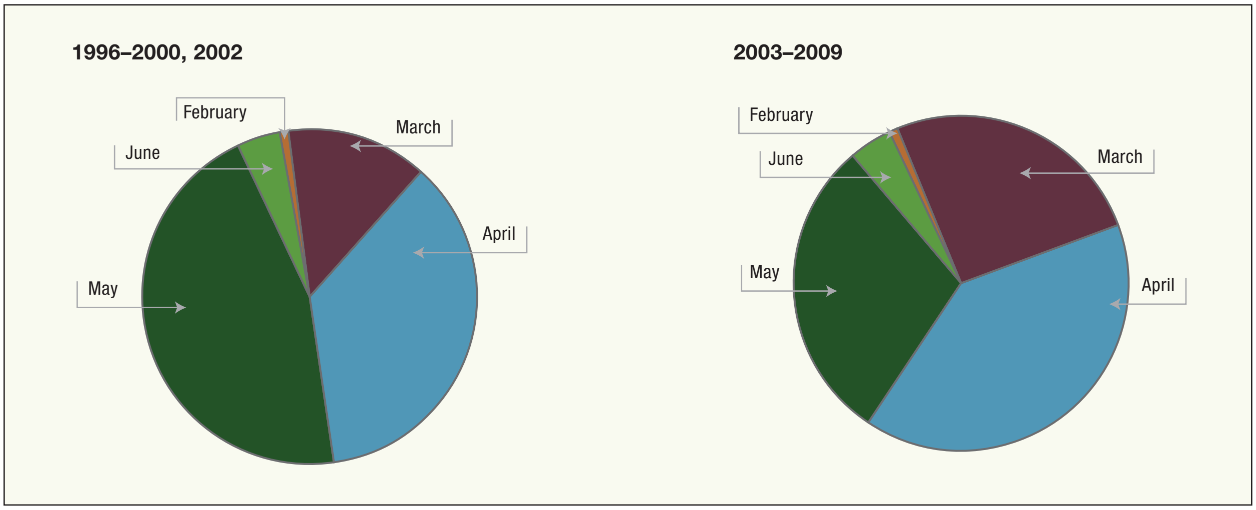

FIGURE 3.1

Average distribution of AMDEs in each of the spring months at Alert (Nunavut), for the years (a) 1996 to 2000 and 2002, and (b) 2003 to 2009. Correcting data for different sampling rates resulted in changes of < 1% (Cole and Steffen 2011).

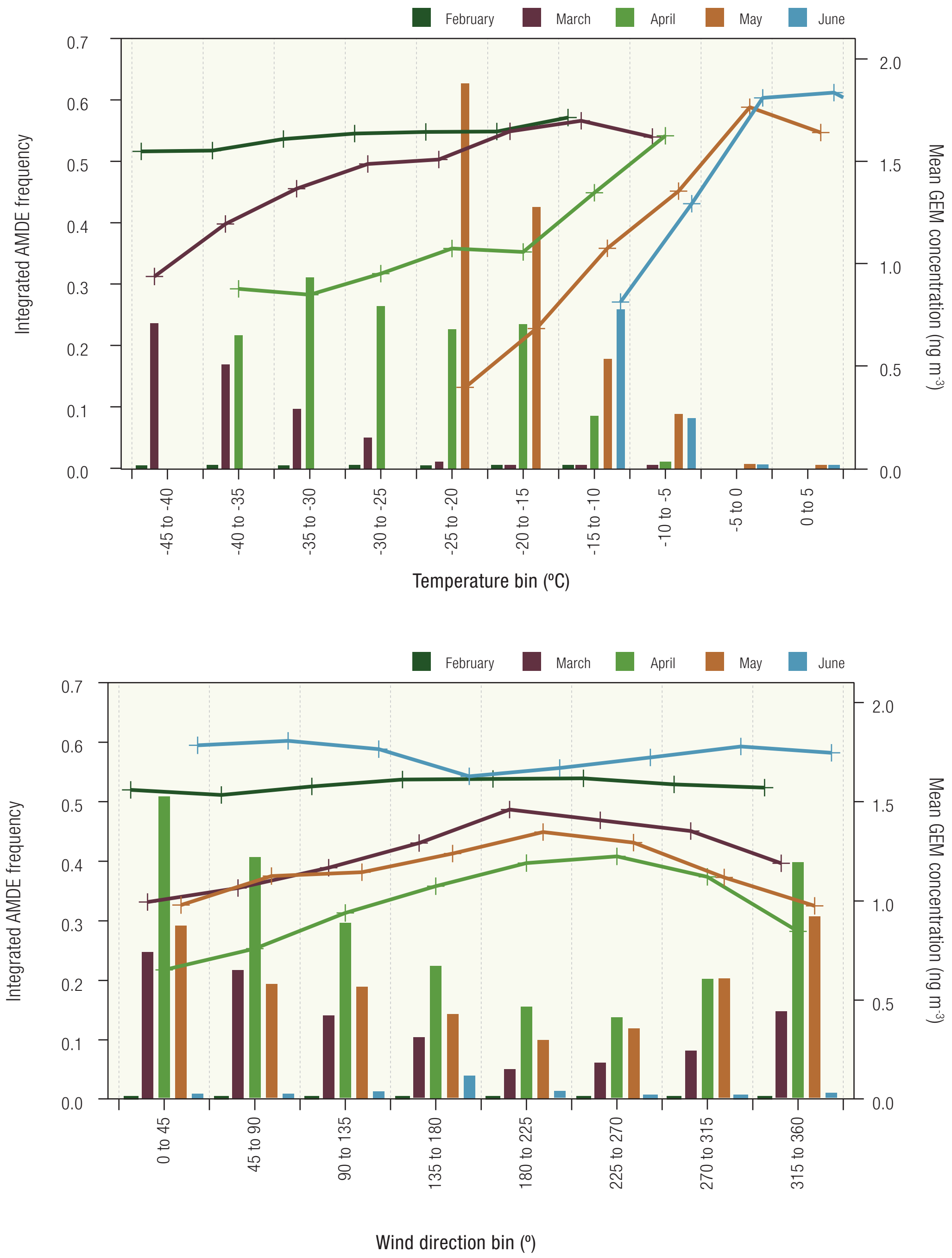

FIGURE 3.2

Frequency of AMDEs (bars) and mean GEM concentrations (lines) at Alert for spring months binned by local temperature and wind direction (source: Cole and Steffen 2010).

3.2.4 Distribution of atmospheric Hg species in the Arctic

The distribution of atmospheric Hg species differs regionally in the Arctic, as shown by a time series of GEM, RGM, and PHg from 2002 to 2009 at Alert (Fig. 3.3) and a time series of GEM, RGM, and PHg measurements from several months in 2004 at Churchill in sub-Arctic Manitoba (Fig. 3.4). The eight years of data from Alert are also summarized in Fig. 3.5, which shows the seasonal cycles of GEM, RGM, and PHg. These figures clearly demonstrate the periodicity of AMDEs in springtime and the significant changes in Hg speciation that occur during these events.

Background atmospheric Hg speciation in low- and mid-latitude rural areas is dominated by GEM which represents about 95–98% of total Hg (THg), with approximately 0.2–1% RGM and 0.2–2% PHg making up the balance (Schroeder and Munthe 1998, Peterson et al. 2009, Rutter et al. 2009). These species emerge from different sources than in the Arctic. The distribution of Hg species in urban areas is reported to be 79–99% GEM, 0.4–13% RGM, and 0.4–8% PHg (Rutter et al. 2009). Cobbett et al. (2007) reported that during the 2005 polar night at Alert, the distribution of Hg species was similar to that of southern rural latitudes (i.e. 95% GEM, 0.63% RGM, and 4.4% PHg). In contrast, during AMDEs, the Arctic springtime atmosphere changed to 88.6%, 5.7%, and 5.7% of GEM, RGM, and PHg, respectively. At Churchill in 2004, the change in distribution was even more dramatic, with GEM, RGM, and PHg distributed on average over three non-AMDE months as 99.2%, 0.5%, and 0.3%, respectively. However, during the AMDE period of March to May, it changed to 71.3%, 10.3%, and 18.3% on average. Although the distribution of atmospheric species during AMDEs is similar to that reported in an urban environment, it should be noted that the high spring concentrations of RGM and PHg in the Arctic are due to oxidation of GEM in the atmosphere, while high levels in urban areas reflect direct industrial emissions of RGM and PHg. Furthermore, since RGM and PHg have a higher deposition velocity than GEM, the springtime shift to high concentrations of short-lived Hg species warrants

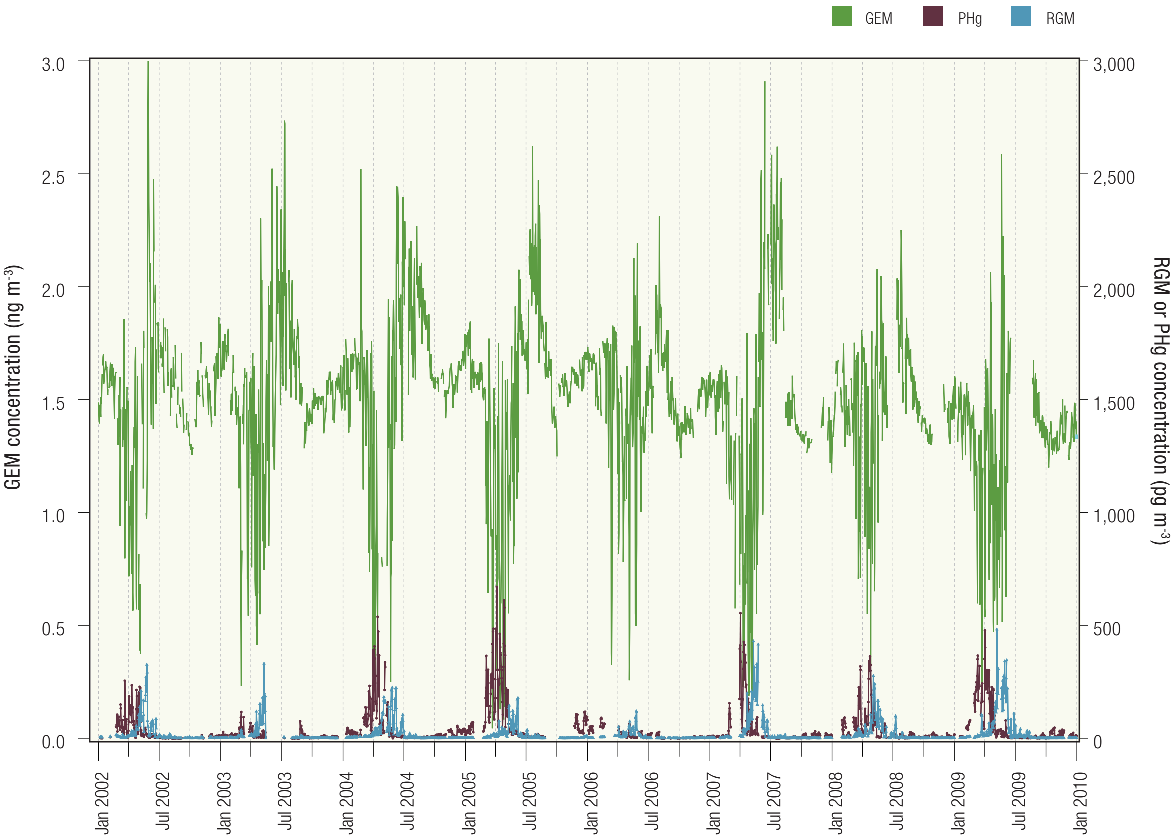

FIGURE 3.3

Daily average GEM, PHg, and RGM concentrations at Alert (Nunavut) from 2002 to 2009. Left and right scales are equivalent (3000 pg = 3 ng) (Steffen 2011).

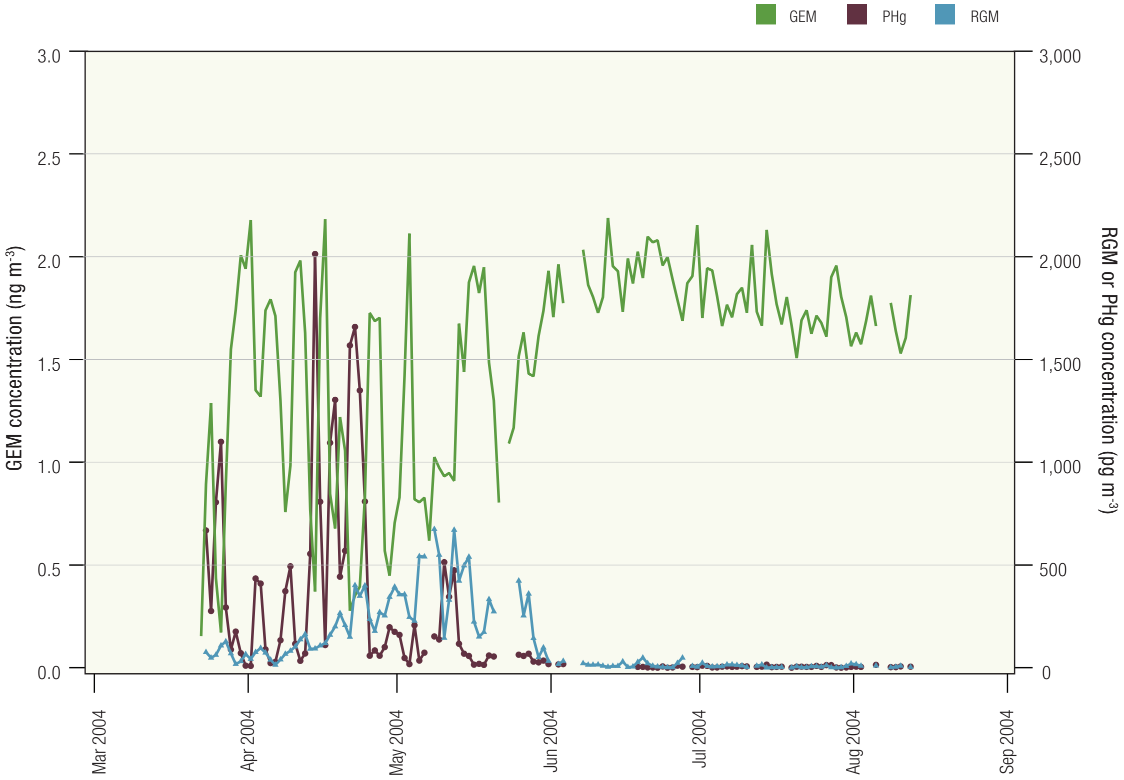

FIGURE 3.4

Daily average GEM, PHg, and RGM concentrations at Churchill (Manitoba), March to August 2004 (data from Kirk et al. 2006). Left and right scales are equivalent (3,000 pg = 3 ng).

investigation to determine the magnitude of springtime atmospheric deposition of Hg to the Arctic environment.

The distribution of GEM, RGM, and PHg during AMDEs differs among Arctic measurement sites and the time in spring. For example, at Barrow in the 2001 campaign, RGM was the dominant species during AMDEs (Lindberg et al. 2002). In Ny-Ålesund in 2003, the distribution was reported to vary throughout the study period but showed a predominance of RGM (Sprovieri et al. 2002, Gauchard et al. 2005), whereas during a more recent study at Ny-Ålesund, the distribution also varied but showed a predominance of PHg (Steen et al. 2009). Some researchers have suggested that the predominance of either PHg or RGM can be an indication of the origin of the air mass (Lindberg et al. 2002, Gauchard et al. 2005) but perhaps the timing in which the sampling occurred more accurately reflects the distribution of these species. At Churchill and Alert, Kirk et al. (2006) and Cobbett et al. (2007), respectively, showed a predominance of PHg at the beginning of spring and a higher predominance of RGM towards the end of spring (Figs. 3.3 and 3.5).

3.3 Deposition of atmospheric Hg

The atmospheric processes involved in the deposition of Hg during AMDEs are not well understood at this time (Cole and Steffen 2010). Cobbett et al. (2007) suggested that a relationship exists between the stability of the atmosphere and the distribution of Hg species during AMDEs, and they reported a significant correlation between increases in PHg and lower air temperatures and humidity. Steffen et al. (2002) reported that not all Hg is lost from the air during AMDEs. Although GEM may be depleted in the air, depending on the wind conditions there may remain some RGM and PHg in the air mass which can be available for transport to another location. This implies that not all depleted GEM is deposited onto the snowpack during an AMDE. More recent work at Alert shows that the deposition of Hg from the air during

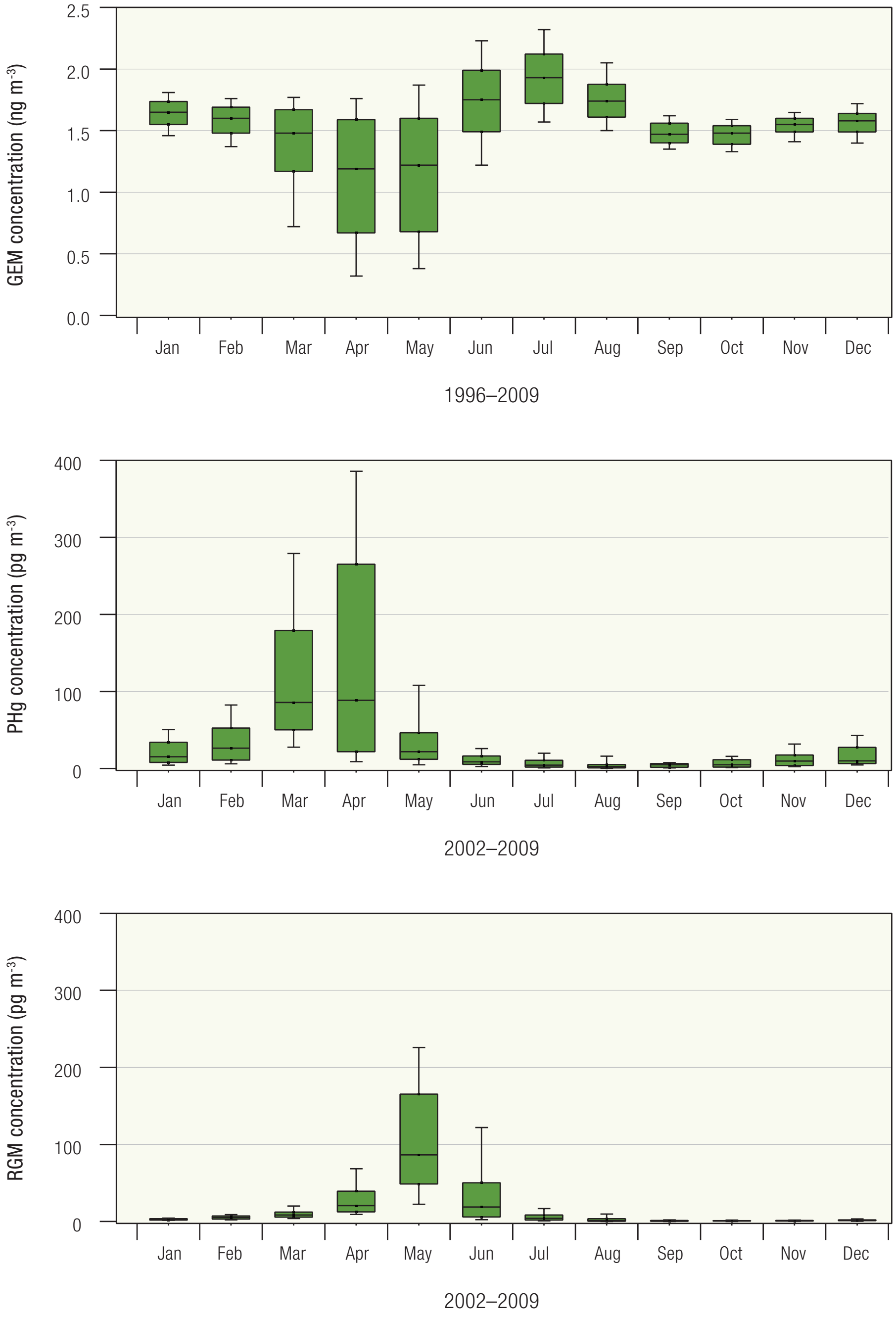

FIGURE 3.5

Seasonal patterns of GEM, PHg, and RGM for Alert (Nunavut) from 2002 to 2009 (Steffen 2011).

AMDEs is dependent on the conditions of the atmosphere such as relative humidity and aerosol contribution (Steffen 2006). Thus, in order to accurately understand the wet and dry deposition processes of Hg from air to snow that result from AMDEs, prevailing atmospheric conditions must be fully characterized. Wet deposition is defined as the scavenging of gaseous and particulate Hg species into atmospheric precipitation while dry deposition is defined as direct deposition to the surface.

Currently, deposition of Hg during AMDEs is typically inferred from concentrations of THg in the snowpack and not primarily through direct measurements of dry or wet deposition. At Alert, snow/ice crystal samples have been collected on a Teflon snow table on an event basis during springtime since 2002 and are likely the closest direct (uninfluenced) measurements of snow that have been collected over time in the Arctic (i.e. scavenging of PHg and RGM by snow). Data from this monitoring setup show that depletion events are often accompanied by an increase in THg concentration in the fallen snow. Additionally, the time when higher levels of THg are found in the snow repeats from year to year. It has been observed in the long-term dataset that the most significant increase of THg in the snow generally occurs when there is a switch from PHg predominance to RGM predominance in the atmosphere (Fig. 3.6). These results show that the highest deposition of Hg through snowfall does not occur throughout the springtime period but rather when the atmosphere is characterized by certain conditions (Steffen 2006). This finding can help explain a report from Ny-Ålesund which found that there were times when no increase in the concentration of THg in the snowpack was observed following AMDEs (Ferrari et al. 2005). Other direct wet deposition measurements—reported as fluxes—were reported by Sanei et al. (2010), who employed a modified Mercury Deposition Network (MDN) precipitation collector in Churchill for over 27 months between 2006 and 2008 (Sanei et al. 2010). Contrary to other observations at Churchill and elsewhere in the Arctic, Sanei et al. (2010) reported that there is little evidence of higher wet deposition of Hg in the springtime during AMDEs. While atmospheric models predict that there should be deposition of Hg during the springtime period at this location, the

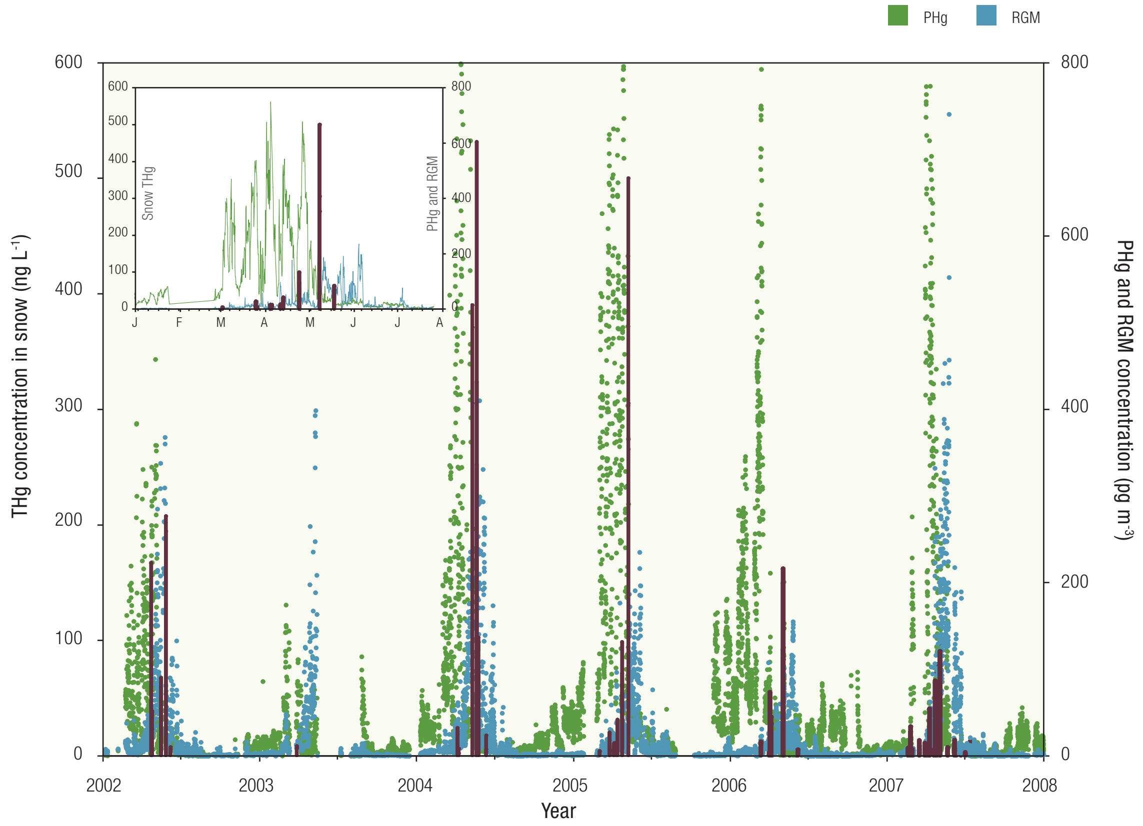

FIGURE 3.6

Atmospheric Hg speciation data (PHg in green, RGM in blue) at Alert (Nunavut) from 2002 to 2007 and THg concentrations in snow (dark purple bars). The insert presents data from January to August 2004 (Steffen 2011).

measurements did not reflect those predictions. Since this finding contradicts what others have reported, further investigation into this discrepancy is warranted.

Other research has examined the deposition of Hg during or after depletion events by measuring the concentration of THg in the snowpack (Poissant et al. 2008, Steffen et al. 2008). Several researchers have reported that when the concentration of THg in the snowpack is elevated following an AMDE, there can be a 50–90% loss of that Hg within a few days (Kirk et al. 2006, Constant et al. 2007), and they have suggested that this loss is a result of photochemical reduction of Hg(II) to GEM and emission to the atmosphere (Lalonde et al. 2003, Poulain et al. 2004, Constant et al. 2007, Outridge et al. 2008, Sherman et al. 2010). A few researchers have measured the atmospheric fluxes of Hg above the Arctic snowpack to quantify the deposition and emission of GEM around AMDEs. In 2000, vertical gradients of GEM concentrations were measured at Alert above the snow surface during springtime. These measurements showed higher concentrations of GEM immediately above the snowpack, thereby suggesting an emission from the surface (Steffen et al. 2002). This evasion was observed for several days and throughout an extended AMDE during which GEM appeared to be emitted from the snow surface and then underwent photooxidation, which depleted it from the air again. In 2005 at Alert, Cobbett et al. (2007) reported a net depositional flux of GEM to the snow during the polar night—between January and March when no AMDEs occur—but, during the spring, there was no *net* GEM deposition to the snow surface and no evidence of *net* emission of GEM following AMDEs (Cobbett et al. 2007). They found significant cycling of Hg between the surface and the atmosphere but an overall net flux of zero was reported throughout the entire season in 2005. A flux chamber study in Barrow reported a total emission of GEM from the surface of 4–7% of the THg content in the snow within a 24-hour period and the authors suggested this was an upper limit to the photoreduction of easily reduced Hg after AMDEs (Johnson et al. 2008). Most recently, Norwegian researchers in Ny-Ålesund reported the same findings of net deposition of GEM in the polar night but found there to be a positive net emission in the spring (Steen et al. 2009). It should be noted that these measurements of GEM flux do not account for fluxes of RGM or PHg, which are the fractions thought to deposit more readily to the surface during AMDEs. Deposition of RGM and PHg, which are unlikely to be emitted from the surface, would increase the overall net deposition of Hg to the surface during the AMDE season.

The aforementioned discrepancies in reports of Hg deposition to the snowpack in the Arctic have led researchers to create a snow exchange model that includes atmospheric deposition (Fig. 3.7). To develop a model parameterization for the fate of deposited Hg in the Arctic, Durnford and Dastoor (2011) analysed the basic mechanisms of the air-snow exchange of Hg using measurements in snow from around the world. Based on field and laboratory data and model results, they presented a conceptual mechanism of the physical and chemical processes governing the fate of deposited Hg to the snow as illustrated in Fig. 3.7. It is hypothesized that all GEM deposited onto snow-covered surfaces is likely revolatilized immediately—as indicated in the top left-hand corner of Fig. 3.7. However, the snowpack likely retains most, if not all, of the deposited PHg. The fate of RGM deposited onto the snowpack is far more complicated because it may undergo a series of chemical reactions within the snowpack. For instance, RGM can be reduced to GEM (section 3.2.2) which may then be oxidized back to RGM again. Oxidation of GEM tends to dominate over reduction of RGM in daylight. Further, hydrogen peroxide acts as both a reductant (in pH neutral snow) and an oxidant (in acidic snow), and the presence of halides in the snowpack is found to stabilize the oxidized Hg in the snowpack. Emitted GEM is sourced only from approximately the top 2 cm of the snowpack, and GEM emission to the atmosphere increases significantly at the onset of snowmelt. Finally, at the onset of snowmelt, a considerable fraction of the snowpack's burden of oxidized Hg exits the snowpack in the subsequent pulse of ionic melt water (Bales et al. 1990, Kuhn 2001). These physical and chemical processes of Hg in the cryosphere vary with differing environmental conditions. For instance, at coastal marine sites, deposition is known to follow AMDEs. Since halogen species are present at higher concentrations around these snowpacks, retention of the deposited Hg may be enhanced. Also, THg concentrations measured in snow under tree canopies are higher than those in adjacent open areas (Poulain et al. 2007). Canopies can reduce revolatilization significantly by lowering the photoreduction of RGM in snow, increasing the fraction of Hg deposited as PHg and the oxidant content of the snowpack, and dampening wind ventilation of snowpack that promotes Hg emission.

According to model results, the concentration of THg in seasonal snowpacks is strongly affected by the burial of Hg with fresh snow—the concentration increases with increasing snow precipitation and its frequency. In contrast, deeper snowpacks apparently dilute the

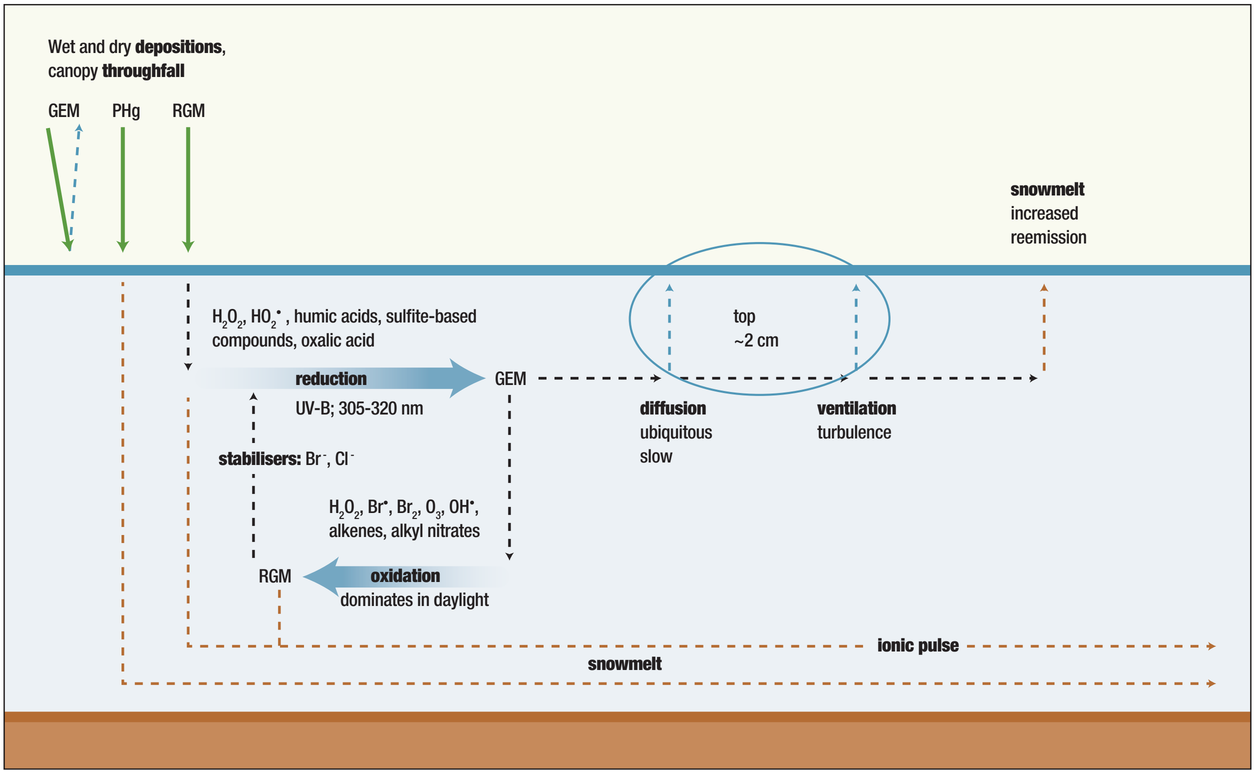

FIGURE 3.7

A schematic of the physical and chemical processes that govern the behaviour of cryospheric Hg (source: Durnford and Dastoor 2011).

concentration of THg in the ionic pulse of melt water from the snowpack. The concentration of THg in long-term cryospheric records varies by latitude possibly due to mid-latitude anthropogenic sources of Hg. The fate of long-term cryospheric Hg may also be governed by snowpack ventilation and the burial of Hg by fresh snowfalls.

Carignan and Sonke (2010) examined Hg deposition along the coast of Hudson Bay. They found that, not only is the snow surface subject to deposition of Hg from AMDEs, but the lichens on trees around Hudson Bay also accumulate this atmospherically deposited Hg. Although this vegetation is known to be a good indicator of atmospheric deposition of Hg (Bargagli and Barghigiani 1991, Horvat et al. 2000, Riget et al. 2000), Hg retention by lichens has only been investigated at a few locations in the Canadian Arctic (section 4.7). In the study by Carignan and Sonke, the THg concentration in lichens decreased with distance from Hudson Bay and showed the same pattern as snow THg concentrations in the area (Constant et al. 2007). These findings, along with those of Douglas and Sturm (2004), show that the concentration of THg in the snow decreases with distance inland from the ocean and indicate that Hg deposition is greater closer to ocean water (see also section 3.4.1).

While much study has concentrated on the exchange of Hg between air and snow on land, there have been few investigations into the cycling of Hg over the sea ice. Elevated levels of THg in frost flowers, hoar frost, sea snow, and seawater over the Arctic Ocean have been reported (Douglas and Sturm 2004, Douglas et al. 2005). A recent publication of measurements over the Beaufort Sea showed a comparison of two snow-covered ice cores and suggested that elevated THg levels in surface snow, presumably due to atmospheric deposition, do not extend down to the sea ice until melting of the snow has begun (Chaulk et al. 2011). More recent studies during International Polar Year (IPY) have occurred over sea ice but the results were not yet available for inclusion in this report.

Modelling results by Hirdman et al. (2009) provide additional support for the hypothesis that AMDEs result in the net deposition of Hg from the atmosphere to the Arctic Ocean. The authors performed a statistical analysis on the results from a Lagrangian particle dispersion model (FLEXPART) and GEM concentrations measured at Ny-Ålesund to identify source regions of high- and low-Hg air masses. It was found that, in the spring, air masses depleted in Hg had undergone low-level transport over the ocean. However, in the summer, air masses with strong surface contact over the Arctic Ocean showed a high Hg concentration. This suggests that the Arctic Ocean is a net sink for Hg in the springtime but a net source in the summer, possibly due to emissions from the snow surface and evasion from the ocean (Hirdman et al. 2009). This conclusion is similar to the findings of Ariya et al. (2004). Hirdman et al. (2009) also found that in the winter, low-Hg concentrations were associated with air masses from the free troposphere. This model analysis has not been replicated with data from the Canadian Arctic but the findings are in agreement with data reported from Alert for both Hg and ozone (Bottenheim and Chan 2006, Cole and Steffen 2010).

The overall impact of atmospheric Hg deposition on the Arctic remains unclear, not only in Canada but across the circumpolar Arctic. A mass balance budget was produced for THg in the Arctic Ocean, and it was concluded that the contribution of the atmosphere to the Hg pool in the Arctic Ocean is less than what is derived for other oceans but is still significant overall (Outridge et al. 2008). Using data collected at Barrow (Alaska), a mass balance budget was developed for the Arctic that showed a net surface gain in Hg in the springtime (Brooks et al. 2006). Other estimates of net deposition of Hg to the Arctic, including deposition from AMDEs, range from 50–300 tonnes (t) of Hg per year (Lu et al. 2001, Banic et al. 2003, Ariya et al. 2004, Skov et al. 2004). Results from these investigations are discussed in more detail in section 3.6.2. It is also clear from this work that there is a strong cycling of Hg at the snow-air interface and within the snowpack and melt water but the processes governing this cycling are not well understood. Some of these issues for marine snow are addressed in more depth in section 6.4 of the marine environment chapter.

3.4 Summary of Canadian atmospheric measurement data and spatial patterns

Air GEM concentrations have been monitored in Canada at approximately ten different sites since 1995. Of the ten sites, GEM has been continuously measured at only three sites between 1995 and the present. In the Canadian Arctic and sub-Arctic, the spatial coverage is even sparser. Alert, Kuujjuarapik, and Little Fox Lake (Yukon) are the only Arctic sites where continuous atmospheric GEM measurements are made at present—measurements at Little Fox Lake began in 2007.

Alexandra Steffen

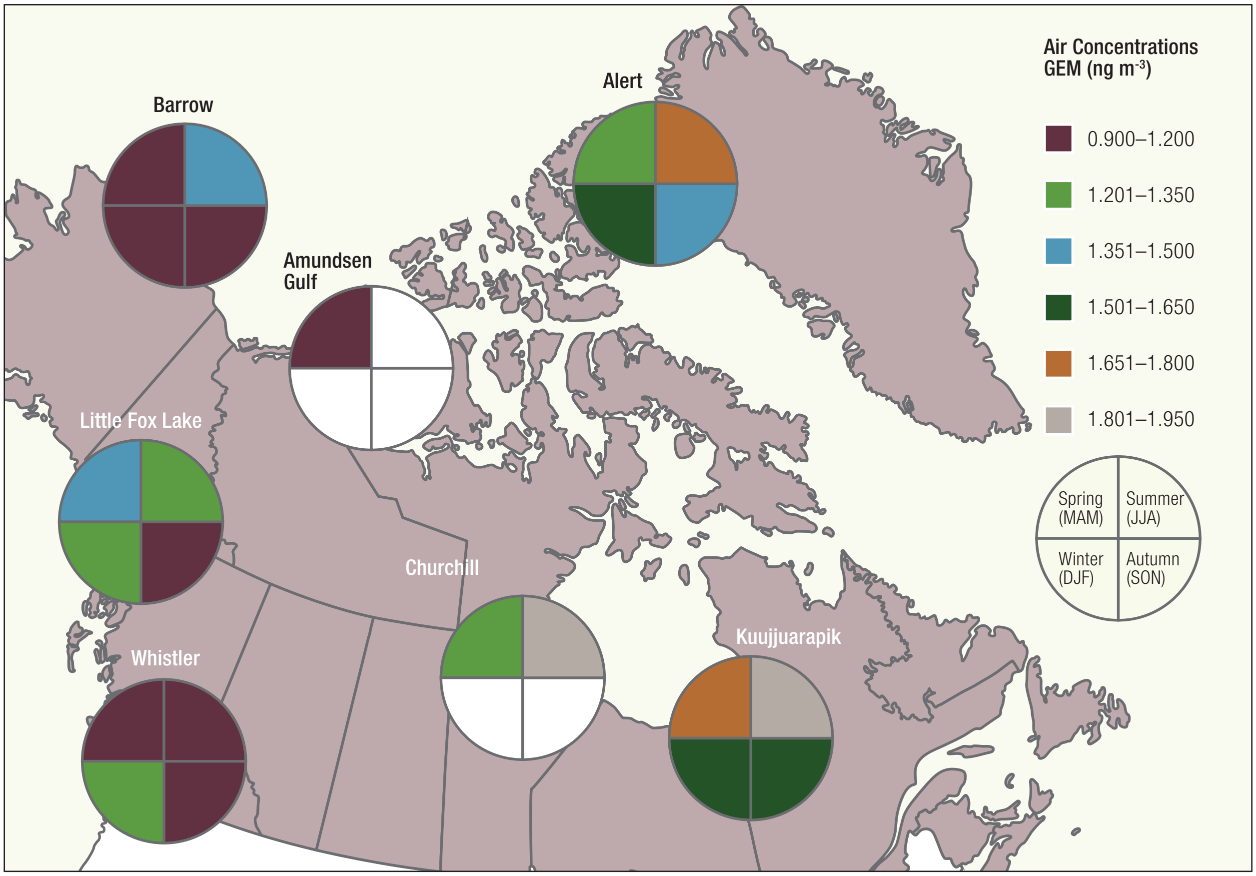

FIGURE 3.8

Overview of seasonal air concentrations of GEM at sites across Canada and Alaska. Data are from Steffen (2011), Pilote (2011), and Kirk et al. (2006).

Supplementary data from short campaigns (< 1 year) are available for Resolute (Nunavut) (Lahoutifard et al. 2005), Churchill (Kirk et al. 2006), and the Amundsen Gulf (Northwest Territories). Continuous monitoring data for speciated Hg (i.e. GEM, RGM, and PHg concurrently) are even more limited. Alert is the only site where speciated measurements have been collected over multiple years. Speciated Hg monitoring has been done for short periods at Kuujjuarapik (Steffen et al. 2003), Churchill (Kirk et al. 2006), on Hudson Bay (Poissant et al. 2006) and around the Amundsen Gulf during the IPY.

3.4.1 Gaseous elemental Hg

Geographic variation was observed in seasonal mean concentrations of GEM from Alert and the Amundsen Gulf in the High Arctic; Little Fox Lake, Churchill, and Kuujjuarapik in the sub-Arctic; and, for comparison, at the Alaskan Arctic site of Barrow and the high altitude temperate site of Whistler (British Columbia) (Fig. 3.8). This map reflects the available data of atmospheric Hg monitoring in the Canadian Arctic and, at the same time, demonstrates the lack of existing spatial coverage. Mean values for each season (winter: December to February, spring: March to May, summer: June to August, and autumn: September to November) and for the entire dataset were calculated over different time periods as summarized in Table 3.2. These measurement data reveal that annual concentrations of GEM are lower at the western sites of Barrow, Little Fox Lake, and Whistler—overall median concentrations of 1.13, 1.31 and 1.14 ng m^{-3}, respectively—compared to eastern sites at Alert and Kuujjuarapik—1.56 and 1.63 ng m^{-3}, respectively (Table 3.2). The annual concentrations for Churchill and the Amundsen Gulf are absent because there are only six months and three months of monitoring data available, respectively, and thus, they cannot be directly compared with annual data from the other sites. There appears to be a concentration difference in GEM with longitude from west to east (Fig. 3.8). In order to

TABLE 3.2 Multi-annual medians and seasonal averages of GEM (ng m^{-3}), RGM (pg m^{-3}) and PHg (pg m^{-3}) concentrations across Canada and Alaska. Data are from Steffen (2011), Pilote (2011), and Kirk et al. (2006). *Note: Although Barrow is not a Canadian site, the data included in this report were collected as a Canada–US collaboration under IPY with Canadian IPY funding.*

Site (measurement period)	Multi-Annual Median GEM	Spring GEM Mean (SD)	Summer GEM Mean (SD)	Fall GEM Mean (SD)	Winter GEM Mean (SD)	Spring RGM Mean (SD)	Spring PHg Mean (SD)	GEM Trend (ng m^{-3} y^{-1})
Alert (1995 to 2009) 82.5° N, 62.5° W	1.56	1.24 (0.53)	1.80 (0.35)	1.49 (0.11)	1.59 (0.17)	50 (72)	70 (65)	-0.009
Barrow (2007 to 2009) 71.3° N, 156.6° W	1.13	1.06 (0.69)	1.40 (0.27)	1.06 (0.12)	1.03 (0.26)			N/A
Churchill (March–August 2004) 58.6° N, 94.2° W	N/A	1.26 (0.61)	1.81 (0.19)	N/A	N/A	213 (199)	342 (495)	N/A
Kuujjuarapik (1999 to 2009) 55.3° N, 77.7° W	1.63	1.72 (0.64)	1.86 (0.58)	1.58 (0.46)	1.56 (0.43)			-0.038
Little Fox Lake (2007 to 2009) 61.3° N, 135.6° W	1.31	1.43 (0.11)	1.27 (0.12)	1.17 (0.14)	1.35 (0.14)			N/A
Whistler (2008 to 2009) 50.1° N, 122.9° W	1.14	1.14 (0.09)	1.17 (0.17)	1.20 (0.25)	1.21 (0.21)			N/A
Amundsen Gulf (March–May 2008) 71.1° N, 124.8° W	N/A	0.94 (0.58)	N/A	N/A	N/A			N/A

TABLE 3.3 Slopes of linear regressions (with 95% confidence limits) for mean annual and seasonal GEM concentrations at sites in Fig. 3.8 in relation to site longitude (Steffen 2011). NS = not significant. No significant trends with latitude were found.

GEM Concentration	Change in GEM Concentration in Relation to Site Longitude (ng m^{-3} degree^{-1})*
Annual mean	0.0053 ± 0.0049
Spring (MAM) mean	NS
Summer (JJA) mean	0.0070 ± 0.0069
Fall (SON) mean	0.0052 ± 0.0025
Winter (DJF) mean	0.0057 ± 0.0041

*A positive slope reflects an increasing trend from west to east.

assess whether this apparent trend is statistically significant, simple linear regressions were performed between the mean seasonal (and annual) GEM concentrations and site latitude or longitude. There was a significant positive relationship between GEM concentration and longitude from west to east for the annual, summer, fall, and winter means (Table 3.3). This relationship was not significant for the spring period likely because of the chemistry that dominates the springtime at Alert, Churchill, and Kuujjuarapik. No significant relationship between GEM concentration and latitude was observed. It should be noted, however, that these trends are based on only five measurement locations with very different temporal coverage and should be treated as being preliminary.

Geographic differences were also observed in the seasonal patterns of GEM among these sites. At Alert, Kuujjuarapik, and Barrow, there were several days in the spring where the GEM concentration dropped rapidly to below 1 ng m^{-3}, typically used as the definition of an AMDE (Fig. 3.9). These AMDEs occurred at Alert and Barrow primarily from March to May, at Kuujjuarapik from February to April, and were not observed at Little Fox Lake or Whistler. Data from one spring and summer at Churchill also showed AMDEs (Fig. 3.4). Those sites with AMDE activity are Arctic and sub-Arctic sites that are in relatively close proximity to sea ice. The sub-Arctic site at Little Fox Lake does not experience AMDEs, which supports the hypothesis that these events occur over or near a maritime source of sea salt (Douglas and Sturm 2004, Douglas et al. 2005).

Sites with AMDEs also experienced rapid increases in GEM concentrations in the late spring and summer that reached average daily concentrations of > 5 ng m^{-3} at Kuujjuarapik (Fig. 3.9). As a result, GEM concentrations peaked in summer at these four sites as shown in Table 3.2. It should be noted that the data from Churchill are for spring and summer only and cannot be compared with fall averages. In addition, while

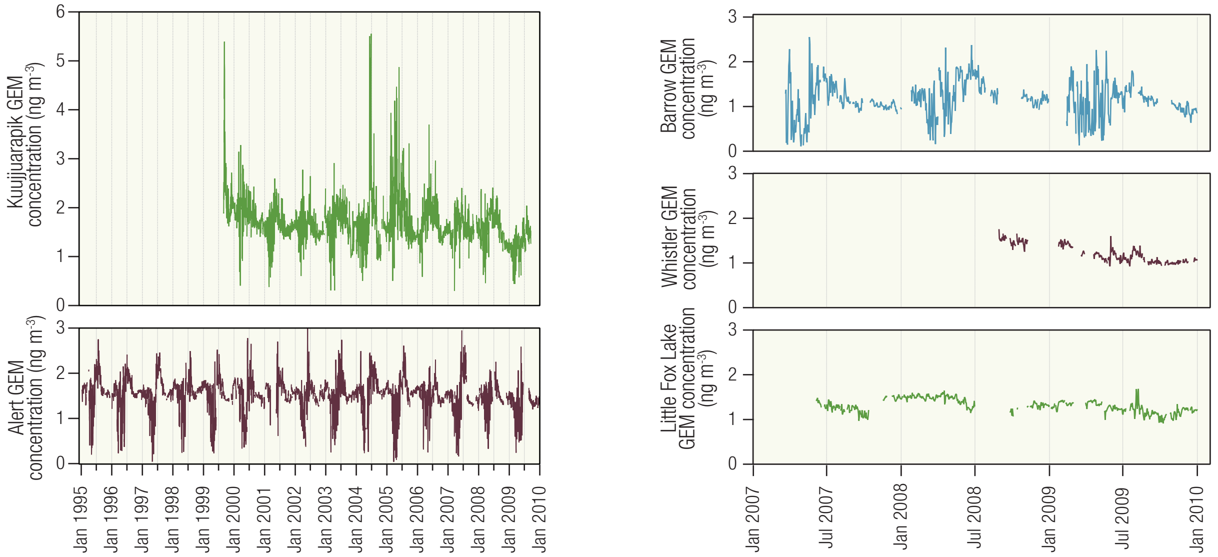

FIGURE 3.9

Time series of GEM daily air concentrations at Alert (Nunavut), Kuujjuarapik (Quebec), Little Fox Lake (Yukon), Whistler (British Columbia), and Barrow (Alaska). Data are from Steffen (2011) and Pilote (2011). Vertical gridlines are spaced six months apart.

AMDEs at Alert depressed the springtime average GEM concentration to the lowest seasonal value, the high GEM peaks in late spring at Barrow and Kuujjuarapik—which are not prevalent until June at the higher-latitude site of Alert—brought the springtime average concentrations up to values that are similar to or higher than fall and winter averages (Table 3.2). Finally, AMDEs and accompanying rapid increases in GEM resulted in high variability of both spring and summer GEM concentrations at sites where AMDEs occur—shown by the high standard deviation of seasonal measurements in Table 3.2. In contrast, at Little Fox Lake and Whistler, where AMDEs were not observed in the time series (Fig. 3.9), the mean GEM concentrations were consistent between seasons and much less variable within each season as reflected by the low standard deviations of the data. Seasonal averages at Little Fox Lake ranged from 1.17 ng m^{-3} in the fall to 1.43 ng m^{-3} in the spring. Whistler exhibited the smallest seasonal difference of all the sites, with seasonal averages ranging from 1.14 ng m^{-3} in the spring to 1.21 ng m^{-3} in the winter.

3.4.2 Reactive gaseous Hg and airborne particulate Hg

Long-term measurements of atmospheric Hg speciation are currently limited to Alert. Measurements of RGM and PHg from 2002 to 2009 are shown with GEM measurements in Figs. 3.3, 3.5 and 3.6. Six months of GEM, RGM, and PHg data were also collected at Churchill (Fig. 3.4) (Kirk et al. 2006). Both sites reveal that RGM and/or PHg values were high in the springtime during periods when GEM is unusually low, consistent with the conversion of GEM to RGM and/or PHg during AMDEs. These springtime means and standard deviations are included in Table 3.2. However, it should be noted that currently these measurements are only operationally defined because there are no existing calibration standards and the exact chemical compounds that comprise RGM and PHg are unknown. Therefore, data from these two sites have not been quantitatively compared. Identification of these chemical species remains an important knowledge gap.

3.5 Temporal trends of gaseous elemental Hg

As global anthropogenic emissions of Hg continue to change, and as the Arctic undergoes dramatic environmental changes, such as shrinking permanent sea ice, the continuous monitoring of atmospheric Hg provides important information about long-term changes in the transport, chemistry, and deposition of this pollutant in the Arctic environment. Worldwide atmospheric measurements of GEM up to the early 2000s suggest that concentrations of atmospheric Hg increased from the 1970s to a peak in the 1980s and then decreased to a plateau around 1996 to 2001 (Slemr et al. 2003). Similarly, a recent reconstruction of GEM levels in firn air (air bubbles trapped in glacier ice) from Greenland indicated that GEM increased from the 1940s to the 1970s and reached a plateau around the mid-1990s (Fäin et al. 2009).

As of 2011, continuous measurements of GEM have been made for over 15 years at Alert and for 11 years at Kuujjuarapik (Fig. 3.9). Temporal trends in these datasets were assessed using statistical techniques to account for large seasonal variability. The techniques included seasonal decomposition (Temme et al. 2004) and the seasonal Kendall test for trend (Cole and Steffen 2010). While analysis of the Alert data to the end of 2004 found no significant trend in GEM air concentrations (Temme et al. 2007), subsequent analysis using additional years of data (to 2007) revealed a decrease of -0.6% per year (Cole and Steffen 2010). No significant long-term trends have been reported for Kuujjuarapik to date.

To update the currently published GEM trends and compare these two sites, the seasonal Kendall test was performed on the GEM concentration measurements at Kuujjuarapik for the period from 2000 to 2009. Measurements from 1999 were excluded because a full year of data was not collected. Alert measurements were also re-analyzed for the same time period, along with GEM concentrations from the mid-latitude site at Saint-Anicet (Quebec) (45.1° N, 74.3° W), for comparison. Overall trends based on the entire dataset were (with 95% confidence limits): -0.9 ± 0.6% per year at Alert, -2.0 ± 0.9% per year at Kuujjuarapik, and -1.9 ± 0.1% per year at Saint-Anicet. Trends at the lower-latitude Quebec site agreed well with the reported decrease in background GEM concentration at Mace Head (Ireland) of -1.8 ± 0.1% per year over the period 1996 to 2009 (Ebinghaus et al. 2011). The rate of decrease at Alert was faster over this 10-year period than was found for 1995 to 2007 period (Cole and Steffen 2010), but is still significantly slower than at the lower latitude sites at Saint-Anicet and Mace Head. It should also be noted that GEM in the southern hemisphere declined at a faster rate of approximately -2.7% per year from 1996 to 2009, based on data from shipboard measurements and monitoring at Cape Point (South Africa) (Slemr et al. 2011).

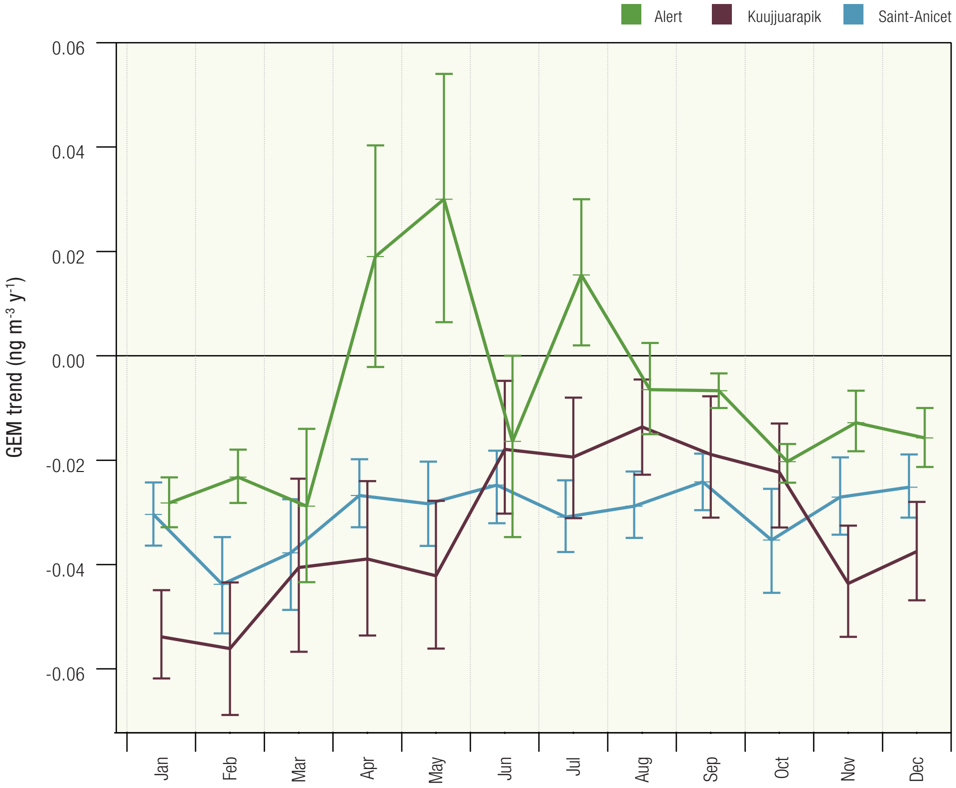

FIGURE 3.10

Trends by month in GEM concentrations at Saint-Anicet, Kuujjuarapik, and Alert over the period 2000 to 2009 (based on daily mean concentrations) using the seasonal Kendall test and Sen's estimator of slope. Data are from Steffen (2011) and Pilote (2011). Error bars show 95% confidence limits of the slopes.

Monthly trends based on the seasonal Kendall test and Sen's estimator of slope were determined for GEM at Alert, Kuujjuarapik, and Saint-Anicet (Fig. 3.10). These trends represent an estimate over the ten-year period of the year-to-year rate of change in GEM concentration in a given month—positive values indicate a concentration increase and negative values indicate a decrease. While the trend in GEM concentration at Saint-Anicet was relatively consistent between different months, trends at Kuujjuarapik and Alert varied from month to month (Fig. 3.10). At Kuujjuarapik, the year-to-year decreases in GEM concentration for the months of November to May were greater than the decreases from June to October. At Alert, concentrations of GEM in April, May, and July increased from 1996 to 2009—though the trend was not significant in April—and decreased in the other months. This result differs from trends between 1995 to 2007 at Alert (Cole and Steffen 2010) that showed increasing GEM levels only in May, and suggests that even ten years of data may be insufficient for establishing a robust trend.

Another study of long-term trends at Resolute Bay examined measurements from 1974 to 2000 of filterable Hg. That is, manual samples of Hg collected by passing air through particle filters and is likely PHg and RGM combined (Li et al. 2009). These authors reported a decrease of approximately 3% per year in total filterable Hg in summer and fall which is similar to the world-wide decrease in Hg emissions from anthropogenic activities between 1983 and 1995. Considerable variability was found in the data during the winter and early spring months suggesting some influence of AMDEs in the samples.

At this time, it is not clear why atmospheric Hg levels have decreased more rapidly at sub-Arctic and temperate locations than at a polar location. GEM trends in polar regions would reflect any changes in local chemistry (e.g., AMDEs), local emissions, or altered transport of polluted air from lower latitudes as well as trends in hemispheric background GEM. Springtime increases in GEM at Alert have previously been attributed to changes in the timing of AMDEs (Cole and Steffen 2010) but it was determined that the combined strength and frequency of AMDEs have not changed significantly. Increases or slower decreases of GEM concentrations in the summer months at Alert and Kuujjuarapik could theoretically be tied to unidentified local or regional emissions and this possibility requires further investigation. Finally, it is also possible that Alert is more strongly influenced by source regions where Hg emissions are increasing (such as Asia) compared with Kuujjuarapik, Saint-Anicet, Mace Head, or Cape Point (section 3.6.4), that inputs from polluted regions are increasing, or that large climate-related variability is impacting the trends (Cole and Steffen 2010). Continued long-term monitoring at locations such as Little Fox Lake and Whistler, where there is no contribution from local AMDE chemistry, will help to distinguish between possible contributing factors to the observed trends in the Arctic.

3.6 Atmospheric Hg models

Atmospheric Hg models simulate the cycling of Hg in the atmosphere starting from emissions to the air and ending with deposition to terrestrial and aquatic surfaces. Emissions to the air include anthropogenic emissions, natural emissions, and re-emissions of previously deposited Hg of both anthropogenic and natural origins. These models are used to estimate ambient Hg concentrations and deposition fluxes, to understand atmospheric transport pathways, to estimate source attribution, to explain long-term trends, and to predict future levels of Hg pollution. The models complement direct measurements by providing spatial coverage and detailed information on Hg budgets in the Arctic environment.

Atmospheric Hg models incorporate emissions of GEM, RGM, and PHg and their parameterized gas and aqueous phase chemistry, phase exchange processes between air, aerosol and clouds, wet and dry deposition, boundary layer and cumulus cloud mixing, and transport. Owing to the global transport of Hg, hemispheric or global scale atmospheric models have been developed to estimate the atmospheric Hg contribution to the Arctic. The global models are typically integrated (advanced temporally) for a few years to establish a balance between the emissions, atmospheric Hg concentrations, and deposition. Current Hg models are primarily constrained by measurements of surface GEM concentration and wet deposition fluxes from North America and Europe where long-term observations with sufficient spatial coverage are available. A limited number of vertical profiles of GEM and fluxes of Hg from terrestrial and oceanic surfaces are also used to constrain the models. Current measurements of RGM and PHg concentrations in the atmosphere are very limited and have a high degree of uncertainty for use in model development (Gustin and Jaffe 2010, Lyman et al. 2010). Since wet deposition of Hg occurs through the scavenging of oxidized Hg species in the atmosphere, it is possible to constrain the total oxidized Hg concentration in the lower troposphere using observed Hg concentrations in precipitation. Of course, Hg modelling in the Arctic is further complicated by AMDE processes.

Environment Canada has developed the Global/Regional Atmospheric Heavy Metals Model (GRAHM) which includes a complete set of meteorological processes and Hg physical-chemical processes (Dastoor and Larocque 2004, Dastoor et al. 2008). Three other atmospheric models have included a detailed Hg component as follows: 1) Danish Eulerian Hemispheric Model (DEHM) (Christensen et al. 2004); 2) Global EMEP Multi-media Modelling System (GLEMOS) (Travnikov and Ilyin 2009); and, 3) a global model driven by data from the Goddard Earth Observing System (GEOS-Chem) (Holmes et al. 2010). All four models have incorporated AMDEs as part of the chemistry. The largest differences among models are found in the values and spatial distribution of natural emissions and re-emissions, the major oxidants of GEM and the reaction products represented and, in the Arctic, the GEM-Br oxidation rates, Br concentrations and parameterization of re-emission of GEM from the snowpack. The Canadian model (GRAHM) was used in this assessment of Hg in the Canadian Arctic. A brief description of the model is presented in the following section.

3.6.1 Factors included in the Global/Regional Atmospheric Heavy Metals Model

Anthropogenic emissions included in this model are derived from the inventory of global anthropogenic emissions of Hg for 2005 (Fig. 3.11; AMAP/UNEP 2008). Details on the methods, data sources, and other information can be found in AMAP/UNEP (2008). Total global emissions from natural sources and re-emissions of previously deposited Hg (from land and oceans) are based on the global Hg budgets by Mason and Sheu (2002). Land-based natural emissions are spatially distributed according to the natural enrichment of Hg. Land re-emissions are spatially distributed according to the historic deposition of Hg and land-use type. Oceanic emissions depend on the distribution of primary production. Both land and oceanic emissions temporally depend on the surface temperature. Gaseous and aqueous phase chemistry of Hg is parameterized in the model and includes GEM, RGM, and PHg species. The primary gaseous oxidants of GEM included in GRAHM are ozone (O_3) and halogen species (Br, Cl, Br_2, Cl_2, BrO). In the aqueous phase, both oxidation and reduction pathways of Hg are included in the model. The oxidants include O_3, hydroxyl radical (OH), and hypochlorous acid

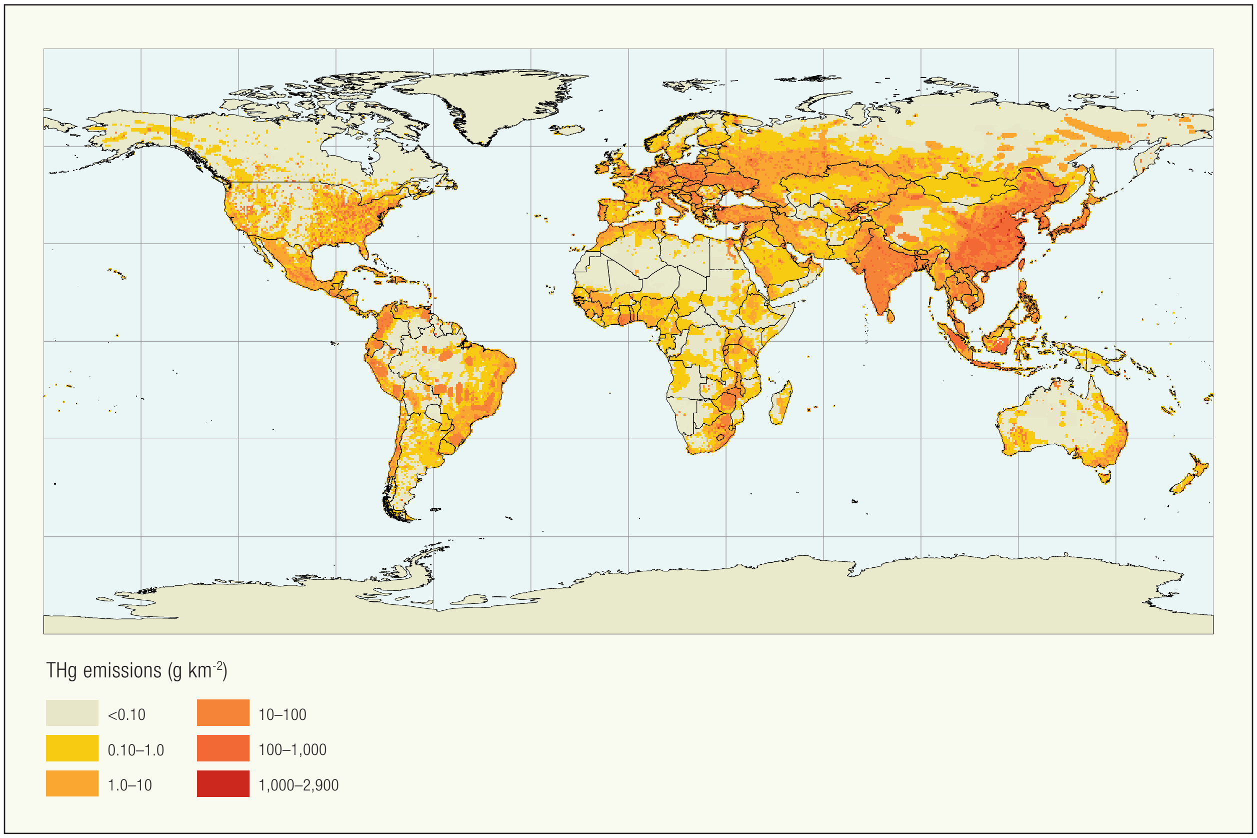

FIGURE 3.11

Global distribution of anthropogenic emissions of THg in 2005 (source: AMAP 2011).

(HOCl)/hyperchlorite ion (OCl^-) while the reductants include dissolved sulphur dioxide (SO_2), HO_2 and photoreduction of dissolved Hg. Photochemical oxidation of GEM by halogens, speciation of products, deposition pathways, and re-volatilization of GEM from snow are also parameterized to estimate the net accumulation of Hg in the Arctic. Scavenging of gaseous and particulate Hg species into atmospheric precipitation (wet deposition) and direct deposition to the earth's surface (dry deposition) are the removal mechanisms employed in the model. The dry deposition velocity is calculated using the resistance analogy (aerodynamic resistance, quasi-laminar sub-layer resistance, and surface resistance) for GEM and RGM, while the dry deposition velocity of PHg is calculated as fine particulate matter.

3.6.2 Model simulations of Hg deposition

In recent years, several models have provided estimates of Hg deposition to the Arctic. Using DEHM, Skov et al. (2004) estimated a deposition of 208 t y^{-1} of Hg to the Arctic area north of the polar circle (66.56° N), of which 120 t y^{-1} was attributed to AMDEs. Using an early version of GRAHM that did not incorporate post-AMDE re-emission but did include other re-emissions, Ariya et al. (2004) estimated that 325 t y^{-1} of Hg is deposited in the Arctic north of 60° including 100 t y^{-1} deposited as a result of AMDEs. Dastoor et al. (2008) estimated a 174 t y^{-1} net accumulation of Hg in snow (± 150–181 t y^{-1}) north of 66.5° latitude. This estimate incorporated bi-directional exchange of Hg fluxes in the Arctic in GRAHM to account for the fast re-emission of Hg from snowpack following AMDEs. Brooks et al. (2006) measured Hg deposition, re-emission, and net surface gain fluxes of Hg at Barrow (Alaska) between March 25 and April 7, 2003, as 1.7 µg m^{-2}, 1.0 ± 0.2 µg m^{-2} and 0.7 ± 0.2 µg m^{-2}, respectively. Dastoor et al. (2008) found excellent agreement between model-derived fluxes and measurements during the Barrow study with rates of 1.8 µg m^{-2} deposition, 1.0 µg m^{-2} re-emission and 0.8 µg m^{-2} net surface gain of Hg. The most recent version of GRAHM yields a deposition rate of 143 t y^{-1} above the Arctic Circle (66.5° N) of which approximately 25 t y^{-1} is deposited between 63°–141° W. In comparison, the yearly net deposition flux estimated by GLEMOS and DEHM in the Arctic are 131 t y^{-1} and 110 t y^{-1}, respectively. Net deposition is defined as yearly gross deposition minus re-emission of seasonally deposited Hg to snowpacks. In the Canadian Arctic, the average deposition flux using GRAHM is simulated to be approximately 4.5 µg m^{-2} y^{-1} in the Arctic (north of 66.5°), approximately 9 µg m^{-2} y^{-1} in the western sub-Arctic (60–66.5° N, 102–141° W), and approximately 12 µg m^{-2} y^{-1} in the eastern sub-Arctic (60–66.5° N, 63–102° W) regions. Figure 3.12 shows the annual net deposition of Hg (left) and annual average surface air concentrations (right) in the Arctic simulated by GRAHM. The model, in general, simulates a north to south deposition gradient with increasing deposition that could be attributed to Br concentrations (which are higher in coastal regions), proximity to emission sources, and increasing precipitation amounts. The eastern sub-Arctic displays higher Hg deposition and lower GEM concentrations compared to the western sub-Arctic. Modelled longitudinal trends in GEM concentrations differed from measurement observations (see section 3.4.1), and further research is needed to clarify

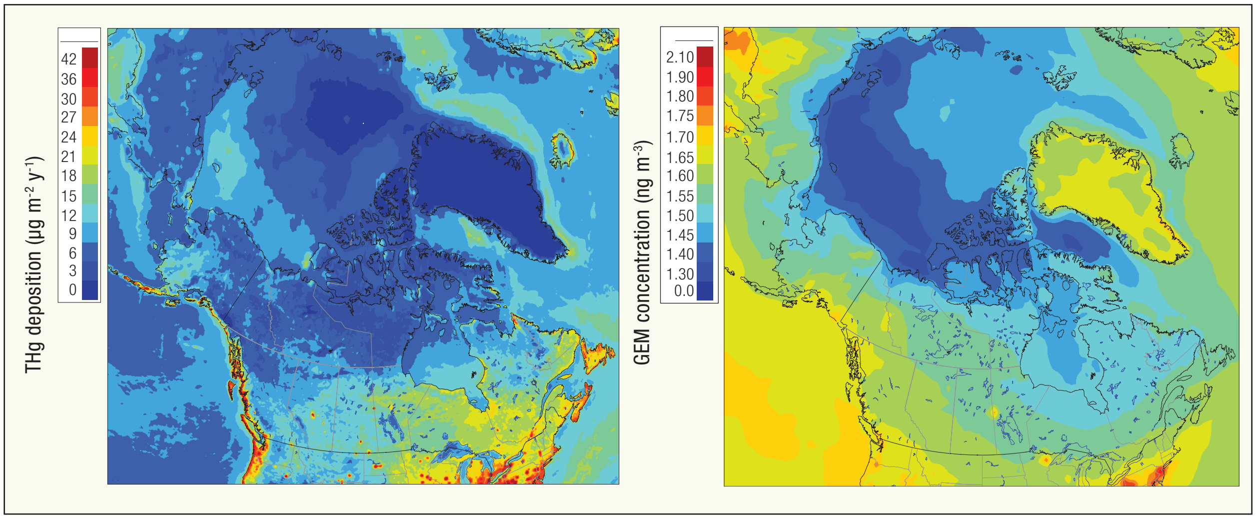

FIGURE 3.12

Spatial distribution of THg deposition (µg m^{-2} y^{-1}; left), and surface air concentration of GEM (ng m^{-3}; right) in the Canadian Arctic simulated by GRAHM in year 2005 (Dastoor 2011).

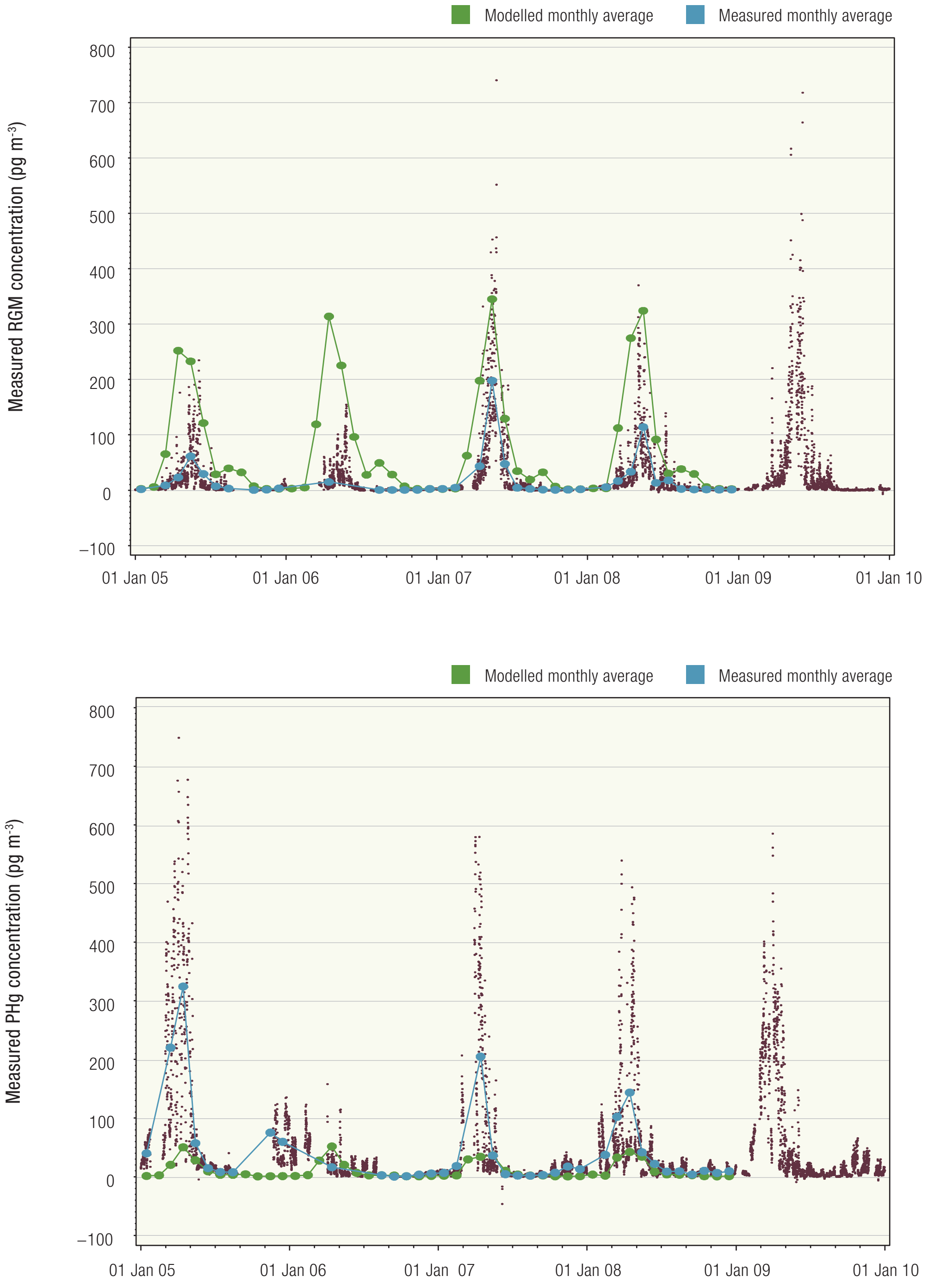

FIGURE 3.13

Surface air concentrations of RGM (top panel) and PHg (bottom panel) simulated by GRAHM and measured at Alert (Nunavut) in pg m^{-3}. Dark purple dots represent the concentrations from measured samples, blue dots represent the monthly average of those measured samples, and green dots represent the monthly average concentration predicted by the model (Dastoor 2011).

east-west variation in GEM concentration in northern Canada. The eastern sub-Arctic appears to be directly influenced by North American sources.

A few datasets of measured wet Hg flux are available from sub-Arctic precipitation collection stations at Churchill and Fort Vermilion (Alberta) in Canada (Sanei et al. 2010), and Kodiak, on the Pacific coast of Alaska (through the MDN, http://nadp.sws.uiuc.edu/mdn). Gross wet deposition fluxes from these stations were compared to model estimates from GEOS-Chem and GRAHM for these sites (Sanei et al. 2010). The modelled wet Hg fluxes for the three sites ranged from 2.1–4.1 μg m^{-2} y^{-1} compared to measurements of 0.54–5.3 μg m^{-2} y^{-1}. The model estimates were closest to the measured fluxes at Kodiak (GEOS-Chem = -47% and GRAHM = +23%). The largest discrepancy between observations and model estimates was found at Churchill. Kirk et al. (2006) measured high concentrations of Hg in snow during AMDEs (over 100 ng L^{-1}). However, Sanei et al. (2010) did not measure high wet deposition fluxes at Churchill during spring using their method (see also section 3.3). The relative contribution of wet and dry deposition at Arctic sites, including Churchill, is unknown. There is a discrepancy between measured and modeled distribution of RGM and PHg at Alert (Fig. 3.13) that can give rise to inaccuracies in the partitioning between dry and wet depositions employed in the model. Uncertainties in the partitioning of modelled dry and wet deposition and/or underestimation of wet deposition fluxes by measurements are possible sources of the discrepancy between measured and modelled estimates of wet deposition at Churchill. Dry deposition flux measurements and additional wet deposition measurements are required to fully evaluate the models and the impact of AMDEs in the Arctic.

3.6.3 Model simulations of Hg concentrations in surface air

Monthly mean GEM concentrations and deposition fluxes derived from GRAHM were compared to measured monthly GEM means from combined years at six Arctic monitoring sites: Barrow (USA), Alert (Canada), Station Nord (Greenland), Ny-Ålesund, (Norway), Andoya (Norway), and Amderma (Russia) (Fig. 3.14). The GRAHM-derived and the measured yearly average GEM concentrations at the six sites are 1.47 and 1.52 ng m^{-3}, respectively. This close agreement between the two averages suggests that very similar physical-chemical processes are occurring throughout coastal circumpolar Arctic regions. The differences between GRAHM and GLEMOS or DEHM estimated mean GEM concentration and Hg deposition in the Arctic are within a factor of 1.2 and 1.3, respectively. The close agreement between the models is not surprising because they are all constrained by the GEM measurements. The air concentrations are determined by the ratio between emission and removal rates (by deposition and chemical reaction), both of which have uncertainties, thus different model configurations can predict similar concentrations. However, the differences in how the removal processes are calculated in

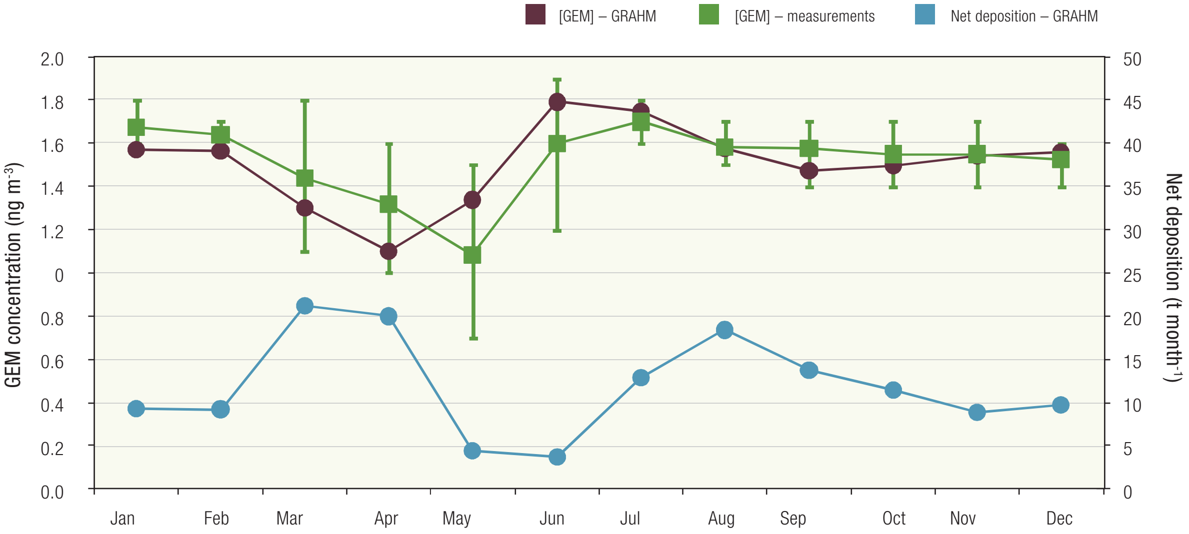

FIGURE 3.14

Monthly mean ambient GEM concentrations (north of 66.5°) simulated by GRAHM and observed concentrations averaged over six Arctic sites: Barrow (USA), Alert (Canada), Station Nord (Greenland), Ny-Ålesund (Norway), Andoya (Norway), and Amderma (Russia) (ng m^{-3}; left scale). Total monthly deposition was simulated by GRAHM (t $month^{-1}$; right scale) (Dastoor 2011).

the various models result in larger uncertainties in model estimates of deposition fluxes as compared to the air concentration estimates.

The model chemical parameterization is capable of simulating the seasonal pattern of oxidized Hg (RGM and PHg) at Alert (Fig. 3.13). The magnitude of oxidation in the springtime is simulated within the measured range, whereas there is notable discrepancy in estimating the partitioning between gas and aerosol phases. Currently, partitioning between RGM and PHg in the model is somewhat arbitrary because the heterogeneous chemistry is a knowledge gap.

3.6.4 Long-range transport and source attribution of Hg deposition in the Canadian Arctic

The origin of atmospherically deposited Hg in the Arctic was analyzed by estimating the contribution from anthropogenic emissions in the source regions shown in Fig. 3.15. Total anthropogenic emissions of Hg (in t y^{-1}) in these regions and the percent contribution to total global anthropogenic emissions are provided. The combined contributions to deposition of natural emissions and re-emissions, anthropogenic emissions, and oceanic emissions were simulated in the model.

Figure 3.16 presents estimates of contributions to the 2005 deposition flux from each source region as well as total contributions from all anthropogenic, terrestrial, and oceanic emissions as absolute values (top panel) and as a percentage of total deposition to Canadian Arctic and sub-Arctic regions (bottom panel). The eastern sub-Arctic region showed the highest deposition fluxes of all the Arctic regions, possibly because of higher precipitation rates, favourable land-use types for dry deposition, and the presence of AMDEs around Hudson Bay.

Figure 3.16 (bottom panel) shows the percent contribution from various source regions to deposition in the Canadian Arctic. Global anthropogenic emissions, terrestrial emissions, and oceanic emissions each contribute 20–28%, 40–45% and 30–34%, respectively, to Hg deposition in Canadian Arctic and sub-Arctic regions. The anthropogenic contribution to Hg deposition is higher (28%) in the High Arctic than the sub-Arctic (20–21%) due to higher contributions from East Asia (15%, 11%) and Europe (3%, 2%). East Asia

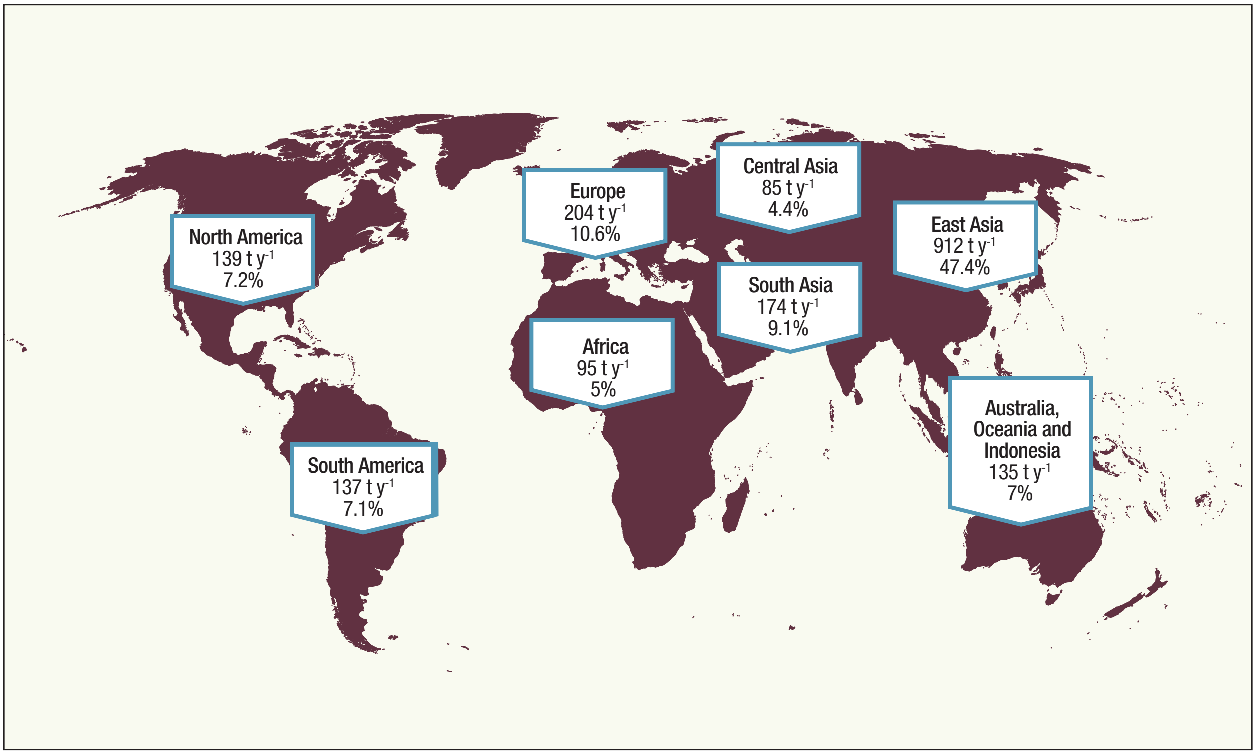

FIGURE 3.15

Location of source regions considered in the source attribution study. Total anthropogenic emissions (t y^{-1}) and percent contributions to global anthropogenic emissions of Hg in these regions for 2005 are shown on the map. Receptor regions in the study are the same as the source regions.

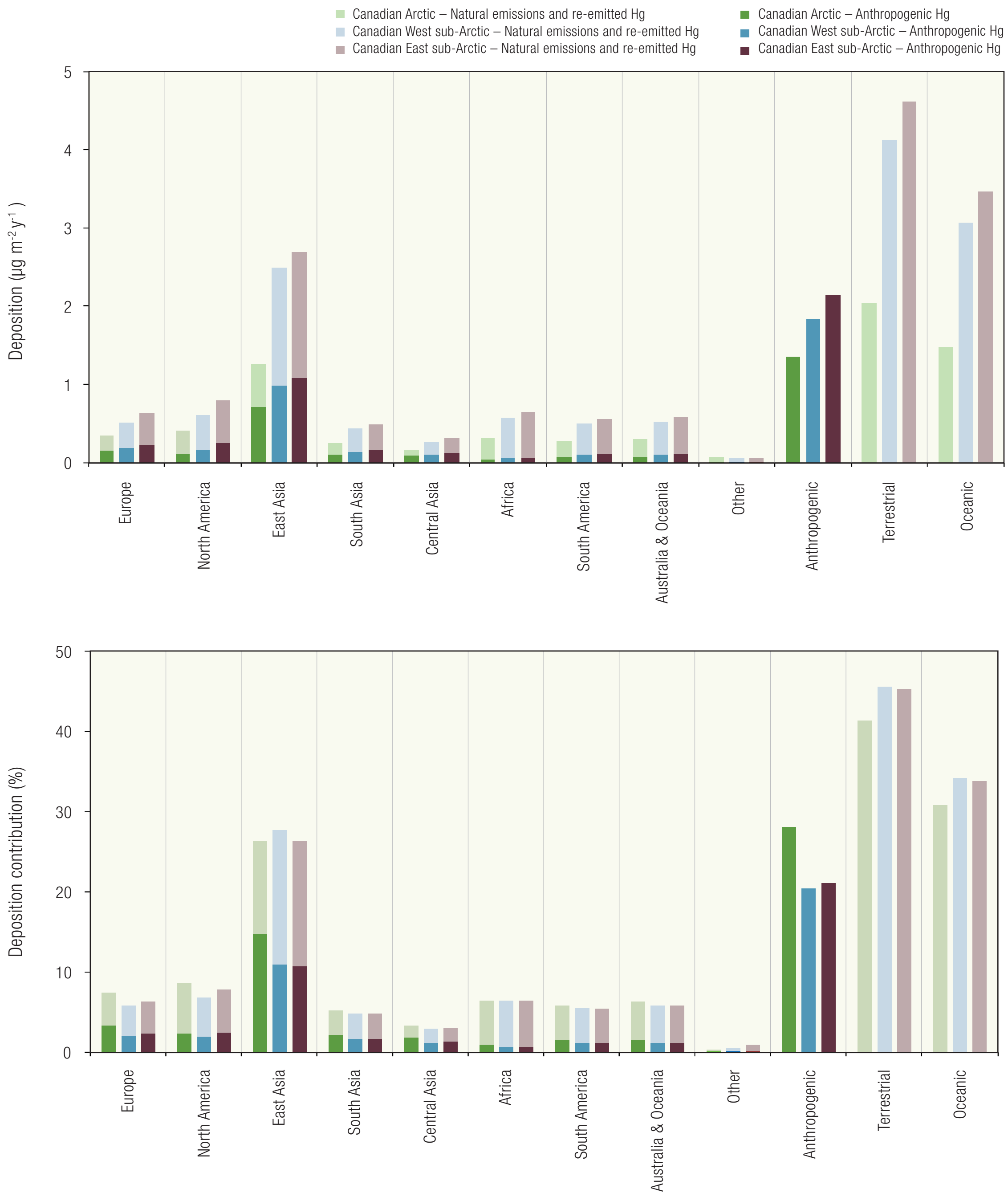

FIGURE 3.16

Contribution of source regions to THg deposition to the Canadian High Arctic, west sub-Arctic and east sub-Arctic simulated by GRAHM in 2005 (Dastoor 2011). Contributions are presented as absolute values (µg m^{-2} y^{-1}; top panel) and as percentages (bottom panel).

contributed the largest proportion of anthropogenic Hg emissions to the Arctic (10–15%) followed by Europe (2–3%), North America (2–2.5%), and South Asia (1.5–2%). An estimated two thirds of deposition to the Arctic comes from natural emissions and re-emitted Hg from land and oceans. Therefore, an accurate representation of these emissions in the models is essential for applying the models to estimate future changes in deposition.

Estimated by combining the results of four models, an *ensemble* average of the spatial distribution of percent Hg deposition from anthropogenic emissions in Europe, North America, East Asia, and South Asia is shown in Fig. 3.17. The figure illustrates that anthropogenic Hg emissions from East Asia have a greater impact on levels of Hg deposition in the Canadian Arctic compared to other source regions. These results suggest that increasing emissions in East Asia could increase deposition of Hg to the Canadian Arctic and offset the decrease in deposition that has resulted from reductions of North American emissions.

Measured concentrations of air Hg and simulated contributions to total concentration from four source regions—Asia, North America, Russia, and Europe—were analyzed for Alert and Little Fox Lake (Durnford et al. 2010). Similar to the results shown in Fig. 3.16, results from this analysis indicate that Asia contributed by far the most Hg at both Alert in 2000 (~30%) and at Little Fox Lake (~35%) in 2008. At Little Fox Lake in 2008, the next largest source regions contributing to atmospheric Hg levels were North America (~10%), Russia (~6%), and Europe (~5%). Most of the variability at Alert and Little Fox Lake was determined by Hg emitted in Asia. Little Fox Lake has been assessed as a good location for monitoring Asian long-range transport. Long-range transport events (LRT), where Hg-rich plumes are transported past the measurement sites, were investigated (Durnford et al. 2010). It was found that Hg-rich plumes originating in Asia cross the Pacific Ocean, mix with Hg-rich plumes originating in North America, and are transported together. Maximum LRT events simulated at Alert were found to come from Russia and Asia. The LRT events simulated at Little Fox Lake primarily reflected Hg transported from Asia followed by Russia.

3.6.5 Recent historic changes and future projection of Hg deposition and surface air concentrations

Long-term temporal changes in atmospheric Hg levels in the Arctic can be used to monitor the effect of changes in anthropogenic Hg emissions worldwide and to model their impact on deposition to the Arctic. It is challenging to relate these observed trends to the spatially heterogeneous changes in global anthropogenic Hg emissions because the temporal trends of Hg include the impacts of changes in weather patterns, land and oceanic emissions, atmospheric chemical composition, land use, and climate. However, modelling the impacts of these changes on the atmospheric cycling of Hg, both independently and in combination, can help explain the observed Hg trends in the Arctic and predict future Hg levels based on projected future emissions. Thus, GRAHM was applied to assess the impact of changing anthropogenic emissions on Hg concentrations and deposition in the Arctic. Figure 3.18 shows the changes in anthropogenic Hg emissions within major emission regions and globally for 1990, 1995, 2000, and 2005. Figure 3.18 also shows the expected changes in global and regional anthropogenic emissions under three future scenarios for 2020 (described as follows). Global Hg inventories for 1990, 1995, 2000 and 2005 were previously developed by the Arctic Monitoring and Assessment Programme (AMAP) but the methods used for developing these inventories were not consistent. Recently, a more comparable set of historical global emissions were developed for the period 1990 to 2005 using a standardized methodology and consistent information for estimating certain emissions (AMAP/UNEP 2008). According to these reanalyzed emission inventories, the emissions in Europe and North America declined most rapidly from 1990 to 2000, and declined less from 2000 to 2005. The emissions from other continents, most notably from Asia, steadily increased from 1990 to 2005. Global anthropogenic Hg emissions declined from 1990 to 1995 by 7.8%, remained steady from 1995 to 2000 and increased by 5.6% from 2000 to 2005. It should be noted here that these inventories do not capture inter-annual changes in emissions which introduce a source of uncertainty to the modelling results, nor do they include natural emissions that may not be constant. As noted previously, Fig. 3.18 also includes expected changes in regional and global anthropogenic emissions under three future scenarios (AMAP/UNEP 2008) described later in the section.

Atmospheric Hg concentration and deposition were simulated from the year 1990 to 2005 based on changes in meteorology and anthropogenic emissions (Figs. 3.19 and 3.20). The trend simulation was repeated twice to analyze the changes. In the first set, the emissions were kept constant at the year 2005 level and only meteorology was allowed to change from 1990 to 2005. In the second set, the meteorology was kept constant at year 2005 conditions and emissions were allowed to change from 1990 to 2005. Comparison of the simu-

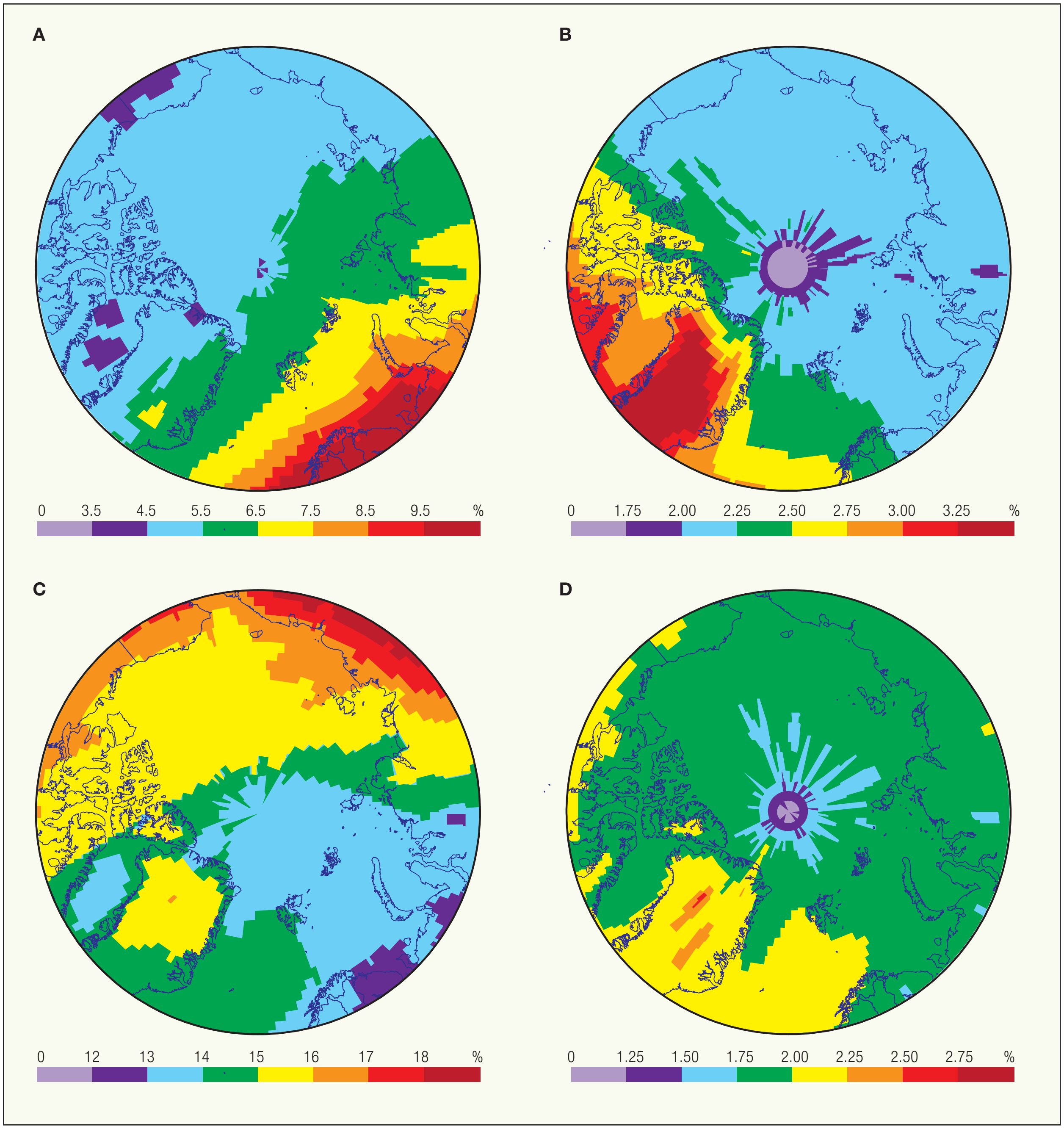

FIGURE 3.17

Spatial distribution of the percentage of THg deposition to the Arctic contributed from the four main source regions determined as a *model ensemble* by averaging four available world-wide global Hg models (source: AMAP 2011). The maps are ordered by source region: Europe (A), North America (B), East Asia (C), and South Asia (D). Note that the scales for each of the regions are different.

lated temporal trends from model versions with and without changing meteorology shows that significant inter-annual variability in deposition and air concentration is driven by inter-annual variability in meteorology, perhaps linked with climate oscillations. These results suggest that temporal trends detected over short periods of time or from intermittent datasets may not be conclusive evidence of changes in emissions. Controlled model simulations with changing meteorology alone and changing emissions alone, for the same time period, can be used to analyse Hg trends as illustrated in Fig. 3.19.

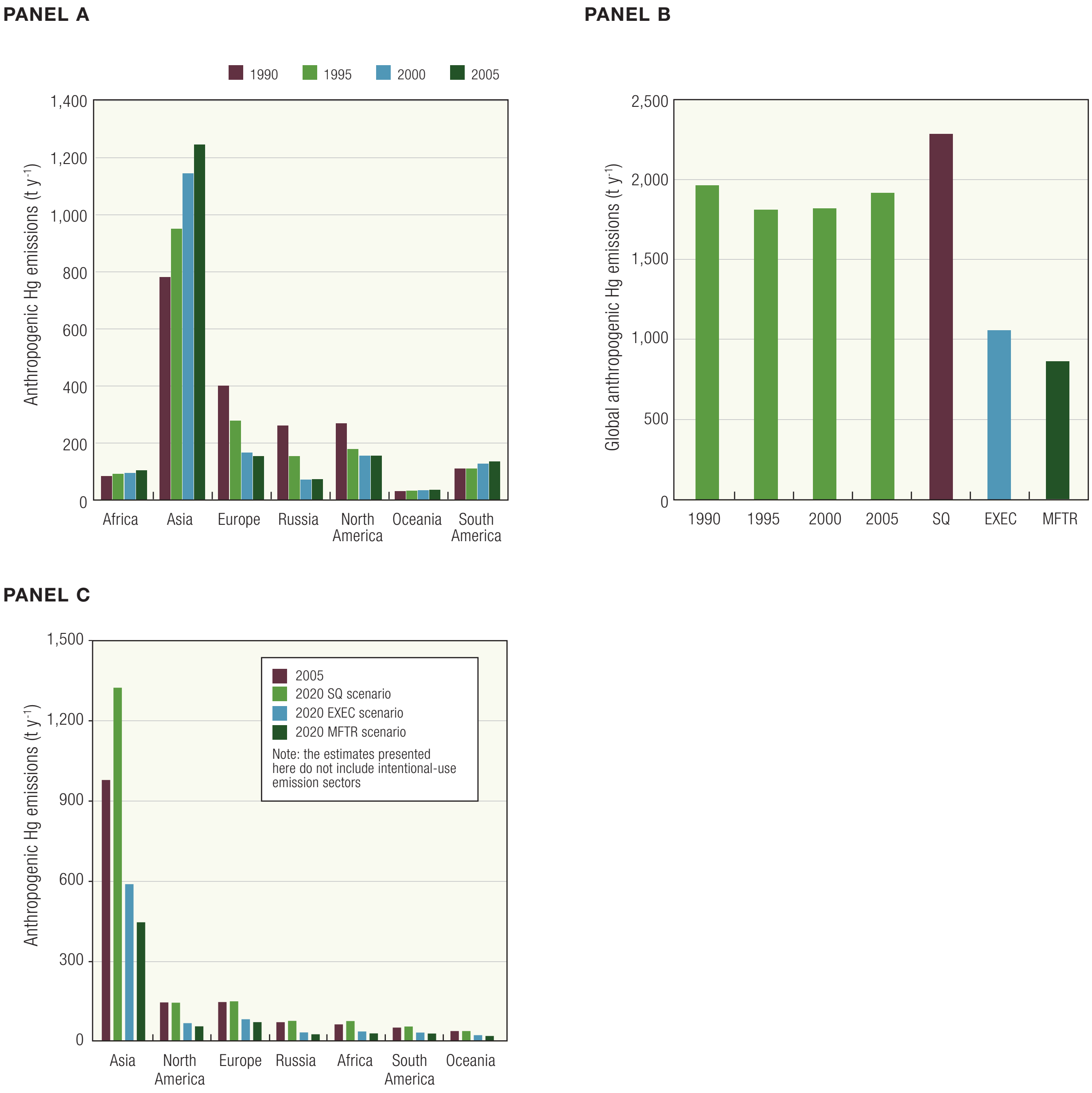

FIGURE 3.18

A) Revised estimates of yearly anthropogenic Hg emissions to air in 1990, 1995, 2000, and 2005 from different regions (t y^{-1}) (source: AMAP 2011). B) Total yearly global estimates of Hg emissions in 1990, 1995, 2000, and 2005 and three future scenarios (t y^{-1}) (Dastoor 2011). C) Regional yearly anthropogenic emissions of Hg under three future scenarios for the year 2020: *Status Quo* (SQ), *Extended Emissions Control* (EXEC), and *Maximum Feasible Technological Reduction* (MFTR) (t y^{-1}) (source: AMAP 2011).

Figure 3.19 shows the average net deposition and surface air GEM concentration from base and controlled simulations for the North American High Arctic (66.6–90° N, 170–10° W), and east and west sub-Arctic regions (east: 60–66.5° N, 100–10° W; west: 60–66.5° N, 170–100° W). Also shown for comparison are the average net deposition and surface air GEM concentrations for the Great Lakes region (40–50° N, 94–74° W). The main conclusion to note from Fig. 3.19 is that the model shows a declining trend in surface air concentrations from 1990 to 2005 for all three regions. As well, a constant or increasing deposition trend is found in

High and sub-Arctic regions as opposed to the declining deposition trend found in the Great Lakes region. These results are consistent with observed concentrations from atmospheric Hg measurement networks in the North American Arctic and mid-latitudes (section 3.5 and the National Atmospheric Deposition Program, NADP). The long-term wet deposition data from North American mid-latitudes that are available from the NADP suggest a small decline between 1997 and 2005. However, dry deposition trends are not known from observations and are model-estimated. In Arctic and sub-Arctic regions, there are no long-term records of direct measurements of wet or dry deposition. The relationship between the trends in emissions and/or atmospheric deposition of Hg and lake sediment Hg fluxes is not fully understood. However, an increasing trend in Hg fluxes to Arctic lake sediments has been observed in recent decades which is in contrast with the measured decline in air concentrations of Hg (Munthe et al. 2011). Climate change has been suggested as a possible explanation for this observation (section 5.4; Outridge et al. 2007, Munthe et al. 2011) although the decrease in atmospheric GEM can alternatively be explained by increased deposition of Hg to the surface through atmospheric transformation of GEM. The modelled trends (Fig. 3.19) show that changes in meteorology in recent years have led to an increase in net Hg deposition in the North American Arctic. This may partly explain the increasing Hg fluxes in Arctic lake sediments.

From 1990 to 2005, the model simulation shows a 15% increase in deposition in the High Arctic due to changes in meteorology, which is offset by decreases in North American and European emissions by 5%, resulting in an overall increase in the deposition by 10%. During this same period in the eastern sub-Arctic, simulations of deposition suggest an increase of 25% from meteorological changes and a decrease of 5% from emission changes, leading to an overall increase of 20% in deposition. Almost no change in deposition is found in the western sub-Arctic where meteorological changes have led to a 2% increase and emission changes have led to a 4% decrease resulting in an overall 2% decline in deposition over the 1990 to 2005 period.

Oxidized Hg is scavenged into cloud water throughout the troposphere where the air is more influenced by long-range transport from Asia in comparison to surface air. An analysis of modelled meteorological trends from 1990 to 2005 reveals that precipitation and solar insolation are increasing at the surface in North American Arctic regions. Increasing precipitation rates result in greater wet depositional fluxes of Hg. Increased surface solar insolation leads to changes in snow cover and depth which reduce GEM evasion from the snowpack and, in turn, leads to increased net deposition of Hg at the surface.

The differences in GEM concentration between the surface air in High and sub-Arctic regions are less pronounced compared to the differences in deposition fluxes between these regions. This is a result of wet and dry deposition being influenced by local meteorology and its changes compared to surface air concentrations of GEM. Declines of 6% and 4% are simulated by the model due to changes in meteorology for High Arctic and sub-Arctic regions, respectively. A decline of 6% due to emission changes in surface air concentrations is simulated by the model for all three regions. The 12% decline in surface air Hg concentrations in the High Arctic was found to result from both changing meteorology and changes in anthropogenic emissions from 1990 to 2005. The simulated decline in the surface air GEM concentration in the High Arctic is comparable with the measured decline at Alert as discussed in section 3.5. Lower Hg evasion from snowpack, likely due to changes in snow cover and depth, may be the primary meteorological cause for a decline in surface air GEM concentrations in Arctic North America.

Recent observations of a faster decline in surface air GEM concentrations in the sub-Arctic and at a few other mid-latitudinal global monitoring sites cannot be explained by the changes in global anthropogenic emissions alone (section 3.5). Decreases in terrestrial emissions of Hg, in response to the decline in anthropogenic emissions, are a possible explanation. However, it is not consistent with the longer estimated response time for the terrestrial environment. The model results presented in Fig. 3.19 suggest that recent declines in surface air concentrations could be related to changing surface meteorological conditions. It should be pointed out that the version of the GRAHM model employed here did not include the impact of changing anthropogenic emissions and/or land use cover other than for snow cover effects on terrestrial evasion rates.

In contrast to High and sub-Arctic regions in North America, significant decreases in Hg deposition (-20% in ~15 years) as well as surface air GEM concentrations (-10% in ~15 years) have been simulated by the model for the Great Lakes region. These declines are probably related mainly to reductions in North American Hg emissions because of the proximity of known anthropogenic sources. Direct emissions of oxidized Hg and GEM from local and global sources contribute to the deposition fluxes in temperate regions. Thus, a larger

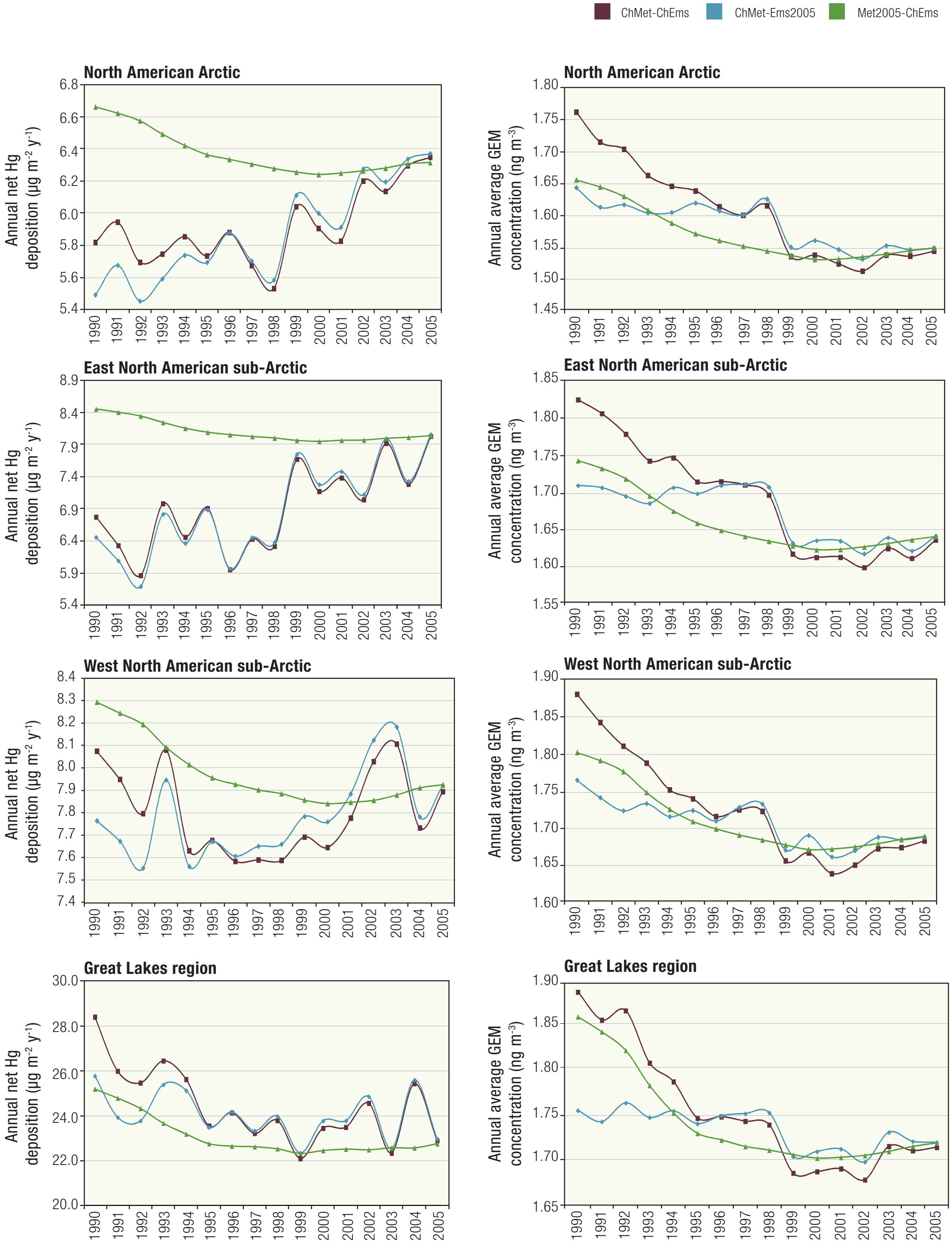

FIGURE 3.19

Modelled yearly trends of net Hg deposition (µg m^{-2} y^{-1}; left panels) and yearly average concentration of surface air GEM (ng m^{-3}; right panels) in the North American High Arctic (66.6–90° N, 170–10° W), east and west sub-Arctic regions (east: 60–66.5° N, 100–10° W; west: 60–66.5° N, 170–100° W), and Great Lakes region (40–50° N, 94–74° W). Three trend simulations were conducted: changing meteorology and changing emissions (ChMet-ChEms), changing meteorology and emissions constant at 2005 levels (ChMet-Ems2005), and changing emissions and meteorology constant at 2005 conditions (Met2005-ChEms). Data are from Dastoor (2011).

decrease in deposition flux was observed in the simulation compared to the decrease in GEM surface air concentration. These results illustrate that Hg trends observed at temperate locations cannot be extrapolated to the Arctic because the changes in the Arctic are dominated by the changes in meteorology and global anthropogenic emissions whereas Hg trends in the temperate regions are more influenced by the regional changes in emissions.

Both the modelled and observed temporal trends of average surface air GEM concentrations from October to December were compared for Alert (Fig. 3.20). Measured GEM concentrations during that period, which represent background levels at Alert (Cole and Steffen 2010), were used for comparison. It should be noted that model estimates represent average concentrations in a region corresponding to the model grid cell area around the measurement site. This comparison confirms that inter-annual variability in meteorology was responsible for the year-to-year variations in GEM surface air concentrations observed at Alert. When compared to the model-predicted trends in GEM concentration generated by changing emissions alone and keeping meteorology constant, the small decline observed in surface air GEM concentrations from 1995 to 2000 and the subsequent increase from 2000 to 2005 are found to be consistent with changes in global anthropogenic emissions during this period. These results indicate that Alert is a good location to detect changes in global anthropogenic Hg emissions. Currently, there are no estimates of anthropogenic emissions available beyond 2005. Therefore, it is difficult to interpret the more recent trend.

Figure 3.21 presents expected changes in deposition and surface air GEM concentrations in 2020 under three future global anthropogenic emissions scenarios (AMAP/UNEP 2008) described as follows.

1. The *Status Quo* (SQ) scenario assumes that current patterns, practices, and uses that result in Hg emissions to air continue in the future. Economic activity is assumed to increase in various regions with no change in emission control practices that are currently employed, leading to increased emissions from several sectors.
2. The *Extended Emissions Control* (EXEC) scenario assumes economic progress at a rate reflecting the future development of industrial technologies and emissions control technologies. Mercury-reducing technologies currently employed throughout Europe and North America would be implemented globally. It further assumes that measures currently committed to in Europe to reduce Hg emissions to air or water would be implemented throughout the world. These include measures adopted under the *Protocol on Heavy Metals* to the *Convention on Long-range Transboundary Air Pollution* (LRTAP), European Union Directives, and agreements to meet Intergovernmental Panel on Climate Change (IPCC) Kyoto targets on the reduction of greenhouse gases causing climate change (which will also result in reductions in Hg emissions).
3. The *Maximum Feasible Technological Reduction* (MFTR) scenario assumes all available solutions and measures are implemented, which will lead to the maximum degree of reduction of Hg emissions

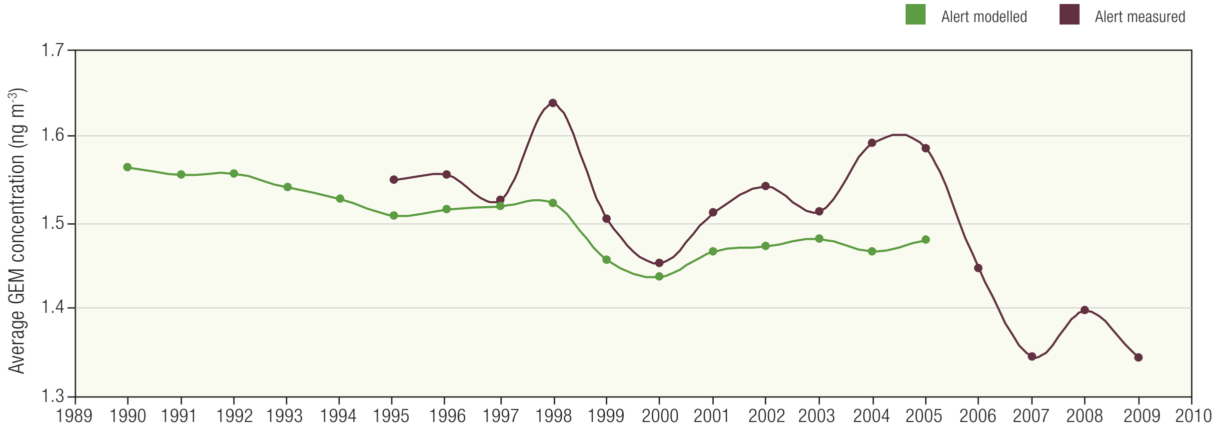

FIGURE 3.20

Comparison of measured (dark purple) and modelled (green) average GEM concentrations (ng m^{-3}) in surface air from October to December at Alert (Nunavut) (Dastoor 2011).

and Hg discharges to any environment. The monetary costs of these measures are taken into account but only as a secondary consideration.

The SQ emission scenario predicts a moderate increase of emissions in all regions except for North America (Fig. 3.18). The EXEC and MFTR scenarios both predict an emission decrease roughly by a factor of two and the difference between these scenarios is not significant. The largest reduction in Hg emissions is expected in Asia. Under the SQ scenario, the overall global anthropogenic emissions are estimated to increase by 19% between 2005 and 2020. Under the EXEC and MFTR scenarios, the emissions are expected to decrease in 2020 by up to 45% and up to 55%, respectively. These scenarios are a first attempt to predict future emissions and depend on a number of variables including economic development, implementation of emission control technologies, regulatory changes, and factors related to global climate change.

The model estimates were performed keeping the global meteorological conditions unchanged in all simulations for 2020 scenarios. The meteorology of 2005 was used as a reference for all the simulations and thus, no impact of climate change was taken into account. In the Canadian High and sub-Arctic, average increases of 3% and 3.1%, respectively, are expected for annual GEM concentrations in 2020 if emissions are to increase under the SQ scenario. In the Canadian High Arctic, average declines of 9.1% and 10.9% can be expected if the emissions are to be reduced according to EXEC and MFTR scenarios, respectively. In the Canadian sub-Arctic, average declines of 9.6% and 11.5% are expected for EXEC and MFTR scenarios, respectively. Differences in simulated changes in Hg concentrations between Arctic sectors are small. The largest estimated increase in Hg concentration is in the western sub-Arctic under the SQ scenario and the decline is largest in the eastern European sub-Arctic and west Asian sub-Arctic (not shown here). In the Canadian High and sub-Arctic, average increases in deposition of 5.2% and 4.8%, respectively, are expected by 2020 under the SQ scenario. Average declines of 14.9% and 18% are simulated under the EXEC and MFTR scenarios, respectively, in the Canadian High Arctic. In the Canadian sub-Arctic, average declines of 16.5% and 19.9% are expected for EXEC and MFTR scenarios, respectively. This comparison between changes in air concentrations and deposition reveals that the expected changes and spatial variations are larger for Hg deposition than for air concentrations of GEM. These results suggest that the changes in anthropogenic oxidized Hg in these scenarios have an influence on deposition in the Arctic and sub-Arctic through direct transport of these species. Stronger variations in deposition are the result of regional differences in meteorology that influence dry and wet deposition as well as regional differences in upper air Hg concentrations because wet deposition scavenges Hg at higher levels. The impact of changes in remote emissions such as from Asia is larger in the free troposphere compared to surface air. The increase in deposition estimates is found to be somewhat larger in the Arctic compared to the sub-Arctic, which is consistent with differences in source attribution for these two regions.

3.6.6 Uncertainties in model estimates of deposition in the Arctic

The two most fundamental parameters that determine the exchange of Hg between the earth's surface and atmosphere are air emissions of GEM and its overall oxidation rate. These parameters are important because of the dominance of GEM in emissions and its physical property of low solubility. GEM is subject to long-range transport whereas RGM and PHg deposit more rapidly near emission sources. Currently, estimates of anthropogenic emissions are more reliable than estimates of natural emissions and re-emissions of GEM. The natural and re-emissions of GEM from terrestrial and ocean surfaces are estimated to account for approximately half to two-thirds of total global GEM emissions (Sunderland and Mason 2007), and as a result, are highly uncertain and represent a major knowledge gap. Although GEM is the dominant species in anthropogenic Hg emissions, current inventories estimate significant emissions of oxidized Hg. The uncertainties in precise partitioning of anthropogenic Hg species at the stack or in-plume have an impact on the extent of the long-range transport of Hg simulated by the models. Current anthropogenic emission inventories lack temporal variations, leading to further modelling uncertainties.

The next major limitation of current atmospheric Hg models is uncertainty in the chemical processes involving Hg in air and at surfaces (Sabir et al. 2011, 2012). The current version of atmospheric Hg chemistry and related kinetics in the models is based on limited laboratory investigations. The uncertainties in gaseous Hg chemistry are associated with the reported kinetic constants and the lack of identified products of Hg reactions. The existing Hg kinetic parameters for the gas and aqueous phases, obtained from both theoretical calculations and laboratory measurements, vary significantly. The application of experimental data to the real atmosphere also has been questioned (Calvert and

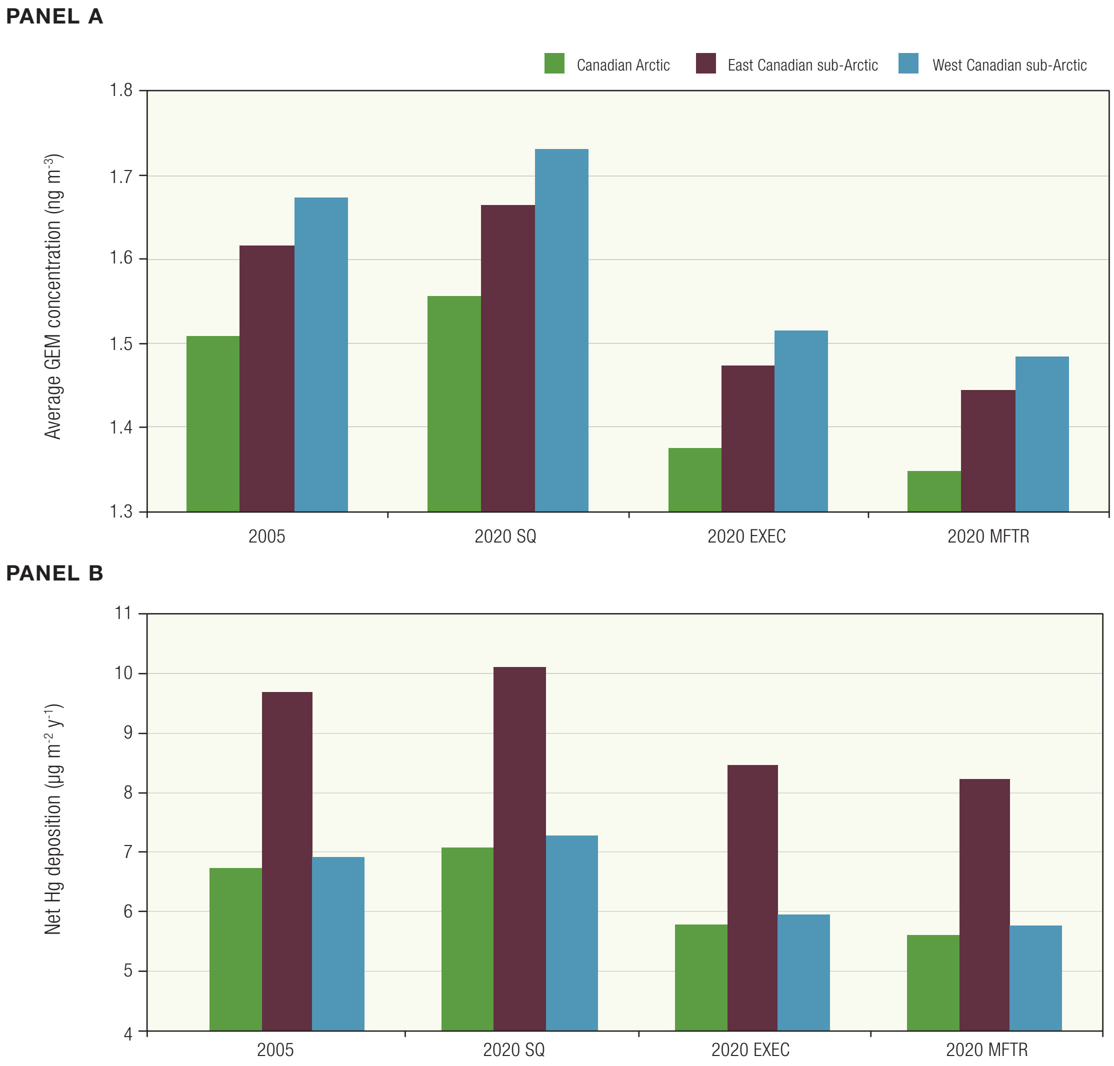

FIGURE 3.21

A) Yearly average concentrations of surface air GEM (ng m^{-3}) simulated in GRAHM in year 2005 and under three future anthropogenic emission scenarios for the year 2020 (SQ, EXEC and MFTR) in the Canadian High Arctic (66.5–90° N) and east (60–66.5° N) and west sub-Arctic (60–66.5° N). B) Simulated yearly net deposition flux (μg m^{-2} y^{-1}) for the same regions (Dastoor 2011).

Lindberg 2005). Mercury is found to exhibit atypical characteristics in environments such as polar regions, the marine boundary layer, and upper troposphere where low concentrations of GEM are observed. Presently, it is not clear whether Hg oxidation pathways related to these observations are limited to the aforementioned environments or are also relevant to the global atmosphere. In addition, there is limited knowledge of the temperature dependence of Hg reactions for the wide range of temperatures in the atmosphere. Laboratory experimental data point to significant heterogeneous Hg chemistry on surfaces that could be relevant in the atmosphere. However, little is known about Hg reactions and equilibrium processes that take place on surfaces. As a result, it is difficult to quantify gas-particle partitioning of Hg in the atmosphere, which leads to uncertainty in model predictions of Hg deposition. Other knowledge gaps related to Hg processes are inadequate knowledge of scavenging characteristics of Hg in liquid and solid condensates, and dry deposition velocities. Uncertainties in model results are also caused by the uncertainties in meteorological parameters.

Given the current knowledge gaps, models rely on observational data to constrain the parameterization of non-anthropogenic emissions and Hg chemical mechanisms by evaluating the model outputs with measurements of Hg species in air and precipitation. Significant progress has been made in reducing the uncertainties in Hg models in the last decade as a result of the increase in measurement data. Long-term surface air measurements of GEM and wet deposition fluxes have been conducted in North America and Europe. Mercury measurements in the marine boundary layer are available from several ocean cruises (Soerensen et al. 2010). A limited number of upper air observations from aircraft measurements have also been used to reduce uncertainties in the model emissions and Hg chemistry. However, a lack of long-term observations outside North America and Europe, limited and uncertain measurements of oxidized Hg species and lack of species identification, vertical profiles, and dry deposition fluxes hinder our ability to fully constrain the Hg model parameters. Environment Canada is currently an external partner in the newly formed Global Mercury Observation System (GMOS) program in which the goal is to establish a global monitoring system that could provide more data in polar regions in addition to temperate areas. Finally, terrestrial and ocean models that include biogeochemical processes of Hg need to be linked to atmospheric Hg models to improve estimates of natural and revolatilized emissions of Hg and more fully assess the impact of changing anthropogenic emissions and climate on Hg deposition in the Canadian Arctic.

3.7 Summary

During Phase III of the NCP, significant advances were made toward understanding the unique processes governing atmospheric Hg transport and fate in the Arctic and sources of Hg to this region. Several of these advances relate to the chemical processes of AMDEs—a phenomenon characterized by rapid depletion of GEM from the lower atmosphere through oxidative processes and subsequent deposition of this Hg on the ground or to the surfaces of particles. It was verified that Br atoms, thought to originate from marine waters, are the most significant oxidants of Hg in the Arctic atmosphere. The speciation of atmospheric Hg changes during springtime AMDEs in the Arctic as GEM is oxidized, leading to large increases in RGM and PHg concentrations. Long-term measurements of Hg species at the High Arctic monitoring station at Alert revealed that the timing of AMDEs has shifted to earlier in the year although the frequency of AMDEs has not changed. The timing of AMDEs was correlated with local temperature, time of year, and wind direction, but these factors alone did not completely account for observed trends and variability of AMDEs. Further study of the factors affecting AMDE chemistry is needed in order to predict their future contributions to Hg deposition in the Arctic.

Field studies on Hg fluxes from the atmosphere to snow indicate that, while a fraction of the Hg deposited during AMDEs is retained in the snowpack, a significant portion is quickly reduced and emitted to the atmosphere. Field and modelling studies suggest that halogen compounds from the marine environment facilitate oxidative and stabilizing reactions with Hg that increase its retention in the snow. Snowpacks rich in halogen species are found near coastal regions, and reactions between Hg and halogens in the snow may in part explain the observation that the extent of Hg re-emission increases with distance from open ocean water. The concentration of THg in seasonal snowpacks is also affected by the burial of Hg with fresh snow. Therefore, snow accumulation of Hg may be higher in regions with higher and frequent snow precipitation. The outcome of these findings is that Hg retention and cycling in the Arctic seems to differ depending on the time of year, proximity to the ocean, and meteorology.

The geographic coverage of air measurements in the Canadian Arctic is still sparse but now includes additional data for Little Fox Lake (Yukon), Churchill (Manitoba) and the Amundsen Gulf (Northwest Territories) to complement the long-term monitoring sites at Alert in the High Arctic and Kuujjuarapik in sub-Arctic Quebec. Annual concentrations of GEM were lower at western Arctic sites of Little Fox Lake, and Barrow (Alaska) and similar to a western temperate site at Whistler (British Columbia). Rapid drops in atmospheric GEM concentration, associated with AMDEs, were observed for the coastal sites at Alert, Barrow, Churchill, and Kuujjuarapik but not for the inland site at Little Fox Lake, which supports the hypothesis that these events occur near marine sources of halogens.

Scientific advances on Hg processes in the atmosphere and exchanges between air, snow, and water were incorporated into global Hg transport models, including Environment Canada's GRAHM model. These models were subsequently used to estimate regional Hg deposition, interpret Hg measurements, and understand past, present, and future sources of Hg to the region. Detailed model simulations using GRAHM for the Alert and Little Fox Lake monitoring sites indicated that Asia likely contributes the most Hg—from anthropogenic, natural, and re-emission sources—to the High Arctic and western Arctic, followed by contributions from North America, Russia, and Europe. Most of the variability in model simulations for both locations was determined by long-range transport of Hg from Asia. Based on model simulations of the short-term dataset from Little Fox Lake and the long-term dataset from Alert, it was concluded that Little Fox Lake is a good location for monitoring Asian long-range transport and Alert is a good location for detecting changes in global anthropogenic Hg emissions.

Using GRAHM, it was estimated that net deposition of Hg above the Arctic Circle (north of 66.5°) occurs at a rate of 143 t yr^{-1}, which indicates that the Arctic is a sink for Hg. Source attribution analyses were performed with the model using Hg emissions data from 2005. In the Canadian Arctic and sub-Arctic regions, global anthropogenic emissions, terrestrial emissions, and oceanic emissions were predicted to have contributed to 20–28%, 40–45% and 30–34% of Hg deposition, respectively. The anthropogenic contribution to Hg deposition was higher in the High Arctic than in the sub-Arctic due to greater contributions to the High Arctic from East Asia. The largest anthropogenic source region with respect to Hg deposition to the Arctic is East Asia followed by Europe, North America, and South Asia. The absolute contribution from East Asia to Hg deposition is slightly higher in the western sub-Arctic region and deposition from North America is slightly higher in the Arctic and eastern sub-Arctic. It was estimated that two thirds of deposition to the Arctic is from natural and re-emitted Hg from land and oceans, which demonstrates the importance of improving our understanding of Hg emissions from these sources.

Air concentrations, measured as GEM, declined from 2000 to 2009 at Alert and Kuujjuarapik. The annual rate of decline at Kuujjuarapik of -2.0 ± 0.9% (95% confidence limits) was comparable to non-Arctic monitoring sites at lower latitudes while the trend for Alert was lower (-0.9 ± 0.6%). A slower rate of decline at Alert may be related to the environmental conditions occurring in the High Arctic or exposure to different pathways of long-range atmospheric transport of Hg. According to modelling analyses in mid-latitude locations of North America, the decline in regional Hg emissions was well reflected by a decline in air concentrations and deposition. However, in the High Arctic, the GRAHM model results suggested that the Hg trends were mostly related to changes in meteorology and global changes in anthropogenic emissions. The changes in meteorology also reflected the impact of climate change between 1990 and 2005 in these simulations. Modelling results showed a slow increasing trend in Hg deposition in contrast to a decreasing trend in surface air GEM concentrations in the Arctic. The increase in net deposition in the Arctic was found to be related to changes in surface conditions such as solar fluxes and temperature. Results from the model and measured GEM concentrations illustrate that Hg trends observed at temperate locations cannot be extrapolated to the Arctic.

If the emissions were to increase along a *status quo* scenario, model simulations projected an average increase in atmospheric Hg deposition of about 5% in 2020 for the Canadian Arctic. Maximum declines in Hg deposition of 18–20% are expected under a scenario where the best control technologies are implemented.

Although tremendous progress has been made toward understanding long-range atmospheric transport of Hg and its deposition in the Arctic, many knowledge gaps still impede our capacity to quantify the significance of this cycling for bioaccumulation of methylmercury in biota and humans. These knowledge gaps include uncertainties in the identification of oxidized Hg species in the air, physical-chemical processes in air, snow and water—especially over the sea ice—and the relationship between these processes and climate change.

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Chapter 4

Terrestrial Environment

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4.1 Introduction

Arctic terrestrial ecosystems support a variety of wildlife including populations of mammals and birds that are important to the culture and traditional diet of northern Aboriginal peoples. Considerable information was collected on Hg in terrestrial mammals and birds during Phases I and II of the Northern Contaminants Program (NCP), with broad geographic coverage across the Canadian Arctic. In general, Hg levels are below levels of concern for wildlife health in large mammals such as caribou, moose, wolf and wolverine (Fisk et al. 2003).

Terrestrial environments are important in the regional Hg cycle because these watersheds supply Hg to freshwater and marine ecosystems. Snow, ice, and soils on land are key reservoirs for atmospheric deposition and, in turn, can become sources of Hg through the melting of the cryosphere (terrestrial ice and snow) and via soil erosion. A major knowledge gap identified in the second *Canadian Arctic Contaminants Assessment Report (CACAR II)* was the lack of information on the fate of Hg following atmospheric deposition, particularly from atmospheric mercury depletion events (AMDEs) (Bidleman et al. 2003). Further study was recommended in order to understand the relationship between abiotic Hg deposition in snow and its bioaccumulation in marine, freshwater, and terrestrial food webs (Bidleman et al. 2003).

Since the *CACAR II*, several important advances have been made regarding the fate of Hg in the terrestrial environment. New data have been collected for snow and ice that provide more information on the flux of Hg to the cryosphere and its post-depositional fate. The monitoring of caribou and moose continued during Phase III of the NCP and included more frequent sampling of caribou to increase the statistical power of temporal trend monitoring for this key terrestrial

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indicator species. Vegetation was also measured for Hg levels to provide information on uptake at the base of Arctic terrestrial food webs.

4.2 Overview of the Hg cycle in the terrestrial environment

Mercury enters the Arctic terrestrial environment primarily as divalent inorganic Hg [Hg(II)] via two main pathways: atmospheric deposition and geological weathering (Fig. 4.1). Methylmercury (MeHg) is also deposited from the atmosphere although this flux is probably small relative to inorganic Hg (see section 4.3.3). The Hg(II) in snow, ice, and soil can have several fates: 1) reduction to elemental Hg [Hg(0)] and subsequent evasion to the atmosphere; 2) microbial methylation in soils and possibly also in snow; 3) transport to aquatic ecosystems via snowmelt, surface runoff, and erosion; and, 4) uptake in vegetation. The current state of knowledge on these complex processes is presented in more detail throughout this chapter.

Mercury enters terrestrial food webs through uptake in vegetation. Dominant vegetation types vary among the diverse terrestrial landscapes in the Canadian Arctic that include barren polar desert, lush tundra meadows and taiga forest (Prowse et al. 2009). Lichens, which are an important food for many Arctic herbivores, have no root system and accumulate Hg mainly from the atmosphere (Estrade et al. 2010). Below the tree line, foliage of trees and shrubs also accumulates Hg mainly from atmospheric deposition directly onto leaves rather than through uptake by roots (Grigal 2002, Ericksen et al. 2003, Graydon et al. 2009). Trees and shrubs play an important role in scavenging Hg and increasing its flux to snow and soils via throughfall and litterfall (Poulain et al. 2007b, Graydon et al. 2008).

4.3 Ice and snow

4.3.1 Concentrations of Hg in Arctic terrestrial snow

Since the discovery of AMDEs in the late 1990s (Schroeder et al. 1998), considerable research has focused on Hg in the Arctic cryosphere because snow and ice are key receptors for atmospheric deposition, and chemical processes occurring at the snow-air interface affect how much Hg enters the environment. The amount of Hg retained in snow and ice reflects, in part, where it is found (St. Louis et al. 2005, Poulain et al. 2007a). Therefore, this assessment presents descriptions of Hg in the cryosphere under both the terrestrial and marine chapters of the report. Concentrations of Hg in lowland terrestrial snow are summarized in this section, while glacier snow and ice

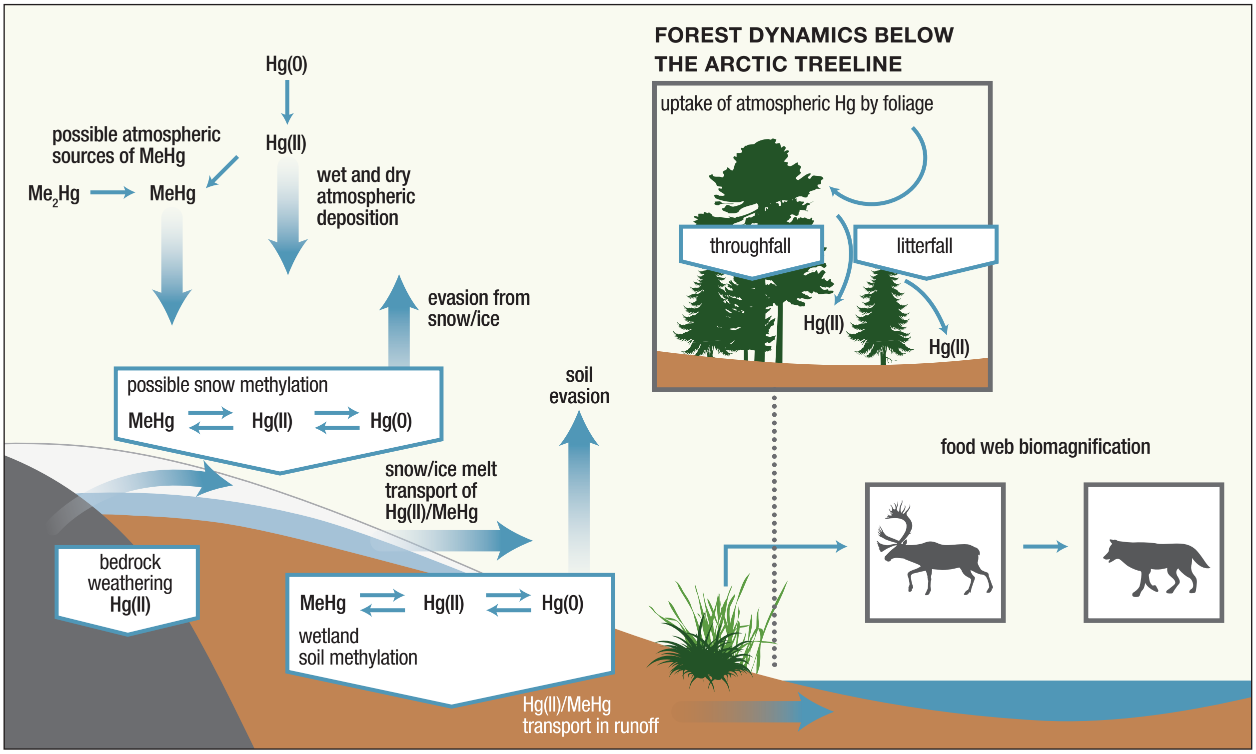

FIGURE 4.1

Schematic of major transformations and pathways in the Hg cycle for the Arctic terrestrial environment.

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are discussed separately in subsequent sections 4.3.2 to 4.3.4. Arctic marine snow and ice are discussed in section 6.4.

Detailed investigations of Hg in terrestrial snow—excluding glaciers—have mainly been conducted at three sites in the Canadian Arctic: Resolute on Cornwallis Island, Alert on Ellesmere Island, and Kuujjuarapik in northern Quebec. These studies revealed that snow total Hg (THg) concentrations can range three orders of magnitude from 0.3 ng L^{-1} to as high as 156 ng L^{-1} at Arctic sites (Fig. 4.2). Although maximum observed concentrations were elevated, average concentrations, determined for three of the studies, were relatively low (< 5 ng L^{-1}).

The high variability in THg concentrations reflects complex processes and transformations within the snow pack. Freshly deposited Hg on snow is highly reactive (Lalonde et al. 2002), and a large portion of the Hg deposited during AMDEs is quickly returned to the atmosphere because of the photoreduction of Hg(II) to gaseous elemental Hg (GEM) within the surface snowpack (Lalonde et al. 2002, Poulain et al. 2004). The presence of halogens in snow may act to retain Hg (section 6.4), and indeed, elevated snow Hg concentrations are more often found in coastal environments where deposition of sea salt can be expected (Douglas and Sturm 2004, Poulain et al. 2007a, St. Louis et al. 2007). Conversely, these higher concentrations may occur primarily because AMDEs are a result of photochemical reactions involving marine halogens and ozone (section 3.2). Burial through snow accumulation, sublimation, condensation, and ice layer formation are other processes that promote retention of Hg in the snowpack (Douglas et al. 2008).

While deposition occurs at the snow surface, Hg can also be redistributed within the snow accumulation. Migration of GEM within the snowpack and subsequent oxidation can result in accumulation of Hg within deeper layers (Dommergue et al. 2003b, Fäin et al. 2006). Melt events may redistribute Hg towards deeper layers away from the high energy surface of the snowpack. Concentrations are commonly highest in the surface layer (Dommergue et al. 2003a, St. Louis et al. 2007), but the middle stratum or depth hoar—a layer of large crystals at the base of the snowpack—can also be dominant reservoirs of Hg in some settings. Therefore, post-depositional processes over the entire depth of the snowpack may influence Hg accumulation.

Less information is available for MeHg in terrestrial snow. Based on studies from Cornwallis Island, Alert, and Kuujjuarapik, snow MeHg concentrations generally range from below detection limit (around 0.02 ng L^{-1}) to 0.2 ng L^{-1} (Dommergue et al. 2003a, Lahoutifard et al. 2005, St. Louis et al. 2005, 2007) but can reach as high as 0.7 ng L^{-1}, as observed in Kuujjuarapik (Constant et al. 2007). Average concentrations on Cornwallis Island, at Kuujjuarapik, and at Alert were < 0.1 ng MeHg L^{-1} (Dommergue et al. 2003a, Lahoutifard et al. 2005,

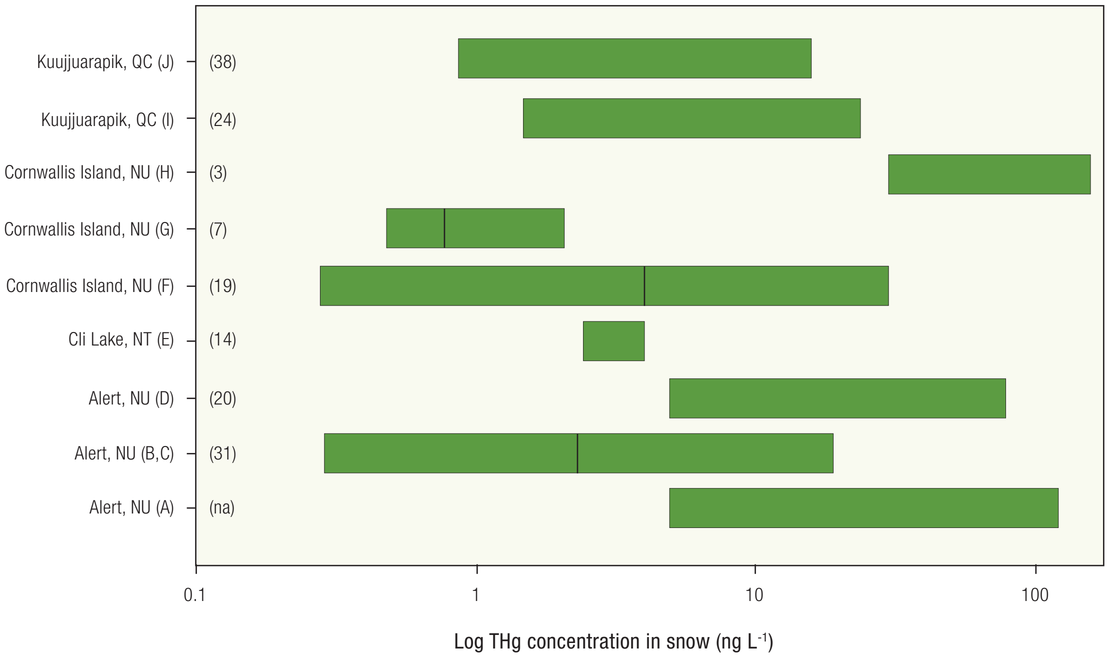

FIGURE 4.2

Ranges of THg concentrations measured in snow from terrestrial sites in the Canadian Arctic (excluding glaciers). Concentrations are presented on a logarithmic scale. Where available, mean concentrations are represented by a line in the bar, and sample sizes are provided in parentheses. References: A = Steffen et al. (2002); B = St. Louis et al. (2005); C = St. Louis et al. (2007); D = Cobbet et al. (2007); E = Evans and Lockhart (1999); F = Lahoutifard et al. (2005); G = Poulain et al. (2007a); H = Lu et al. (2001); I = Dommergue et al. (2003a); J = Constant et al. (2007).

St. Louis et al. 2005, 2007). As with THg, MeHg concentrations also vary with depth in terrestrial snowpack, and the middle stratum or depth hoar were sometimes found to have a higher concentration than the surface (Dommergue et al. 2003a, St. Louis et al. 2005, 2007). The origin of MeHg in terrestrial Arctic snow has not been determined, and several mechanisms for its production have been proposed (section 4.5.2).

4.3.2 Glaciers as natural archives of atmospheric Hg deposition

Glacier snow and firn offer several advantages for monitoring atmospheric Hg deposition compared to other environmental matrices. First, any Hg found in snow is almost exclusively from the atmosphere, provided samples are collected in the accumulation zone where contributions from local contamination sources are minimized. However, this is not the case for lake sediments, for example, where Hg can also be supplied from catchment drainage. Second, the accumulation zone of glaciers preserves a temporal sequence of net atmospheric Hg deposition that can be reconstructed by the analysis of cores. The degree of preservation of the sequence and the temporal resolution that can be attained depend on the snow accumulation and summer surface melt rates at the sampling site. Third, chemical, physical, or biological change—termed diagenesis—is limited for non-volatile components due to the nature of ice.

Firn is snow that has survived at least one summer without being converted to ice. It is the intermediate state between seasonal snow and glacier ice, and its density increases with depth of burial from approximately 0.40 g cm^{-3} to that of glacier ice (0.92 g cm^{-3}). In many glaciers, firn layers are interspersed with discrete layers or lenses of infiltration ice, which are formed by the refreezing of surface meltwater in the firn during summer months. Studies at Summit (central Greenland) have shown that dynamic, photochemically-driven exchanges between air and snow affect the distribution of Hg in glacier firn and interstitial air down to 2 m. However, below this depth, Hg(0) levels in firn air are close to mean atmospheric concentration, and the THg concentration in firn layers shows little variability (Fäin et al. 2006, 2008). Thus, short-term changes—hourly to seasonal—in the Hg content of the snowpack and firn are smoothed out at depths greater than 2 m. On Arctic glaciers with steeper densification gradients and lower firn permeability, this depth limit is probably shallower. It is possible to estimate the net annual deposition of Hg from the atmosphere using firn cores by integrating at least five years of snow and firn accumulation, which on most glaciers exceeds 2 m. Averaging over depth also compensates for some post-depositional effects such as elution and redistribution by summer surface melt that can impact the firn down to several metres on sub-polar glaciers.

It should be noted that net atmospheric Hg accumulation rates in glacier firn are not necessarily representative of all snow-covered Arctic landscapes. Glacier sampling sites are often tens to hundreds of kilometres from the nearest coastline and greater than 1–3 km above the marine atmospheric boundary layer—the part of the atmosphere that has direct contact with the ocean. Yet, some of the processes governing the transfer and retention of atmospheric Hg in snow over coastal and marine areas, such as AMDEs, are confined to approximately the lowermost 1 km of the troposphere (Steffen et al. 2008). Nevertheless, glacier snow and ice are unique in providing the opportunity to infer spatial and temporal trends in atmospheric Hg deposition over the Arctic in the absence of long-term instrumental records of accumulation in snow.

4.3.3 Spatial trends of Hg in glacier ice and snow

Concentrations and fluxes of THg

Snow and firn were investigated on glaciers in Arctic Canada and Greenland to document spatial trends in atmospheric Hg deposition under NCP and International Polar Year initiatives. Snow and firn samples were collected from several glacier sites along a north-to-south transect in the northeastern Canadian Arctic, and from the location of the international NEEM (North Greenland Eemian) deep ice-coring project in northern Greenland (Fig. 4.3). In the following, results are presented for sites where a suite of samples covering at least eight consecutive years of deposition were obtained or where such information was found in published studies. The sites are NEEM (northern Greenland), Summit (central Greenland; Mann et al. 2005), Agassiz Ice Cap and Mount Oxford Icefield (northern Ellesmere Island), Devon Ice Cap (Devon Island), and Penny Ice Cap (Baffin Island). To augment the spatial coverage, data were added from Prince of Wales Icefield and John Evans Glacier on central Ellesmere Island (St. Louis et al. 2005) even though they do not satisfy the requirement for multi-year averaging. A spatial comparison was conducted using mean Hg deposition over approximately the last decade. The possible error on the estimated time interval represented by the snow and firn sample suites is ≤ 1 year, based on Monte-Carlo simulations. Temporal variations in Hg deposition reconstructed from snow and firn cores are discussed separately in section 4.3.4.

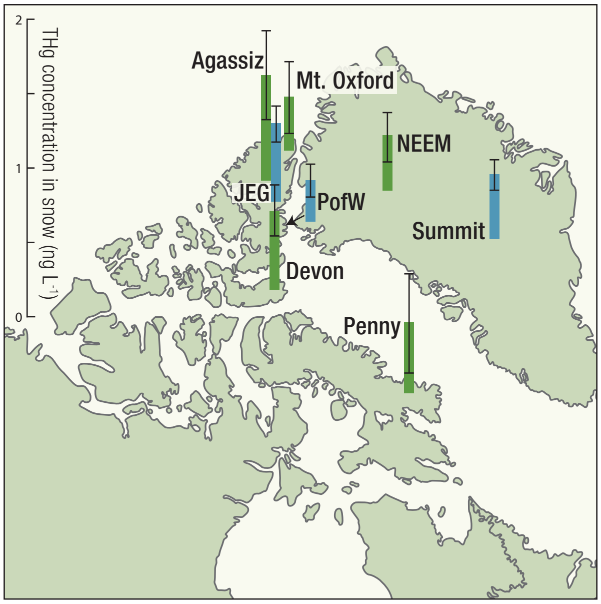

FIGURE 4.3

Mean concentrations of THg in snow and firn layers on Canadian Arctic glaciers and the Greenland Ice Sheet. Green bars are data from Zdanowicz and Zheng (2010) and represent averages computed for at least eight years of accumulation in the past decade. Blue bars are based on data from St. Louis et al. (2005) for Ellesmere Island (JEG = John Evans Glacier, PofW = Prince of Wales Icefield) and Mann et al. (2005) for central Greenland. Error bars are ± 1 standard deviation. See Table 4.1 for details.

Mean THg concentrations in snow from different glacier sites varied within a relatively narrow range of 0.31–0.84 ng L^{-1}, and the average for all sites was 0.54 ± 0.09 ng L^{-1} (Fig. 4.3). The lowest mean Hg concentrations (≤ 0.5 ng L^{-1}) were found in northern Greenland and on Ellesmere Island at Prince of Wales and Mount Oxford icefields. Surprisingly, the highest mean value (0.84 ng L^{-1}) was also found on northern Ellesmere Island at Agassiz Ice Cap. Given the variability of data at each site (standard deviation > 20% of the mean), the observed differences cannot be regarded as statistically meaningful. Hence, it would appear that over the past decade the mean THg concentration in snow deposited on Arctic glaciers has been relatively uniform in space over a broad area encompassing central and northern Greenland, and the Canadian Arctic Islands.

Observed THg concentrations in glacier snow and firn (< 1 ng L^{-1}) were much lower than those reported in other snow surveys of marine or lowland terrestrial environments (sections 4.3.1 and 6.4). Concentrations in spring surface snow at coastal and marine Arctic sites are often an order of magnitude higher and can exceed 100 ng THg L^{-1}. Elevated concentrations in surface snow in spring result from AMDEs, but as several studies have shown (e.g., Kirk et al. 2006), much of this Hg is subsequently re-emitted back to the atmosphere, such that the apparent disparity with Hg measurements from glacier firn may not be as large as it seems.

For each of the glacier sites in Table 4.1, the average net accumulation rate of THg (F_{THg}) in snow over the recent past was calculated as:

$$F_{THg} = [THg] \cdot \lambda$$

The flux estimates represent the net accumulation of THg in firn after any post-depositional modifications have taken place (e.g., by photolytic re-emission). The net snow accumulation rate (λ) for recent decade(s) was obtained from published studies or was estimated using standard methods, specifically seasonal indicators (e.g., oxygen stable isotopes) and/or reference markers in firn cores such as the 1963 peak in snow radioactivity. The mean value of net snow accumulation at each of the glacier sites was used, and monitoring data as well as ice-core studies indicate that the mean rate has not varied meaningfully over recent decades at most of the sites concerned. An exception is the Agassiz Ice Cap summit, where snow accumulation in recent years was affected by drifting. This factor is accounted for in the calculations.

A north-to-south gradient in Hg flux was found, with F_{THg} increasing from < 0.1 µg m^{-2} y^{-1} north of 80° (Agassiz, Mount Oxford) to ≥ 0.2 µg m^{-2} y^{-1} on Penny Ice Cap (67° N) (Table 4.1; Fig. 4.4). The mean F_{THg} for all sites was 0.11 ± 0.02 µg m^{-2} y^{-1}. In absolute terms, these geographic differences are small and the uncertainties large, yet, at the southernmost latitude, the highest value of F_{THg} is twice the lowest and northernmost flux. The apparent latitudinal variation in F_{THg} generally follows the gradient of snow accumulation rate (λ) across Canadian Arctic glaciers, with λ increasing at lower latitudes. This trend suggests that relative to dry deposition, the wet deposition contribution to F_{THg} may increase at lower, wetter latitudes. Another possibility is that both wet and dry THg deposition increase from north to south, as might be expected if part of the Hg deposition is from southern pollution sources. It is noteworthy that sulphate accumulation trends on Canadian Arctic glaciers follow a spatial pattern similar to that of F_{THg}, and that pattern has been linked to southerly advection of polluted air incursions from the eastern United States and Canada (Goto-Azuma and Koerner 2001).

The THg distributions in snow and firn profiles from various Arctic glaciers have shown that the enrichment of THg in winter snow layers which could be attributed to AMDE deposition is confined to the top few centimetres of snow and is highly variable in time and space between sites. Although THg concentrations in these near-surface snow layers are often much higher than in underlying strata, their contribution to the THg burden stored annually in firn is comparatively small. At least half, and typically more, of the THg that ends up in firn storage is actually found in the underlying summer or autumn snow layers, which usually make up most of the total annual snow accumulation. On Penny Ice Cap, for example, where $\lambda = 0.34$ m y^{-1}, the THg in late winter or spring snow layers amounts to < 25% of the annual burden (Zdanowicz et al. 2010). Snow samples collected on the ice cap in summer (June) also had a mean THg concentration of 1.2 ± 0.3 ng L^{-1}, which was twice the mean concentration found in winter snow layers. If melting resulted in some elution into the snowpack, June snow samples may have underestimated the THg concentration in surface snow. At this site, and possibly at others in the same latitude, atmospheric Hg accumulation is therefore primarily associated with precipitation in the warmer and moister half of the year. The contribution of AMDE-related THg deposition to glacier snow may be larger at the colder and drier northernmost sites such as Agassiz Ice Cap.

Glacier estimates of F_{THg} were compared to annual Hg accumulation rates inferred from other natural media reported in the literature, in particular lake sediments and peat (Table 4.2). Present-day net Hg accumulation in glacier firn—which is almost exclusively from atmospheric sources—is lower, by one or two orders of magnitude, than net accumulation rates in lake sediments and peat, even when the latter are corrected for factors such as catchment size or sediment focussing. Net Hg accumulation rates are strongly dependent on the relative retention capacity of the medium, with peat retaining the most and glacier snow and firn retaining the least.

TABLE 4.1 Concentrations and mean annual accumulation rates of THg and MeHg in glacier snow and firn. Data are from Zdanowicz and Zheng (2010) unless otherwise stated.

THg										
Region/Site		**Lat. (°)**	**Elevation (m)**	**Sampled Medium**	**Interval (y)**	***n***	**[THg] μ ± σ (ng L^{-1})**	**λ (m y^{-1})**	**F_{THg} μ ± σ (μg m^{-2} y^{-1})**	
Greenland										
	NEEM	77	2,447	snow pit + firn core	8	51	0.42 ± 0.20	0.26	0.11 ± 0.05	
	Summit[1]	73	3,240	firn core	13	35	0.51 ± 0.12	0.20	0.10 ± 0.01	
Canada										
Ellesmere Island										
	Mount Oxford Icefield	82	1,680	snow pit	8	55	0.42 ± 0.29	0.19	0.08 ± 0.06	
	Agassiz Ice Cap	81	1,670	snow pit + firn core	8	44	0.84 ± 0.36	0.10	0.08 ± 0.04	
	John Evans Glacier[2]	79	N/A	seasonal snow	< 1	15	0.60 ± 0.14	0.15	0.09 ± 0.04	
	Prince of Wales Icefield[2]	78	N/A	seasonal snow	< 1	29	0.31 ± 0.12	0.30	0.09 ± 0.04	
Devon Island										
	Devon Ice Cap	77	1,800	firn core	10	38	0.63 ± 0.21	0.24	0.15 ± 0.05	
Baffin Island										
	Penny Ice Cap	67	1,860	snow pit + firn cores	12	145	0.72 ± 0.52	0.34	0.24 ± 0.18	
MeHg										
Region/Site		**Lat. (°)**	**Elevation (m)**	**Sampled Medium**	**Interval (y)**	***n***	**[MeHg] μ ± σ (ng L^{-1})**	**λ (m y^{-1})**	**F_{MeHg} μ ± σ (μg m^{-2} y^{-1})**	**THg as MeHg μ ± σ (*n*) (%)**
Greenland										
	NEEM	77	2,447	snow pit + firn core	8	51	0.04 ± 0.21	0.26	0.01 ± 0.05	8.3 ± 6.2 (45)
Canada										
Ellesmere Island										
	John Evans Glacier[2]	79	~ 100–1,100	seasonal snow	< 1	15	0.06 ± 0.01	0.15	0.01 ± 0.05	19.6 ± 3.7 (15)
	Prince of Wales Icefield[2]	78	~ 200–1,800	seasonal snow	< 1	29	0.09 ± 0.05	0.30	0.03 ± 0.05	38.3 ± 24.4 (29)
Devon Island										
	Devon Ice Cap	77	1,800	firn core	10	38	0.02 ± 0.01	0.24	< 0.01	3.3 ± 2.4 (16)
Baffin Island										
	Penny Ice Cap	67	1,860	snow pit + firn cores	12	12	0.11 ± 0.07	0.34	0.04 ± 0.02	19.6 ± 13.6 (132)

[1] Summit data from Mann et al. (2005).

[2] Mean THg concentrations calculated from data in St. Louis et al. (2005). Annual deposition rates calculated from THg concentrations and mean snow accumulation rates reported in Sharp et al. (2002) and Kinnard et al. (2008). The derived values of F_{THg} may be underestimated.

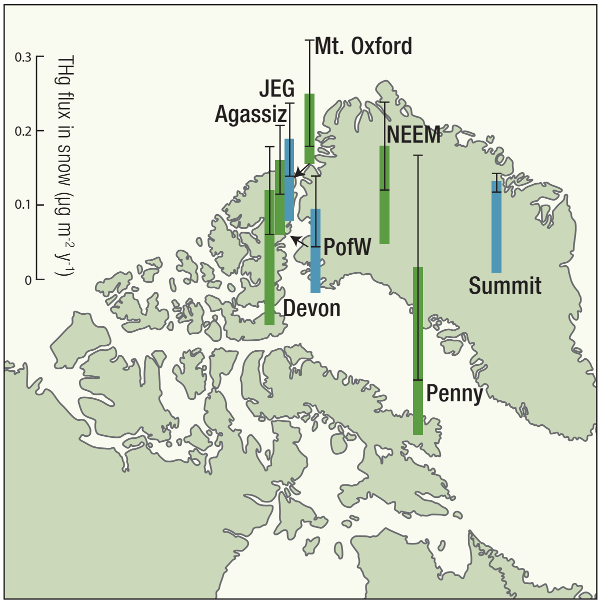

FIGURE 4.4

Mean annual accumulation rates (F_{THg}) for THg in snow and firn layers on Canadian Arctic glaciers and the Greenland Ice Sheet. Green bars are data from Zdanowicz and Zheng (2010) and represent averages computed for at least eight years of accumulation in the past decade. Blue bars are based on data from St. Louis et al. (2005) for Ellesmere Island (JEG = John Evans Glacier, PofW = Prince of Wales Icefield), and Mann et al. (2005) for central Greenland. Error bars are ± 1 standard deviation. See Table 4.1 for details.

Deposition rates of Hg inferred from glacier samples were further compared with limited measurements of direct wet deposition and atmospheric transport model simulations for the Arctic (Table 4.2). Glacier estimates of F_{THg} are much closer to, albeit lower than, measured wet deposition rates measured at latitudes of 58–68° N compared with accumulation rates inferred from peat and sediments at polar latitudes. The comparison with atmospheric transport model simulations shows that the range of model-projected total (wet and dry) Hg deposition rates over the Canadian Arctic islands and Greenland (approximately 2–12 µg m^{-2} y^{-1}) generally exceeds the net deposition rates inferred from glacier firn by at least one order of magnitude, even in simulations that neglect the fast oxidation pathway for Hg(0) associated with AMDEs. Though the conclusion for Greenland seems robust, the usefulness of such comparisons for Arctic Canada is limited by the spatial coarseness of existing models (150 km^2 for DEHM, 1° x 1° for GRAHM, 2.5° x 2.5° for MSCE-Hg) (see Table 4.2 for model descriptions). These findings support the contention of Sanei et al. (2010) that global transport models presently overestimate the total net atmospheric contribution to the Hg budget of the Arctic. Furthermore, this overestimation is not uniquely linked to the representation of AMDEs in the models.

Long-term chronologies of Hg deposition recorded in Arctic environmental archives indicate that Hg fluxes vary by one or two orders of magnitude among different media, namely glacier snow and ice, peat, and lake sediment. In addition, glacier measurements suggest that current atmospheric transport models for Hg may overestimate contributions from the atmosphere to those environments. The accumulation zones of glaciers are special environments, largely lifeless polar deserts, which only represent a small areal fraction of Arctic lands (< 2%), except in Greenland. However, given the scarcity of direct instrumental observations, glaciers provide convenient surrogate estimates of total atmospheric Hg deposition over terrestrial sectors of the Arctic. To develop a more refined Hg mass balance for the Arctic, further research is needed that includes direct measurements of wet and dry deposition from the atmosphere.

Concentrations and fluxes of MeHg

There are less data available for MeHg in glacier ice cores and snow (Table 4.1) than for THg. Consequently, limited inferences can be made on spatial patterns of deposition for this Hg species. Generally, the MeHg concentration in snow and firn varied within a narrow range ≤ 0.1 ng L^{-1}, and the estimated net deposition rates (F_{MeHg}) did not exceed 0.05 µg m^{-2} y^{-1}. High variability was observed at each glacier site (Table 4.1) and there was no clear geographical pattern. However, there were large site-to-site differences in the proportion of THg as MeHg, varying from 3% on Devon Ice Cap to 38% on the Prince of Wales Icefield about 420 km to the north. Potential sources of MeHg in polar snowpacks include atmospheric sources, specifically demethylation of dimethylmercury (Me_2Hg) or abiotic methylation of Hg(II), and/or post-depositional, *in situ* methylation by microbial processes (St. Louis et al. 2005, Larose et al. 2010). In the upper accumulation zone of large polar glaciers, there is typically very little biological activity due to the lack of mineral nutrients to support microbial life (Hodson et al. 2008). Hence, MeHg found

TABLE 4.2 Estimated annual rates of atmospheric Hg deposition in the Arctic from different environmental media and atmospheric models.

Region	Sampled Medium or Model	Period	F_{THg} ($\mu g\ m^{-2}\ y^{-1}$)	References
Annual net Hg deposition rates inferred from environmental sampling media				
Southern Baffin Island (67° N)	Glacier snow and firn	Last ~ 10 years	< 0.3	Zdanowicz and Zheng (2010)
Canadian High Arctic (77–82° N)	Glacier snow and firn	Last ~ 10 years	< 0.1	Zdanowicz and Zheng (2010)
Central Greenland (73° N)	Glacier snow and firn	Last ~ 10 years	~ 0.1	Mann et al. (2005)[1]
Northern Greenland (77° N)	Glacier snow and firn	Last ~ 10 years	~ 0.1	Zdanowicz and Zheng (2010)
Canada (51–64° N)	Lake sediments	Last ~ 10 years	15–39	Muir et al. (2009)
Canada (66–83° N)	Lake sediments	Last ~ 10 years	5–18	Muir et al. (2009)
Greenland (66–67° N)	Lake sediments	Last ~ 10 years	5–10	Bindler et al. (2001)
Alaska (68° N)	Lake sediments[2]	Last ~ 10 years	2–5	Fitzgerald et al. (2005)
Norway (58–69° N)	Peat	Last 100 years	1–7	Steinnes et al. (2005)
Sweden (57–59° N)	Peat	Last ~ 10 years	5–20	Bindler (2003)
Annual gross Hg deposition rates inferred from measured wet deposition				
Churchill (Manitoba) (58° N)	Precipitation	2007–2008	0.5–0.8	Sanei et al. (2010)
Churchill (Manitoba) (58° N)	Precipitation	2006	1.4	Outridge et al. (2008)
Pallas (Finland) (67° N)	Precipitation	1996	2.1	Berg et al. (2001)
Toolik LTER (Alaska) (68° N)	Precipitation	1988	0.9–2.1	Fitzgerald et al. (2005)
Annual Hg deposition rates projected by atmospheric transport models				
Canadian Arctic Islands	GRAHM, with AMDE	2001	1.5–6	Dastoor et al. (2009)
Greenland Interior	GRAHM, with AMDE	2001	0.8–3	Dastoor et al. (2009)
Canadian Arctic Islands	DEHM, with AMDE	2001	9–12	Christensen et al. (2004)
	DEHM, no AMDE	2001	3–6	
Greenland Interior	DEHM, with AMDE	2001	< 3–9	Christensen et al. (2004)
	DEHM, no AMDE	2001	< 3–6	
Canadian Arctic Islands	MSCE-Hg, with AMDE	1996	5–12	Travnikov and Ryaboshapko (2002)
	MSCE-Hg, no AMDE	1996	2–5	
Greenland Interior	MSCE-Hg, with AMDE	1996	< 2–3.5	Travnikov and Ryaboshapko (2002)
	MSCE-Hg, no AMDE	1996	< 2–3.5	

[1] Calculated from data in Mann et al. (2005) as per Table 4.1.

[2] Corrected for soil input.

Note: The approximate ranges of F_{THg} are provided.

GRAHM = Global/Regional Atmospheric Heavy Metals Model

DEHM = Danish Eulerian Hemispheric Model

MSCE-Hg = EMEP Meteorological Synthesizing Centre East model for Hg

AMDE = atmospheric mercury depletion event

in glacier snow and firn collected in the accumulation zone is far more likely to be from atmospheric sources. The Prince of Wales Icefield, where the MeHg:THg ratio was highest, is located next to the North Water polynya, a year-round source of marine aerosols, and a possible source for the high proportion of MeHg found in snow at this site (St. Louis et al. 2005). However, the flux of primary sea-salt aerosols in snow, as represented by chloride ions, was comparable on the Prince of Wales Icefield, Devon Ice Cap, and the Greenland Ice Sheet (Kinnard et al. 2006, Dibb et al. 2010) even though the proportion of MeHg in snow at these sites was strikingly different (Table 4.1). This variation suggests that atmospheric MeHg deposition in snow at these sites is controlled by some other factor(s) than, or in addition to, marine aerosols. Further research is needed to explain the observed geographic differences in the ratio of MeHg to THg in glacier snow and firn.

4.3.4 Temporal trends of Hg in glacier ice and snow

Temporal trends of THg were reconstructed from ice and snow collected on two Canadian High Arctic ice caps: the Mount Oxford Icefield and the Agassiz Ice Cap (Fig. 4.5). Strict protocols were used for field sampling and laboratory processing to avoid potential Hg contamination to snow and ice samples and to ensure analytical accuracy and precision. Age dating was based on multiple parameters, including ion chemistry, accumulation rate, references to volcanic events, and finally linked to chronologies already established in other ice caps in the Canadian High Arctic. Accuracy of age dating for the Mount Oxford samples was estimated at ± 0.5 years.

Ice and snow samples collected from the Mount Oxford Icefield in 2008 contained THg concentrations varying from about 0.25 ng L^{-1} to < 2 ng L^{-1} (Fig. 4.6). The peaks of THg at and near the surface may reflect the effect of AMDEs during polar spring. Those higher concentrations of THg may be unstable and gradually revolatilize to the atmosphere over time (Ferrari et al. 2008, Johnson et al. 2008). Higher THg concentrations in surface and near-surface snow have also been observed at other glaciers in the Canadian High Arctic including the Devon Ice Cap and the Grise Fiord Glacier, and at sites in Svalbard, Norway (Fäin et al. 2006, Zdanowicz et al. 2010). In sub-Arctic Churchill (Manitoba), this phenomenon was found to be even more severe where over 100 ng L^{-1} of THg was measured in surface snow during the AMDE period (Kirk et al. 2006). However, elevated Hg concentrations in surface snow are not typically observed in non-AMDE areas at lower latitudes (Lu et al. 2001, Lalonde et al. 2002).

The Mount Oxford snow pit showed a broad peak from the mid-1990s to around 2005 for samples below 2 m from the surface (Fig. 4.6). Calculated THg fluxes confirmed this broad peak (Fig. 4.6), suggesting higher atmospheric deposition during that period. Given the strict sampling and analytical protocols used during the study, this unusual temporal trend is not likely an artefact. To confirm this broad peak and explore the geographic extent of this pattern, a longer THg archive was collected the following year in 2009 from the nearby Agassiz Ice Cap where the altitude is similar but the precipitation rate is much lower.

Short-core samples to 15.5 m depth from Agassiz Ice Cap contained THg concentrations that were almost twice as high as those in Mount Oxford pit samples (Fig. 4.6). However, both archives exhibited the same broad peak located between the mid-1990s to about 2005. Furthermore, surface and near-surface samples contained higher THg concentrations in both archives. These similarities suggest that: 1) the broad peak between 1995 and 2005 is not an artefact; and, 2) transient, higher THg concentrations exist in surface and near-surface snow layers because of AMDEs and require a certain time period for re-emission back to the atmosphere. The difference in THg concentrations between the two ice fields is likely related to the rate of precipitation.

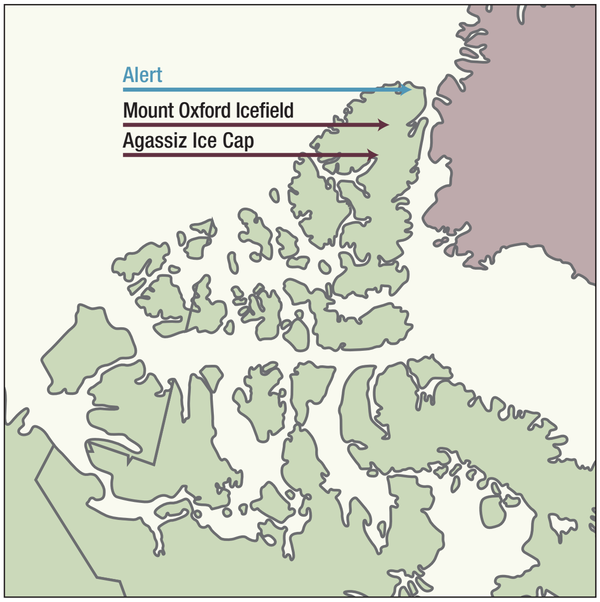

FIGURE 4.5

Locations of the Mount Oxford Icefield (82.2° N and 72.7° W; 1,680 m above sea level) and the Agassiz Ice Cap (80.7° N and 73.1° W; 1,670 m above sea level) where ice cores were collected for temporal Hg trends. The GEM monitoring station run by Environment Canada at Alert is about 100 km north of Mount Oxford Icefield.

Average concentrations and fluxes of THg in snow and ice at Agassiz and Mount Oxford were calculated excluding the unstable surface and near surface layers above 2 m. For Agassiz Ice Cap, the average concentration and flux over 55 years were 0.66 ± 0.46 ng L^{-1} and 0.063 ± 0.27 µg m^{-2} y^{-1}, respectively (n = 164). For Mount Oxford Icefield, the average concentration and flux over 20 years were 0.38 ± 0.17 ng L^{-1} and 0.073 ± 0.023 µg m^{-2} y^{-1}, respectively (n = 118). The mean THg concentrations and fluxes, averaged over more than 20 years, are 10–25% lower than the values calculated for the most recent eight years (Table 4.1),

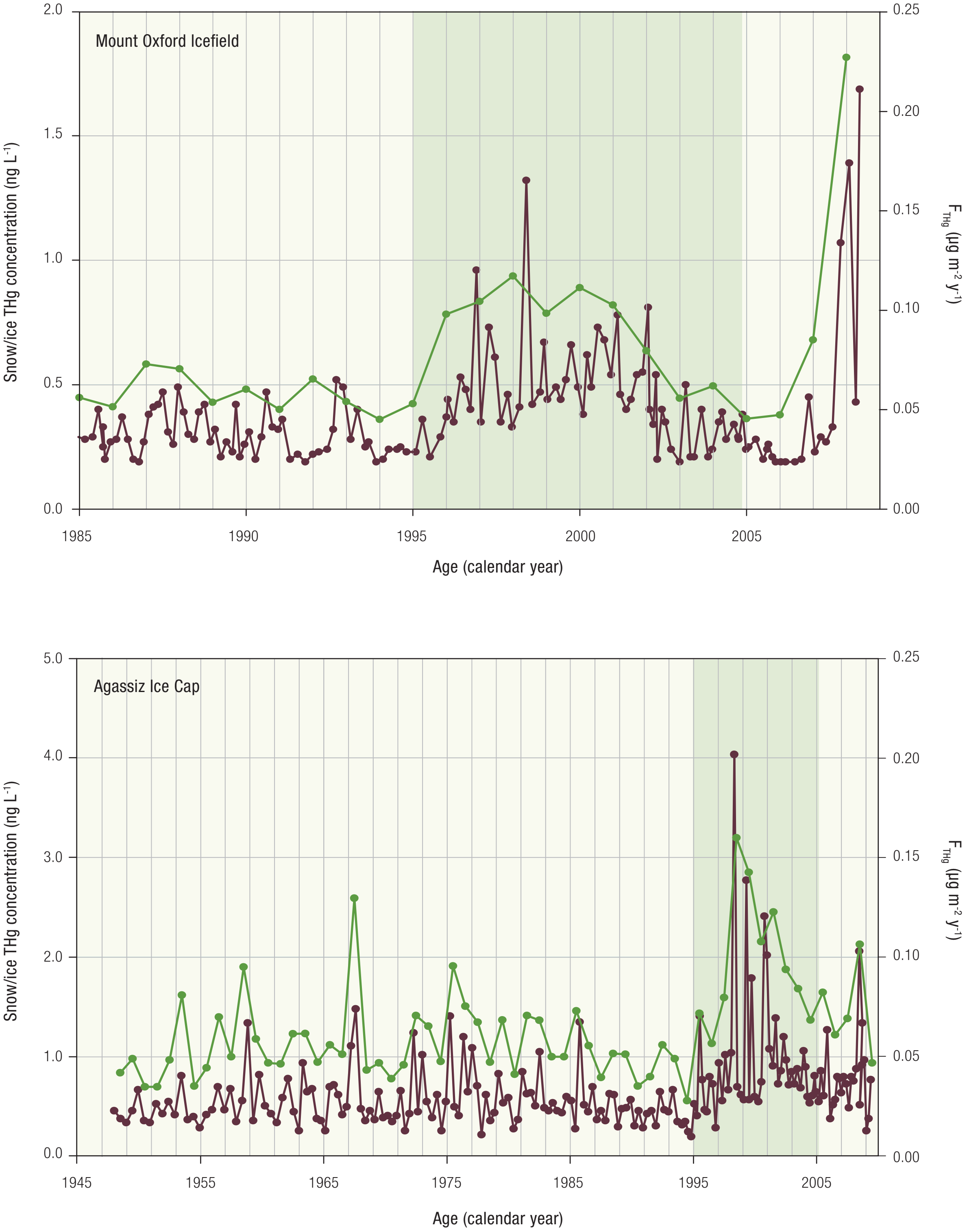

FIGURE 4.6

Temporal distribution of THg reconstructed in ice and snow samples over a ~20-year period at the Mount Oxford Icefield (upper panel) and a ~60-year period at Agassiz Ice Cap (lower panel) (Zheng 2010). A broad peak in concentration (dark purple line) and flux (green line) between 1995 and 2005 is highlighted with a dark green background.

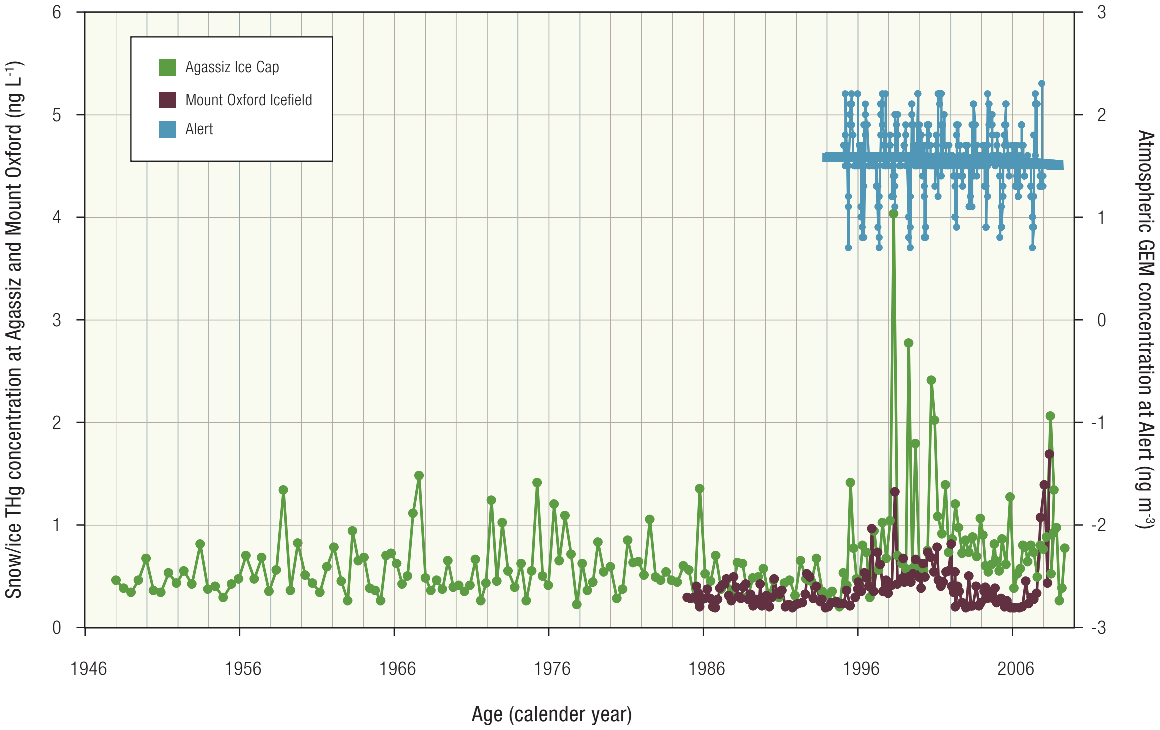

FIGURE 4.7

Comparison of temporal trends of THg concentrations in ice and snow at Agassiz Ice Cap and Mount Oxford Icefield (Zheng 2010) with direct measurements of atmospheric GEM at Alert by Environment Canada.

which probably reflects the influence of higher Hg deposition rates already noted in the interval from approximately 1995 to 2005. Altogether, the long-term mean THg concentration and flux in firn and ice at Agassiz Ice Cap and the Mount Oxford Icefield are comparable to those reported at other Arctic glacier sites (Table 4.1) and are similarly low compared with other environmental archives of THg, namely peat and lake sediment (section 4.3.3; Table 4.2).

The most extensive atmospheric monitoring of Hg in the Canadian Arctic has been carried out at Alert by Environment Canada for almost two decades (see Chapter 3). There is clear evidence from the glacier archives of large decadal variation in net deposition while only a minor declining trend of -0.9% per year was observed with direct monitoring of atmospheric GEM (Fig. 4.7; section 3.5). The lack of correspondence may be due to a difference in what is being measured by these two approaches: the air record represents the atmospheric concentration of GEM while the temporal distribution in ice and snow reflects the net deposition of THg from the atmosphere (except the surface and near-surface layers of snow). This inconsistency between measurements highlights the need for more research using a variety of methodological approaches in order to better quantify temporal trends in net contributions of atmospheric Hg to the Arctic environment.

4.4 Arctic soils

4.4.1 Concentrations of Hg in soil, peat, and permafrost

Few published studies currently exist on Hg in Canadian Arctic soils. The most comprehensive investigations were conducted on wetland soils of Cornwallis Island (Loseto et al. 2004) and wet sedge tundra on Devon Island (Oiffer and Siciliano 2009). Wetland soils typically had low THg concentrations, averaging 46 ng g^{-1} and ranging from 10–250 ng g^{-1} (Loseto et al. 2004). The proportion of THg as MeHg was also low (< 1%), and MeHg concentrations averaged 0.07 ng g^{-1}. Higher MeHg concentrations ranging from 0.34–3.07 ng g^{-1} were observed in sedge meadow soils on Devon Island (Oiffer and Siciliano 2009). In those meadows, the soil was monitored for temporal trends and a decline in MeHg was found during the summer study period.

Coastal permafrost soils were investigated along the Beaufort Sea by Leitch (2006). Cores of permafrost were collected at Tuktoyaktuk in the Northwest Territories and on Herschel Island, King Point, and Komakuk Beach along the Yukon coast. Concentrations of THg ranged from 26–303 ng g^{-1} at different depths in sectioned permafrost cores, and median values ranged from 61–114 ng g^{-1} for each core. These concentrations were generally higher than the THg in frozen peat cores from Bathurst Island, which averaged < 50 ng g^{-1} (Givelet et al. 2004). The flux of Hg from coastal erosion of permafrost soils is a significant source to the Arctic Ocean (Outridge et al. 2008; section 6.6.1).

4.4.2 Historical trends of Hg recorded in High Arctic peat

Peat deposits from Bathurst Island (Nunavut) were investigated for their long-term record of pre-anthropogenic Hg accumulation (Givelet et al. 2004). Two cores were collected from peat hummocks, and thinly sliced sections were analyzed for THg content and dated using ^{14}C measurements. The peat profiles indicated a relatively constant accumulation rate of THg of 0.5–1.5 µg m^{-2} y^{-1} across a long time frame of 5,900 to 800 calibrated years before present. Distributions of conservative lithogenic elements in the cores were not correlated with the Hg profiles, suggesting that atmospheric deposition rather than mineral dust inputs was the source of Hg accumulation (Givelet et al. 2004). A pre-anthropogenic atmospheric deposition rate of approximately 1 µg m^{-2} y^{-1} (inferred from the peat cores) is consistent with historical fluxes observed in peat from Greenland and also from bogs at temperate latitudes (Givelet et al. 2003, Shotyk et al. 2003). The authors of the Bathurst Island study concluded that, before the Industrial Era, the Arctic was not a more important sink for global atmospheric Hg than temperate latitudes because of the similarity in Hg accumulation rates in peat from those two zones. It should be noted that the use of Arctic peat as an environmental archive of Hg deposition is controversial because of potential effects of annual freeze-thaw cycles on the long-term stability of the accumulated Hg—see Bindler et al. (2005) and Shotyk et al. (2005) for more detailed discussion.

4.5 Biogeochemical cycling of Hg

Snow, ice, and soil on land are important abiotic sinks for atmospheric Hg deposition which, in turn, may transfer Hg to freshwater and marine ecosystems. Mercury can undergo several biogeochemical transformations within these matrices that influence its fate (Fig. 4.1). The main reactions are the oxidation or reduction of inorganic Hg and the formation or decomposition of MeHg. The reduction of Hg(II) to Hg(0) is a transformation of fundamental importance because of its role in emitting Hg back into the atmosphere. Microbial methylation is also a key process that transforms Hg(II) into its more toxic organic form.

4.5.1 Mercury reduction

Fluxes of Hg to the atmosphere from snow, ice, and soil are driven by Hg reduction processes. Photoreduction, which is the main mechanism thought to drive Hg emissions from snow and ice, is discussed in the atmospheric chapter because of its importance following AMDEs (section 3.3). In soil, Hg(II) originating from atmospheric deposition and geological weathering may also undergo reduction and subsequent evasion to the atmosphere. This section focuses on Hg reduction in soils.

There is currently little information on Hg reduction and evasion from Canadian Arctic soils. In temperate regions, Hg(II) reduction may be driven by sunlight penetrating into the top few millimetres of soil, by the presence of abiotic reductants such as humic and fulvic acids, or by reactions mediated by microbes (Gabriel and Williamson 2004). Reduction of Hg(II) is important under low redox conditions in lower soil horizons or water-saturated soils (Gabriel and Williamson 2004, Obrist et al. 2010). Sunlight intensity, temperature, and soil moisture are other environmental factors that affect the rate of Hg(0) evasion from soil (Carpi and Lindberg 1998, Schlüter 2000). Oxidation of Hg(0) by abiotic or microbial processes may reduce net evasion from soil. However, this aspect of the Hg biogeochemical cycle is also poorly resolved.

Estimates of Hg(0) evasion from Canadian Arctic soils are not available. Limited data exist for moss-covered substrates and bedrock at two sites: 1) MacMillan Pass (Yukon), where the shale is naturally-enriched in Hg; and, 2) Kuujjuarapik (Quebec), which has background levels of geological Hg (Schroeder et al.

2005). Emissions of gaseous Hg from moss-covered mercuriferous shale in Yukon (mean: 1.5 ng m^{-2} h^{-1}, range: -0.60–10.63 ng m^{-2} h^{-1}) were considerably higher than from moss-covered substrate near Kuujjuarapik (mean: 0.08 ng m^{-2} h^{-1}, range: -0.58–1.3 ng m^{-2} h^{-1}). Gaseous Hg emissions from exposed mercuriferous shale were even higher (9.1–213.5 ng m^{-2} h^{-1}). On average, 0.44 ng m^{-2} h^{-1} was emitted from exposed sandy substrates in Kuujjuarapik. Measurements made at various sites across Canada, mostly south of 60° N latitude, revealed a positive correlation between the flux of gaseous Hg to the atmosphere and the THg concentration of the mineral substrate (Schroeder et al. 2005).

Mechanisms of Hg reduction and estimates of evasion from terrestrial substrates are significant knowledge gaps in the Arctic Hg cycle. Minor emission rates might be expected from Arctic soils because of low temperatures, low solar radiation, and low microbial activity. However, climate change may impact many of the yet unidentified environmental conditions that influence soil Hg(II) reduction. Terrestrial ecosystems in the Canadian Arctic represent a vast surface area, and flux measurements between its terrestrial substrates and the atmosphere would improve our understanding of polar terrestrial ecosystems as sources or sinks of Hg in the regional and global cycle (section 3.6.6).

4.5.2 Methylmercury production

Pathways for Hg methylation in Arctic terrestrial ecosystems remain poorly characterized, and several novel processes have been suggested to explain elevated concentrations of MeHg observed in Arctic snow (Barkay and Poulain 2007). The presence of microbial cells, dissolved organic substrates, and bioavailable inorganic Hg suggest it is possible that microbes methylate Hg in snow (Barkay and Poulain 2007, Constant et al. 2007, Larose et al. 2010). Another mechanism, proposed by St. Louis et al. (2005, 2007) is the photodecomposition of volatile Me_2Hg in the atmosphere (Fig. 4.1). Concentrations of Me_2Hg measured in Arctic sea water and estimates of evasion fluxes suggest that open water areas in the ocean could be a relevant source to the atmosphere (St. Louis et al. 2007). Some of this Me_2Hg may ultimately be deposited as MeHg on terrestrial snow in coastal areas. Other possible mechanisms for MeHg accumulation that have not been confirmed are sunlight-induced methylation in the presence of labile organic matter in snow, atmospheric methylation by abiotic reactions (Hammerschmidt et al. 2007), or microbial methylation on aerosols in the atmosphere (Barkay and Poulain 2007).

Water-logged terrestrial soils are also potential sites for MeHg production in the Arctic. In temperate environments, wetlands can be major sources of MeHg, which is formed by the activity of sulphate- and iron-reducing bacteria (Gilmour et al. 1992, Fleming et al. 2006). Loseto et al. (2004) showed that once thawed and incubated at typical Arctic summer temperatures (4–8°C), MeHg concentrations increased 100-fold in Arctic wetland soils. While sulphate-reducing bacteria are thought to be the predominant MeHg producers in temperate anoxic environments, the genes responsible for sulphate reduction could not be detected from all the Arctic wetland sites studied by Loseto et al. (2004). This suggested either some issue with extraction and amplification of the genomic DNA or that sulphate reducers are in fact not the most dominant methylators in Arctic wetlands. Hammerschmidt et al. (2006) evaluated the biogeochemical cycling of MeHg in lakes and tundra watersheds of Arctic Alaska (68° N). They concluded that MeHg supply from tundra watersheds was modest relative to sediment MeHg production in lakes. Oiffer and Siciliano (2009) evaluated the potential for wet sedge meadow soils on Truelove Lowland (75° N), which are typical of Arctic landscapes, to act as sources or sinks for MeHg. They reported that Hg was mostly associated with natural organic matter with low sulphide content and observed significant methylation after inorganic Hg was amended to the soil. This finding suggests that there is a potential for soil production of MeHg during spring melt.

The decomposition of MeHg reduces its concentration in snow and soils. While demethylation rates have not been measured in Arctic terrestrial environments, this process may occur via abiotic or microbial pathways (Barkay and Poulain 2007). Lahoutifard et al. (2005) observed a decrease in snowpack MeHg concentration during spring time which was negatively correlated with temperature. Constant et al. (2007) observed rapid demethylation of snow MeHg at night which ruled out photodecomposition at that site. Further study is required to identify production and loss processes of MeHg in Arctic terrestrial ecosystems.

4.6 Food webs

Terrestrial food webs in the Arctic generally consist of three trophic levels: primary producers (plants and lichens), herbivores, and carnivores or scavengers. Information is currently lacking on the transfer of Hg through terrestrial food webs in the Canadian Arctic, although it is likely that biomagnification occurs between consumers and their diet. Rimmer et al. (2010) showed a pattern of Hg biomagnification at successive trophic levels in a southern montane forest food web, with the highest concentrations observed in raptors. Evers et al. (2005) found a similar pattern in avian freshwater communities in northeastern North America and also noted high levels of Hg in insectivorous birds.

Given these patterns of biomagnification, it would be expected that the highest levels of Hg occur in top trophic-level consumers in the Arctic as well. A typical Arctic terrestrial food web consists of lichens, barren-ground caribou, and wolves. While caribou have higher THg concentrations than lichens (Gamberg 2009, 2010b), wolves do not necessarily have correspondingly higher concentrations (Gamberg and Braune 1999). For example, dry weight (dw) concentrations of THg in caribou kidney averaged 2.8 ± 1.7 µg g^{-1} (Annex Table A2.1) while those reported for wolf kidney from several Canadian Arctic regions averaged 1.0 ± 0.7 µg g^{-1} (converted from wet weight concentrations assuming 77% moisture) (Gamberg and Braune 1999). Arctic fox in Alaska also showed similar levels of kidney THg (Dehn et al. 2006). This lack of apparent biomagnification may occur because wolves also feed on moose and small mammals, which tend to have considerably lower Hg body burdens than caribou (Gamberg et al. 2005b). A more detailed investigation into the transfer of Hg within this Arctic food web would be useful.

Recent studies have shown that terrestrial food webs in Arctic coastal areas can be subsidized by marine resources. Arctic foxes in northern Manitoba will feed in the marine environment during years with low lemming abundance (Roth 2003). In Alaska, satellite-tagged foxes have been observed to travel long distances over sea ice, foraging for several months at a time (Pamperin et al. 2008). Wolves will also feed on marine mammal carcasses along the Alaskan coastline (Watts et al. 2010). While no investigations have been conducted on the effects of marine feeding on Hg transfer to terrestrial carnivores and scavengers, a study in Svalbard showed that Arctic foxes linked to a marine-based diet fed at a higher trophic position and had higher tissue contaminant levels than those with more terrestrial-based feeding (Fuglei et al. 2007).

The role of insects and insectivores in the trophic transfer of Hg also remains poorly resolved in Arctic terrestrial food webs. Among Arctic mammals, only shrews and bats eat appreciable amounts of insects, but the latter is an obligate insectivore in this environment. Arctic birds, however, have a much greater diversity in diet. In the boreal Arctic, some passerines (warblers, thrushes, vireos and others), hummingbirds, woodpeckers, and raptors (kestrels and shrikes) eat insects to varying degrees while nightjars, flycatchers, and swallows are aerial insectivores that eat almost exclusively flying insects. Rails, dippers, and blackbirds eat mainly aquatic larvae of insects. Insects could potentially represent an important route of Hg exposure to some Arctic wildlife.

4.7 Vegetation

Mercury accumulation in plants has received relatively little study in the Canadian Arctic. Two recent investigations, one in Yukon (Gamberg 2009) and one in Nunavut (Choy et al. 2010), both found that lichens had the highest concentrations of THg among all vegetation sampled and ranged from 49 ng g^{-1} in Yukon to 231 ng g^{-1} in Nunavut. Although concentrations were generally higher in lichens from the Nunavut site, this variation may be due to differences in species collected from each location. An earlier study in Nunavut (Chiarenzelli et al. 2001) measured the same concentration of THg (72 ng g^{-1}) in the curled snow lichen as was found in Yukon for the same species (Gamberg 2009). Yet, the crinkled snow lichen had twice as much THg as the same species in Yukon.

Lichens are an important primary producer in the Arctic and a significant pathway for Hg transfer to barren-ground caribou because it is their main winter food source (Gamberg 2009). Crête et al. (1992) found that concentrations of THg in lichens increased eastward in Quebec and were higher in tundra biomes than forest. Based on their findings, Crête et al. (1992) predicted that the Leaf River caribou herd (tundra biome) would be more exposed to THg than the nearby George River herd (boreal forest biome). Some years later, Robillard et al. (2002) confirmed this prediction showing that adult caribou from the Leaf River herd had two to three times higher renal and hepatic Hg concentrations than adult George River caribou. Lichens have no root

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system and obtain their nutrients from the atmosphere; in doing so, they also absorb airborne Hg (Poissant et al. 2008). Crête et al. (1992) hypothesized that lichens on the tundra would be exposed to more Hg deposition due to the absence of trees and the rarity of vascular plants that could intercept particulate deposition. Flowering plants in the Arctic tend to have lower THg than lichens while sedges and willows show the lowest concentrations (Gamberg 2009, Choy et al. 2010). The latter vegetation types do not provide a major source of Hg to Arctic herbivores because of their low Hg concentrations.

Carignan and Sonke (2010) reported that THg concentrations in tree lichens in Quebec were highest near Hudson Bay (up to 2.1 $\mu g\ g^{-1}$ dw) and decreased along a gradient inland in association with lower tissue concentrations of the halogens bromine, chlorine, and iodine. They concluded that the high THg measured in tree lichens close to Hudson Bay likely resulted from AMDEs. Tree lichens appear to incorporate particulate and total gaseous Hg throughout the year, without being much affected by Hg re-emission processes. The authors hypothesized that the uptake of Hg in lichen tissue and its complexation with lichenic acids prevent it from further photoreduction and re-emission to the atmosphere. Lower concentrations of THg reported by Choy et al. (2010) for terrestrial lichens from another Arctic coastal area—presumably also affected by AMDEs—could be explained by the insulating effect of snow cover for much of the year, as compared with tree lichens which are directly exposed to atmospheric Hg all year long.

The proportion of THg as MeHg in caribou forage plants sampled in Yukon ranged from 0.5–31%, and higher proportions were observed in most plant groups collected from a coastal location on the North Slope as compared to an inland location (Gamberg 2009). This difference may be related to AMDEs, which occur in coastal areas (Steffen et al. 2008). However, the mechanism is not yet clearly understood.

4.8 Terrestrial animals

4.8.1 Distribution of Hg in tissues and organs

Terrestrial animals (including birds) in the Arctic typically have very low levels of Hg, particularly when compared to marine animals. Caribou are an exception because they often consume large amounts of lichen. In general, kidney and liver contain the highest levels of Hg among body organs and tissues. These organs are the body's natural filters for toxins and waste. Interestingly, renal Hg concentrations in terrestrial animals are consistently higher than hepatic concentrations (Annex Table A2.1) while the reverse is true for animals in both marine and freshwater environments (Kim et al. 1996, Fisk et al. 2003, Gamberg et al. 2005b). This pattern is likely related to the dominant form of Hg in the diet—inorganic Hg versus MeHg—and differences in demethylation mechanisms among species.

The kidney is often used for monitoring Hg in terrestrial species because of high concentrations in this organ. Muscle is usually of greatest concern when considering wildlife commonly hunted for food but this tissue is less suitable for monitoring purposes because muscle Hg levels are about three orders of magnitude lower. Muscle tissue of caribou and moose mainly contain MeHg, which accounts for about 75% and 90% of THg, respectively (Gamberg 2010a). These proportions are somewhat lower than the average of 100% reported for muscle tissue in marine mammals (Wagemann et al. 1997) and are likely related to the form of Hg in the diet.

4.8.2 Caribou

In general, barren-ground and woodland caribou have a winter diet of primarily lichen (Kelsall 1968, Bloomfield 1980). This results in a greater range of Hg concentrations among them than is found in other terrestrial herbivores across the Canadian Arctic. Mean renal concentrations ranged from 0.30 μg g^{-1} THg dw in the woodland Klaza caribou herd to 6.2 μg g^{-1} in the barren-ground

BOX 4.1 Yukon hunters donate organs to science

Between 1994 and 2010, Yukon hunters donated organ and tissue samples from over 2,000 moose and caribou to the NCP. Each year, a request was made to the public to donate kidney, liver, muscle, and tooth samples from their successful moose and caribou hunts and the response was overwhelmingly positive. Yukon Environment was an essential partner in this project, accepting samples at their main offices as well as offices in the smaller communities.

All participating hunters received a letter summarizing the research and the age of their particular moose or caribou. Each hunter's name was also put into an annual draw for a charter flight with a local aircraft company. Hunters reported that they appreciated the opportunity to help scientists with their research while local Conservation Officers reported that this project was highly regarded and provided valuable opportunities for positive contact and information exchange with hunters.

For the NCP, the crucial support of hunters made it possible to assess moose and caribou from a broad geographic range and in large enough numbers to make confident conclusions from the research and monitoring. It would not have been possible for one scientist, or even a team of scientists, to collect samples on their own from so many moose and caribou.

Yukon Government

As a result of this program, scientists have concluded that moose and caribou in Yukon are not of concern in terms of Hg and, for the most part, do not need to be monitored any further. However, the Porcupine caribou, along with the Qamanirjuaq caribou herd in the eastern Arctic, will continue to be regularly monitored to track any long-term changes that may occur.

Scientists with the NCP sincerely appreciate the support of local Yukon hunters!

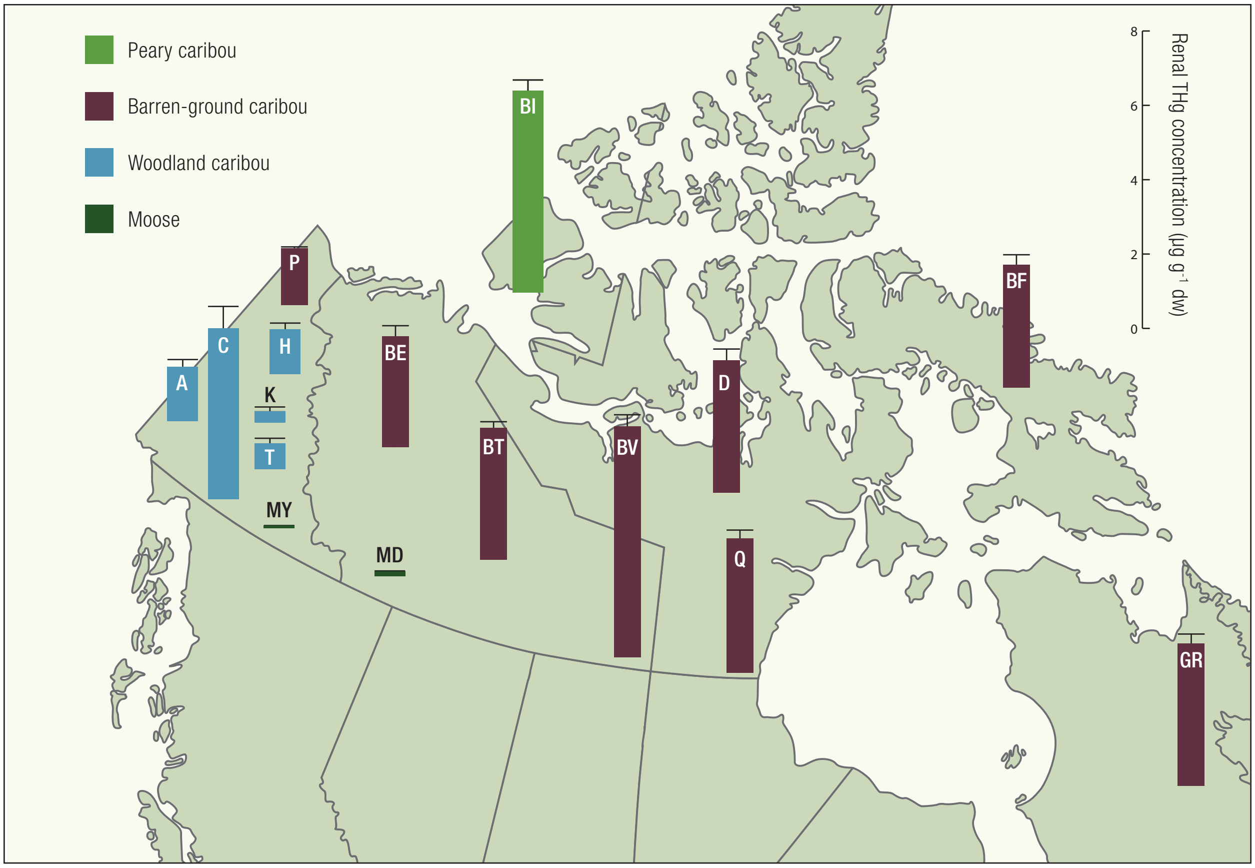

FIGURE 4.8

Renal THg concentration (µg g^{-1} dw) in Arctic caribou and moose collected from 1999 to 2009 with a minimum sample size of five. Abbreviations for the caribou herds are: A = Aishihik, BE = Bluenose East, BF = Baffin Island, BI = Banks Island, BT = Bathurst, BV = Beverly, C = Carcross, D = Dolphin and Union, GR = George River, H = Hart River, K = Klaza, P = Porcupine, Q = Qamanirjuaq, and T = Tay. Abbreviations for the moose herds are: MD = Deh Cho and MY = Yukon. Data are from Gamberg et al. (2005b), Gamberg (2008, 2009a, 2010a, 2010b), Larter and Nagy (2000), and Pollock et al. (2009).

Beverly herd (Fig. 4.8). Differences in THg concentrations among caribou herds may be due to atmospheric Hg deposition patterns across the Arctic (Dastoor and Larocque 2004) and/or differing proportions of diet consisting of lichens (Larter and Nagy 2000). The winter diet of Peary caribou contains very little lichen and is largely made up of grasses, sedges, and forbs (Parker 1978, Shank et al. 1978, Larter and Nagy 1997). Therefore, lower concentrations of Hg were expected in these caribou, and it is not clear why their THg levels were relatively high (Fig. 4.8).

Seasonal monitoring of the Porcupine caribou herd in Yukon revealed that spring-collected females had higher renal concentrations of THg than fall-collected females (Gamberg 2006). Seasonal variation in kidney size has been used to explain differences in cadmium concentrations in caribou. Kidneys are at their largest in the fall (Crête et al. 1989, Gamberg and Scheuhammer 1994), and this may provide at least a partial explanation for differences in THg concentrations. If dietary intake of Hg does not change appreciably over one season, a heavier kidney in the fall would have a lower concentration of Hg. Seasonal variation in diet may also play a role. A model estimating Hg intake by Porcupine caribou (Gamberg 2009) indicated that Hg intake is highest in the late fall and winter when lichens make up most of the diet and is lowest in the summer months when the caribou are feeding more on grasses, sedges, and forbs (Fig. 4.9). There is a potential spike of Hg intake in the fall when mushrooms are available, but this modelling result needs to be confirmed.

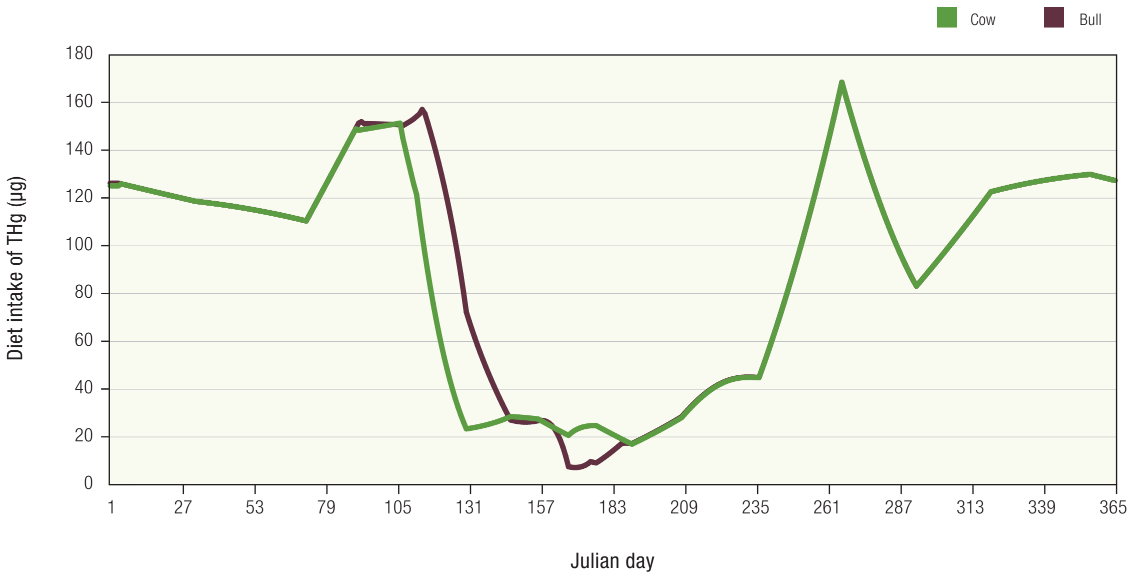

FIGURE 4.9

Average THg intake (µg) over a calendar year (presented as Julian days) for a typical Porcupine caribou bull and cow (Gamberg 2009).

Pollock et al. (2009) found a different seasonal trend for George River caribou in Labrador, which had lower renal THg concentrations in caribou taken in late winter (February) than those taken in the fall. Although no specifics on their diet were given, the authors attributed this pattern to seasonal differences in diet. Unlike other northern ungulates, the summer habitat of George River caribou rather than winter ones appear to be particularly constraining for foraging (Messier et al. 1988). Low fat reserves of females during the first month of lactation and in the fall, combined with low calf growth, suggest that caribou are nutritionally stressed during summer (Huot 1989, Crête and Huot 1993, Manseau 1996). Therefore, it would be expected that these animals have lower kidney weights in the fall than in the spring along with correspondingly higher THg concentrations in the fall.

Fall-collected female Porcupine caribou had higher concentrations of renal THg than males. Females are physically smaller than males but have higher energetic demands. They eat more food and hence, proportional to their body weight, females accumulate more Hg than males, which results in higher renal concentrations (Fig. 4.10; Gamberg 2009). This sex difference has also been demonstrated for Hg in mink where females have a smaller body size than males (Gamberg et al. 2005a).

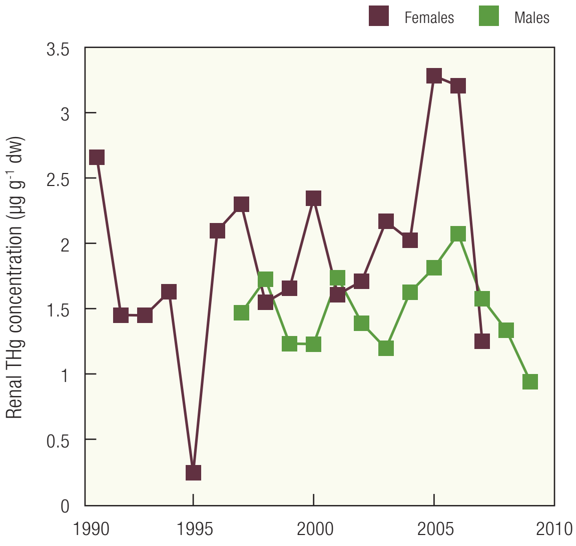

FIGURE 4.10

Renal THg concentration (µg g^{-1} dw) in male and female Porcupine caribou collected during fall between 1991 to 2009 (Gamberg 2010a).

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There was no statistically significant linear trend in renal THg in the Porcupine caribou herd over the last two decades, although there was discernable annual variation (Fig. 4.10). This dataset demonstrates the necessity for annual sampling in wildlife populations because sporadic monitoring could result in erroneous conclusions depending on which years were sampled (Bignert et al. 1993). The annual variation in THg levels in the Porcupine caribou herd appears somewhat cyclic and is likely driven by environmental factors. Concentrations of THg in male and female caribou varied in a similar fashion. However, males lagged behind females by about one year (Fig. 4.10). Female caribou may respond more quickly to the environmental factors that control Hg bioaccumulation or may be more sensitive to those particular drivers. One hypothesis to explain the cyclic inter-annual variation in THg levels of both male and female caribou is the influence of the Pacific Decadal Oscillation—a pattern of climate variability over the Pacific Ocean—on atmospheric Hg deposition and its subsequent uptake in lichen (Gamberg 2010b). An association with the Pacific Decadal Oscillation is suggested by a decrease in caribou renal concentrations when the index is in a negative mode and an increase when the index is in a positive mode. Further analysis is required to investigate this potential relationship and the underlying processes.

4.8.3 Other terrestrial animals

Arctic hare and a variety of rodents sampled from the Canadian Arctic had notably low concentrations of THg (Annex Table A2.1). One exception was a renal THg concentration of 2.94 μg g^{-1} dw in a composite sample of four red-backed voles from Yukon (Gartner Lee 2005). This concentration is more similar to those in caribou than to those in other rodents. However, it is difficult to determine the cause for the relatively high value. Moose also had very low levels of THg, both in Yukon and the Northwest Territories (Fig. 4.8; Gamberg 2010b). Their diet consists primarily of willow, which is Hg-poor. Arctic fox had somewhat higher concentrations of hepatic THg (mean: 1.11 μg g^{-1} dw) than terrestrial herbivores (Hoekstra et al. 2003), perhaps due to their opportunistic scavenging of fish and/or marine mammals (Fuglei et al. 2007). In contrast, Choy et al. (2010) measured a low whole-body concentration (0.23 μg THg g^{-1} dw) in ermine, again likely related to their diet which consists almost exclusively of small rodents.

Choy et al. (2010) found that snow buntings had low whole-body concentrations of THg (mean: 0.18 μg g^{-1} dw). Their accumulation of Hg was not influenced by proximity to a seabird colony that has contaminated the local coastal area with Hg through the high-density release of guano. Recently published data on willow ptarmigan collected in northern Canada from 1985 to 1994 revealed similarly low hepatic and renal THg concentrations (mean: 0.07 and 0.23 μg g^{-1} dw, respectively) (Pedersen et al. 2006).

4.9 Summary

In recent years, much new information has been collected on the movement and fate of Hg in the terrestrial cryosphere. Terrestrial snow at Arctic sites had THg concentrations that ranged three orders of magnitude from < 1 to > 100 ng L^{-1}. The high variability in THg reflects atmospheric Hg deposition—mainly during AMDEs—and post-depositional processes, including photoreduction and re-emission to the atmosphere, and burial and redistribution in the snowpack. Concentrations in snow, firn, and ice (< 1 ng THg L^{-1}) at several Arctic glaciers were much lower than those reported in snow surveys of lowland terrestrial environments. The atmospheric flux of THg to glaciers increased from north-to-south, likely due to latitudinal change in wet deposition and/or particulate scavenging. At Penny Ice Cap, the southernmost glacier studied, THg accumulated in snow primarily during the summer or autumn when AMDEs do not occur. These findings suggest that wet deposition of atmospheric Hg may be more important at lower Arctic latitudes. Atmospheric THg deposition rates inferred from glacier snow and ice are lower by one or two orders of magnitude than net accumulation rates in sediments and peat, as well as flux estimates from atmospheric models. A peak flux of THg was observed between the mid 1990s and 2005 in snow and ice archives from Agassiz and Mount Oxford, but this temporal trend was not observed through direct monitoring of atmospheric GEM at Alert. These discrepancies suggest that in order to better quantify atmospheric Hg contributions to the Arctic, there is a need for additional measurements of wet and dry deposition and more refined modelling of Hg deposition processes in different environments.

Less information is available for MeHg in snow, although concentrations were typically < 0.2 ng L^{-1} at glacier and lowland sites. Several mechanisms have been proposed to explain the occurrence of MeHg in snow including *in situ* microbial methylation as well as atmospheric MeHg deposition, and further work is needed to confirm these hypotheses.

Few published studies currently exist on Hg in Canadian Arctic soils. Data from two High Arctic islands and the Beaufort Sea coast suggest there can be considerable regional variation in soil THg and MeHg concentrations. Limited investigations indicate that Arctic soils have the capacity to methylate Hg and to emit Hg(0) to the atmosphere. However, the extent of this processing and implications for the local and regional Hg cycles remain unknown.

In general, terrestrial animals in the Canadian Arctic have low levels of THg, and caribou have the highest concentrations among the species studied. Recent research on Hg accumulation in Arctic vegetation indicates that lichens contain more THg than other plant types such as shrubs, flowers, and willows. The importance of lichen in the diet of caribou may explain their higher THg burdens. Monitoring of THg levels in the Porcupine caribou herd indicates no significant change has occurred over the last two decades, although there is discernable annual variation that is likely driven by yet-unidentified environmental factors. Seasonal variation in caribou THg concentrations as well as differences between males and females point to the importance of bioenergetics and seasonal diet. Further investigation of the pathways and processes of bioaccumulation is required to better understand the geographic and temporal trends of Hg in caribou.

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CHAPTER 5

Freshwater Environment

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5.1 Introduction

The Canadian Arctic contains vast freshwater resources that cover about 140,000 km^2 of land north of 60° latitude (Prowse et al. 2009). Freshwater fish are important in the diet of many Northerners, particularly in the Northwest Territories, Yukon, and northern Quebec. Over the last two decades, extensive monitoring of Hg in freshwater fish has been conducted across the Canadian Arctic, and in some lakes, concentrations of Hg in top predator species exceed Health Canada's consumption guideline for commercial sale of fish. While trophic position, age, and size of fish have been identified as important factors, no geographic trends were discernable for the elevated Hg levels (Fisk et al. 2003). In the previous *Canadian Arctic Contaminants Assessment Report (CACAR II)*, additional study was recommended to better explain the high lake-to-lake variability of Hg in freshwater fish and to investigate potential influences of atmospheric mercury depletion events (AMDEs) and climate change on food web bioaccumulation of Hg (Fisk et al. 2003).

Several important advances have been made in our understanding of the Hg cycle in Arctic freshwater ecosystems during Phase III of the Northern Contaminants Program (NCP), particularly on Hg concentrations in water, Hg fluxes to sediment, methylmercury (MeHg) cycling, bioaccumulation in food webs, temporal changes in fish Hg levels, and impacts of climate change. Water concentrations of Hg are now available for Arctic lakes, ponds, and several large rivers. Mercury profiles in lake sediments across the Canadian Arctic provide new estimates of deposition rates to fresh waters. Production and loss rates of MeHg in ponds were modelled, and snowmelt fluxes of MeHg to lakes were investigated. To determine factors influencing lake-to-lake variability in Hg bioaccumulation, food webs were studied in Arctic lakes where landlocked char or trout are the top predator fish. A novel approach to tracing Hg sources using Hg stable isotopes was also applied to some of these Arctic lake food webs. Recent studies examined potential influences of climate change on Hg bioaccumulation in fish and on Hg sequestration in lake sediments. Impacts of thawing permafrost on Hg transport to lakes are under investigation in the Mackenzie River basin. Annual monitoring of key fish species during Phase III of the NCP has further strengthened the temporal trend datasets for the freshwater environment.

5.2 Overview of the Hg cycle in Arctic freshwater ecosystems

Freshwater ecosystems receive Hg, primarily as divalent inorganic Hg [Hg(II)], by direct atmospheric deposition and delivery in runoff from the watershed (Fig. 5.1). Methylmercury is also deposited from the atmosphere although this flux is probably small relative to inorganic Hg. Aqueous Hg(II) is sequestered, transformed, or removed from the system by: 1) reduction to elemental Hg [Hg(0)] and evasion to the atmosphere; 2) binding to particles in the water column, followed by sedimentation; 3) microbial methylation in sediment or the water column; and, 4) hydrological removal via the outflow of lakes or flushing in streams and rivers. Snowmelt or wetlands may also be significant sources of MeHg. Following entry into the food web by uptake in microbial organisms, MeHg bioaccumulates in individuals and biomagnifies in consumers at higher trophic levels. Aqueous MeHg is broken down to inorganic forms of Hg by photodemethylation in the water column and by microbial demethylation in sediment. The current state of knowledge on these complex processes is presented in more detail throughout this chapter.

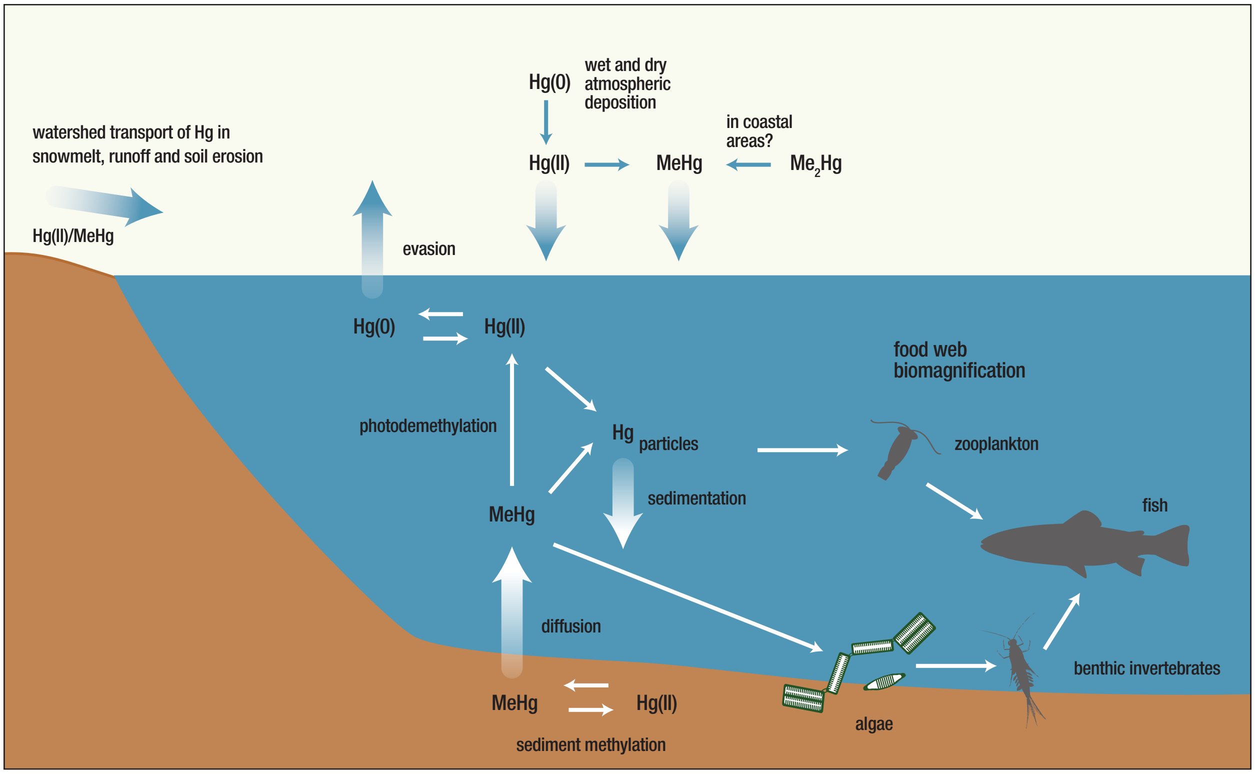

FIGURE 5.1

Schematic of major transformations and pathways in the Hg cycle for Arctic freshwater ecosystems.

5.3 Lake and river water

5.3.1 Ecosystem and geographic variation in water Hg concentrations

Water concentrations of total Hg (THg) and MeHg were compiled to examine geographic and ecosystem variation in fresh waters of the Canadian Arctic (Table 5.1; Fig. 5.2). Most available data are for lakes and large rivers although some ponds, streams, and wetlands have also been sampled in recent years. The most extensively sampled areas are two High Arctic islands (Cornwallis, Ellesmere) and the Mackenzie River basin in the Northwest Territories.

Water concentrations of THg ranged widely from 0.1–19.8 ng L^{-1} in different water bodies of the Canadian Arctic (Fig. 5.2; Table 5.1). Lakes, ponds, and wetlands from several Arctic regions had relatively low concentrations—generally less than 3 ng L^{-1}—whereas rivers often had considerably higher levels. Elevated concentrations were mostly measured in the Mackenzie River or its tributaries and were largely associated with particulate matter. The Mackenzie had more particulate Hg during periods of high flow due to increased erosion in the basin (Leitch et al. 2007). Concentrations of THg were generally lower in other rivers and streams including sites on Cornwallis Island and the Nelson and Churchill rivers (Table 5.1). Geographic variation in hydrology and drainage basin characteristics may influence concentrations of THg in flowing waters.

Water concentrations of MeHg in Arctic water bodies ranged two orders of magnitude from < 0.02 ng L^{-1} (below analytical detection) to 3.1 ng L^{-1} (Fig. 5.2; Table 5.1). The majority of lakes, rivers, streams, and wetlands (for which data are available) had low MeHg concentrations (≤ 0.1 ng L^{-1}). At sites on Ellesmere Island where high concentrations were observed, ponds showed great potential for MeHg production. These shallow water bodies had characteristics known to enhance microbial Hg methylation, namely warm water temperatures and high concentrations of labile organic matter (St. Louis et al. 2005). Elevated MeHg concentrations were also observed in Mackenzie Delta lakes (Table 5.1) and near the outflow of the Churchill River where vast wetlands are the suspected source (Kirk and St. Louis 2009). In lakes along the Mackenzie River, water concentrations of THg and MeHg, as well as the proportion of THg as MeHg, were higher in

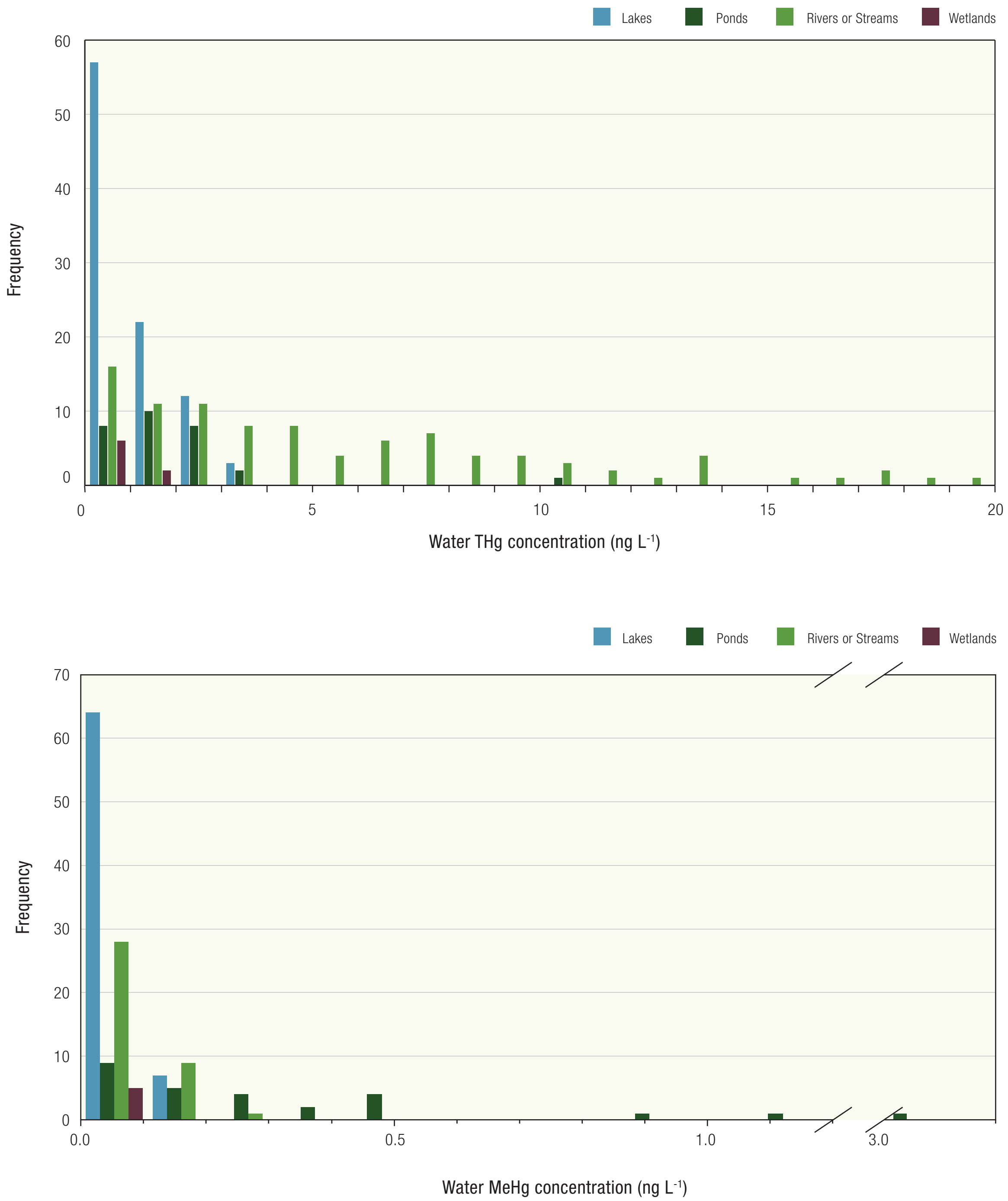

FIGURE 5.2

Frequency distributions of unfiltered THg and MeHg concentrations in water from different freshwater ecosystems in the Canadian Arctic (data sources in Table 5.1).

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smaller lakes where dissolved organic carbon (DOC) concentrations were also higher (Evans et al. 2005). Wetlands on Cornwallis and Ellesmere islands were typically water-logged soils overlaid with grass, sedge, or moss and had low aqueous MeHg (Loseto et al. 2004b; see section 4.5.2). Biogeochemical processing of MeHg in Arctic fresh waters is discussed in section 5.5.

5.3.2 Mercury dynamics and fluxes in the Mackenzie River, and influence of the delta

The Mackenzie is the second largest river in Canada, with an immense water discharge averaging more than 10,000 m^3 sec^{-1}. It is also a turbid river that transports a heavy sediment load north to the Arctic Ocean. The dynamics of Hg in rivers are related to the intensity of water discharge and therefore the seasonality of the river system (Benoit et al. 1998, Domagalski 2001, Lawson et al. 2001). In the Mackenzie River, the strong seasonality of its river flow, its large and diverse watershed (1.8×10^6 km^2), and the effects of ice and large floodplain areas combine to determine Hg concentrations and export to the Beaufort Sea.

Seasonality and hydrology of the Mackenzie River

The Mackenzie River has a seasonal flow pattern that is unique to large north-flowing rivers. During the winter period (October to May), the river is at low flow and receives little surface runoff because its basin is covered with snow and ice. From May to June, thaw progresses south to north in the Mackenzie River basin and large quantities of snowmelt water are delivered to still frozen northern areas of the river and its large delta floodplain. This meltwater *wave* breaks up the ice cover in downstream areas and creates ice jams that enhance local river levels. It also causes back-flooding of the low-lying floodplain in the Mackenzie River delta (Marsh and Hey 1989). By June, after peak flooding and the breakup of ice jams, river water levels decline and excess water in the delta floodplain drains back through channels to the ocean. From July to October, water levels and flow decline as the river receives less water from its watershed. However, flows can increase periodically throughout the summer due to rain storms in the Mackenzie River basin. Strong windstorms from the coast can also cause flooding in the delta floodplain by *pushing* river water upstream and increasing river water levels.

TABLE 5.1 Concentrations of Hg in waters of rivers, streams, lakes, wetlands, and ponds in the Canadian Arctic. Ancillary measurements of water pH and DOC concentrations were included if available.

Location	Date	*n*	pH	DOC (mg L^{-1})	MeHg (ng L^{-1})		THg (ng L^{-1})		Source
					Unfiltered	Filtered	Unfiltered	Filtered	
Rivers and Streams									
Yukon River, YT	2004	3	7.9 ± 0.1	2.8 ± 1.8		< 0.04		2.2 ± 2.4	1
Yukon River tributaries, YT	2004	14	7.8 ± 0.2	5.1 ± 5.1		< 0.04–0.11		1.5 ± 1.2	1
Mackenzie River, NT	2003–2005	37			0.09 ± 0.03	0.08 ± 0.04	7.0 ± 4.3	2.8 ± 2.1	2, 3
Mackenzie River, NT	2007–2010	6			0.08 ± 0.05	0.03 ± 0.01	13.8 ± 8.0	1.4 ± 0.7	4
Mackenzie River tributaries, NT	2003–2005	20				0.07 ± 0.04	6.3 ± 2.9	2.6 ± 1.9	2
Peel River, NT	2007–2010	1			0.13 ± 0.12	0.03 ± 0.02	18.8 ± 12.2	1.6 ± 0.9	4
Cornwallis Island streams, NU	1994–2006	7	8.2 ± 0.1	1.5 ± 1.1	0.07 ± 0.06		1.2 ± 1.1		5, 6, 7
Ellesmere Island streams, NU	2005	4			0.04 ± 0.03		1.1 ± 0.7		8
Devon Island river, NU	2006	1	8.0	0.9	0.05		0.2		5
Churchill River, MB	2003–2007	1		20.5	0.18 ± 0.09	0.14 ± 0.07	2.0 ± 0.8	1.7 ± 0.7	9
Nelson River, MB	2003–2007	1		15.1	0.05 ± 0.03	0.04 ± 0.02	0.9 ± 0.3	0.5 ± 0.2	9
Large rivers in Nunavik, QC	2005–2007	3					1.1 ± 0.1	1.4 ± 0.2	10
Baker Lake outflow, NU	2005–2007	1					0.72		10
Lakes									
Cornwallis Island, NU	2002–2007	18	8.1 ± 0.1	1.1 ± 0.6	0.04 ± 0.01		0.6 ± 0.3		6, 7, 11, 12
Devon Island, NU	2006	6	8.3 ± 0.3	2.0 ± 1.6	0.04 ± 0.02		0.5 ± 0.2		11
Somerset Island, NU	2005–2007	1	7.8	0.5	0.02		0.7		11, 12
Ellesmere Island, NU	2003, 2005–2007	25	8.2 ± 0.4	2.8 ± 2.7	0.05 ± 0.03		0.9 ± 0.5		13, 8, 12
Mackenzie Valley, NT	1998–2002	18	8.1 ± 0.4	11.5 ± 5.4	0.07 ± 0.04		1.8 ± 0.7		14
Mackenzie Delta, NT	2004, 2010	6			0.10 ± 0.05		2.3 ± 0.6	0.7 ± 0.2	3, 4
Kent Peninsula, NU	2005–2007	3		4.6 ± 2.1			0.5 ± 0.1		12
Victoria Island, NU	2005–2007	1		5.6			0.6		12
Nunavik, QC	2005–2007	2		1.2 ± 0.7			3.2 ± 0.03		12
Wetlands									
Ellesmere Island, NU	2002	2	7.2–7.5	2.2 ± 2.4	< 0.02–0.08		0.7 ± 0.1		6
Cornwallis Island, NU	2002	4		1–4.5	0.06 ± 0.03		1.0 ± 0.2		6
Devon Island, NU	2006	4	7.0 ± 0.2	22.4 ± 8.2				4.4 ± 1.3	15
Ponds									
Cornwallis Island, NU	2006	1	8.5	0.9	< 0.02		0.4		11
Devon Island, NU	2006	4	8.3 ± 0.2	4.6 ± 2.0	0.08 ± 0.05		1.0 ± 0.4		11
Ellesmere Island, NU	2003, 2005	21	8.4 ± 0.4	18.7 ± 12.1	0.53 ± 0.68		2.2 ± 2.0		8, 13

Notes: mean concentrations are presented (± 1 standard deviation), *n* = number of sites sampled, DOC = dissolved organic carbon.

Sources: 1 = Halm and Dornblaser (2007), 2 = Leitch et al. (2007), 3 = Graydon et al. (2009), 4 = Graydon and Emmerton (2010), 5 = Chételat (2010), 6 = Loseto et al. (2004a), 7 = Semkin et al. (2005), 8 = Lehnherr (2010b), 9 = Kirk and St. Louis (2009), 10 = Hare et al. (2008); 11 = Chételat et al. (2008), 12 = Gantner et al. (2010a, 2010b); 13 = St. Louis et al. (2005), 14 = Evans et al. (2005), 15 = Oiffer and Siciliano (2009).

Winter low flow: Hg immobilization

During winter, little watershed material is transported into the Mackenzie River and its tributaries because of minimal surface water movement. The Mackenzie River itself is ice-covered during this time and receives Hg-poor water from oligotrophic lakes and deeper groundwater. During under-ice, low flow conditions in early May, both THg (filtered: 1.0 ± 0.9 ng L^{-1}, unfiltered: 7.8 ± 6.3 ng L^{-1}) and MeHg (filtered: 0.02 ± 0.01 ng L^{-1}, unfiltered: 0.04 ± 0.01 ng L^{-1}) concentrations are generally low in the downstream reaches of the river (Graydon and Emmerton 2010). River Hg concentrations in early May are approximately two to eight times lower than concentrations during higher flow periods later in spring.

Spring freshet high flows: watershed Hg mobilization and the role of particles and organic matter

Water concentrations of Hg in the Mackenzie River are overwhelmingly dominated by particle-bound Hg that originates mostly from mountain-fed rivers to the west of the Mackenzie River proper including the Liard, Arctic Red, and Peel (Leitch et al. 2007). Rivers draining in mountain areas strongly erode catchment and bank material and mobilize elements that associate closely with particles, including Hg (Benoit et al. 1998). Tributaries draining in the mountainous western basin of the Mackenzie River flow intensely during the short snowmelt season and deliver large quantities of suspended rock and soil material. These mountain tributaries (e.g., the Peel and Liard rivers) can have up to two times higher particle-bound Hg concentrations than the low-relief, plains-influenced Mackenzie River (Leitch et al. 2007, Graydon and Emmerton 2010). After integrating water from flood-prone mountain tributaries and the effect of in-channel ice scour, Hg in the lower Mackenzie River is predominantly particle-bound (73–87%; Leitch et al. 2007, Graydon and Emmerton 2010). Large increases in dissolved and particulate concentrations of THg and MeHg (two- to three-fold) are observed in the river during the spring thaw period compared to summer low flow when mountain tributaries deliver much less water to the river (Fig. 5.3; Leitch et al. 2007, Graydon et al. 2009). In other systems, there is also evidence that Hg bound to DOC may be exported from tributaries with extensive wetland and organic soil influences within their watershed (e.g., Hare et al. 2008, Kirk and St. Louis 2009). This process may be an important supply of Hg to the Mackenzie River because it has vast areas of wetlands and organic-rich soils in its catchment (Carrie et al. 2009) that contribute water and DOC to the river during the snowmelt period (Carson et al. 1998, Emmerton et al. 2008).

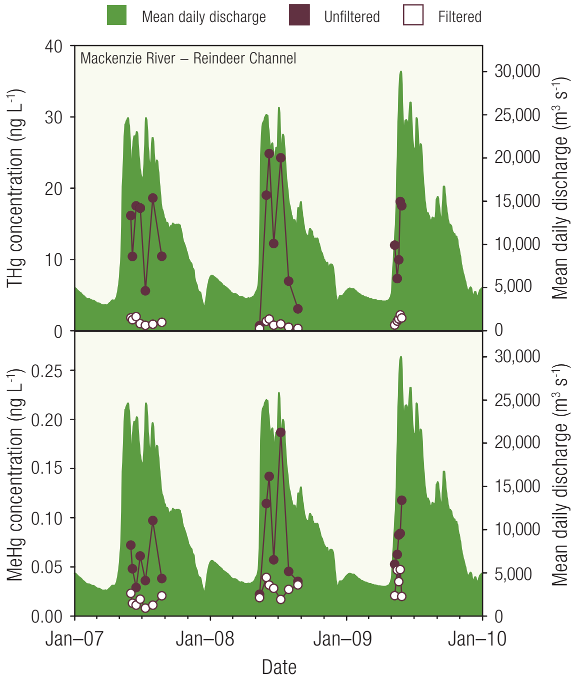

FIGURE 5.3

Filtered and unfiltered concentrations of THg and MeHg in relation to water discharge in the lower Mackenzie River at Reindeer Channel between 2007 and 2009 (Graydon and Emmerton 2010).

Summer and autumn declining flows: event-driven mobility of Hg

After the ice clears and river discharge peaks, runoff within the Mackenzie River basin originates from lower in the soil profile, and erosion decreases as river velocities and water levels decline. These factors combine to deliver less organic matter and sediment to the river water, and concentrations of THg and MeHg also decline rapidly (Leitch et al. 2007, Graydon et al. 2009). Summer precipitation events can mobilize organic matter and sediment through surface runoff and increase Hg concentrations in the Mackenzie River for short periods of time. There is also evidence of mid-summer spikes in Hg concentration in the lower river as high as freshet concentrations, possibly due to coastal storm surges (Graydon and Emmerton 2010).

These storms create dynamic backwater conditions in the lower Mackenzie River and its delta floodplain which can erode and flood low-lying organic soils and increase Hg concentrations locally.

Estimates of Hg export from the Mackenzie River

The geography, hydrology, and climate of the Mackenzie River create an extremely challenging environment for accurate river flow measurement, and by association, estimation of riverine Hg export. Several factors have affected quantification of Hg export from the Mackenzie River including: 1) a lack of water Hg concentration data during winter and rising water, ice-influenced periods; 2) a lack of accurate discharge measurements from the river during dynamic ice-influenced periods; 3) large variability in Hg concentrations and discharge between years (Leitch et al. 2007); and, 4) calculation of Hg export is currently only possible upstream of the Mackenzie delta floodplain, although it is unclear if river Hg concentrations may be affected during passage through the delta. Graydon et al. (2009) found little change in unfiltered MeHg and a 16% decrease in unfiltered THg concentrations downstream of the delta when off-channel storage of river water in delta lakes was considered. There is also preliminary evidence that MeHg concentrations increase in delta lakes in the summer. These sites of Hg methylation are possible sources of MeHg to the Mackenzie River (Graydon et al. 2009).

At present, there are few estimates of THg and MeHg export from the Mackenzie River. Leitch et al. (2007) calculated an annual export of 2,900, 1,200 and 2,400 kg y^{-1} of THg in 2003, 2004, and 2005, respectively, and an annual export of 22 and 7 kg y^{-1} of MeHg in 2003 and 2004, respectively. Graydon et al. (2009) calculated estimates for 2.5 months of export (June to August 2004) that totalled 1,200 kg THg and 8 kg MeHg. Using advanced river modelling and more comprehensive seasonal sampling of the Mackenzie delta, revised estimates of Hg export from the Mackenzie River are expected in the near future (Graydon and Emmerton 2010).

5.3.3 Speciation of Hg in the upper Yukon River basin

Flowing from its upper reaches in British Columbia and Yukon into Alaska (USA), the Yukon River drains the fourth largest watershed in North America (8.6×10^5 km^2) and discharges into the Bering Sea. The United States Geological Survey measured concentrations of THg and MeHg in waters of the upper Yukon Basin in Canada, at three stations on the Yukon River and in 14 of its tributaries (Halm and Dornblaser 2007). This work was part of a larger, comprehensive study of the entire Yukon River basin (Schuster et al. 2011). In the summer of 2004, filtered and particulate fractions of Hg were determined on two occasions at each station in Canadian waters.

On average, THg concentrations were equally partitioned between the dissolved and particulate fractions (49 ± 28%, $n = 32$). However, the observed range in particulate THg (< 0.06–26.3 ng L^{-1}) was much higher than for the dissolved fraction (0.3–6.4 ng L^{-1}), and elevated THg levels in river water were primarily associated with particulates. Dissolved MeHg was below analytical detection at the majority of stations, and the maximum observed concentration was 0.11 ng L^{-1} in Fortymile River. Particulate MeHg concentrations were also generally below analytical detection, and a maximum of 0.07 ng L^{-1} was observed. Dissolved and particulate fractions of Hg were associated with water quality characteristics in rivers of the upper Yukon River basin. River water DOC ranged widely from 0.8–20.6 mg L^{-1} among stations and explained 27% of the variation in dissolved THg (Fig. 5.4). Similarly, dissolved MeHg concentrations were positively correlated with DOC (Spearman correlation coefficient = 0.58, $p < 0.001$) and were only measurable (i.e. above analytical detection) in high-DOC waters (6.9–20.6 mg L^{-1}). In contrast, the concentration of particulate THg was strongly related to the amount of suspended sediment in water (Fig. 5.5). Dissolved THg was not correlated with suspended sediment concentration (Fig. 5.5). Trends for particulate MeHg could not be determined because it was above the detection limit in only a few samples.

Suspended sediment in the rivers was primarily composed of inorganic material, with organic carbon representing < 5% of suspended particle concentrations. However, for a subset of rivers where particulate organic carbon (POC) concentrations in water were available ($n = 16$), POC was strongly correlated with both suspended sediment ($r^2 = 0.92$, $p < 0.001$) and particulate THg ($r^2 = 0.83$, $p < 0.001$). Particulate THg was likely associated primarily with the organic fraction, even though it represented a small portion of the total suspended material. Likewise, dissolved THg and MeHg were associated with DOC, such that both Hg fractions were coupled with watershed organic carbon. In the Yukon River, DOC is primarily terrestrial in origin and a substantial portion is leachate from recent plant production (Guo and Macdonald 2006, Spencer et al. 2008). These observations are consistent with a more detailed analysis for the outlet of the Yukon

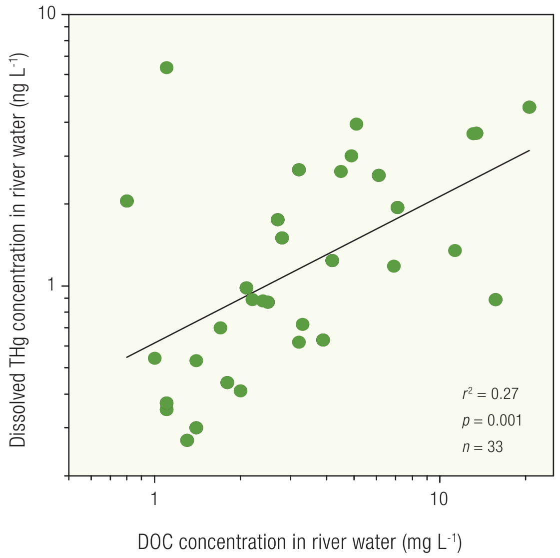

FIGURE 5.4

Relationship between log-transformed concentrations of dissolved THg and DOC in river waters of the upper Yukon River basin (data from Halm and Dornblaser 2007).

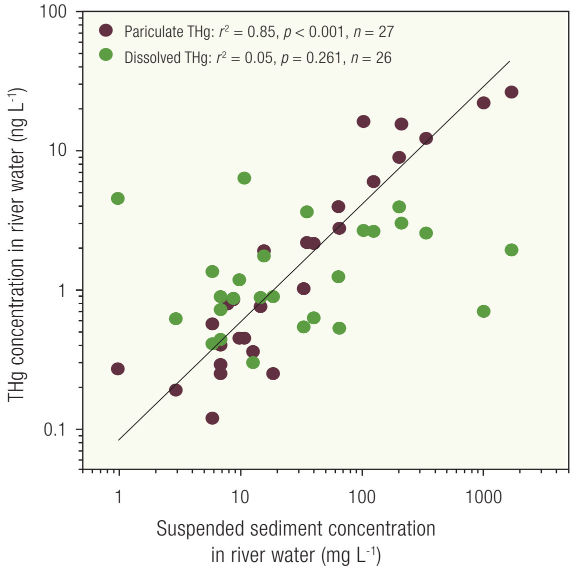

FIGURE 5.5

Influence of suspended sediment on concentrations of particulate and dissolved THg in river waters of the upper Yukon River basin (data from Halm and Dornblaser 2007). All variables were log-transformed.

River basin in Alaska where water concentrations of dissolved and particulate Hg were strongly related to organic carbon (Schuster et al. 2011). Further investigation should characterize the transport processes and coupling of organic carbon to watershed delivery of Hg, particularly given that large reservoirs of organic matter in wetlands and permafrost of northern rivers will likely be impacted by climate change (Schuster et al. 2011).

5.3.4 Exports of Hg from the sub-Arctic Nelson and Churchill rivers

Annual exports of Hg were determined for two Canadian sub-Arctic rivers that flow into the Hudson Bay—the Nelson and the Churchill (Kirk and St. Louis 2009). In the 1970s, roughly 75% of flow from the Churchill River was diverted into the Nelson River for hydroelectric power development. In recent years, discharge from the Nelson River (3,550 m^3 sec^{-1}) has been seven times more than that from the Churchill River (550 m^3 sec^{-1}) (Kirk and St. Louis 2009). Based on continuous measurements from 2003 to 2007, THg and MeHg concentrations were low in the Nelson River (mean ± standard deviation: 0.88 ± 0.33 and 0.05 ± 0.03 ng L^{-1}, respectively) but higher in the Churchill River, particularly for MeHg (1.96 ± 0.8 and 0.18 ± 0.09 ng L^{-1}, respectively). Hence, the Churchill River may be an important source of MeHg to organisms feeding in its estuary. A large portion of THg in the Nelson River was particulate-bound (39 ± 23%), which likely reflects high sediment loads from large regions of cropland in the Nelson River catchment as well as bank erosion due to high flows post-diversion for hydrological power production. In the Churchill River, however, most THg was in the dissolved form (78 ± 15%) and was likely DOC-bound Hg originating in surrounding wetlands. In fact, both the Nelson and Churchill rivers had high DOC concentrations and were therefore large exporters of DOC to Hudson Bay (1480 ± 723 and 392 ± 309 x 10^3 t y^{-1}, respectively). Despite higher Hg concentrations in the Churchill River, average THg and MeHg exports to Hudson Bay from the Churchill River (37 ± 28 and 4 ± 4 kg y^{-1}, respectively) were less than half of exports from the Nelson River (113 ± 52 and 9 ± 4 kg y^{-1}) because of differences in flow. Interestingly, combined Hg exports to Hudson Bay from the Nelson and Churchill rivers are comparable to estimated THg inputs from spring snowmelt on Hudson Bay ice (177 ± 140 kg y^{-1}) but are about 13 times greater than MeHg inputs from snowmelt (1 ± 1 kg y^{-1}) (Kirk and St. Louis 2009). Together, Hg inputs from the rivers and snowmelt were estimated to contribute approximately 16% to the THg pool in Hudson Bay waters but account for a lesser portion (6%) of the MeHg pool.

5.3.5 Snowmelt delivery of Hg to High Arctic lakes

Snow is a large reservoir that accumulates inorganic Hg and MeHg over the long Arctic winter. As spring temperatures warm the snowpack above freezing, Hg is rapidly leached and transported in runoff to fresh waters downstream (Lindberg et al. 2002, Dommergue et al. 2003, Lahoutifard et al. 2005). Intensive sampling of inflow streams to Amituk Lake (Cornwallis Island) showed that stream water concentrations of THg were highest at the onset of spring melt (1.4–4.4 ng L^{-1}) and decreased through spring to a summer low of 0.02–0.4 ng L^{-1} (Loseto et al. 2004a, Semkin et al. 2005). Water concentrations of MeHg in those streams also followed the same decline during the spring melt (Loseto et al. 2004a). The high Hg concentrations in stream water coupled with high discharge resulted in important amounts of THg and MeHg being delivered during spring (Loseto et al. 2004a, Semkin et al. 2005). Snowmelt Hg can undergo several fates during transport: 1) uptake by microorganisms; 2) adsorption onto particles in soils and stream channels; 3) methylation of inorganic Hg; 4) reduction to elemental Hg and emission to the atmosphere; or, 5) flushing into lakes and ponds.

In the Canadian High Arctic, snowmelt is a dominant source of Hg to lakes. Semkin et al. (2005) calculated that surface run off was responsible for almost all the THg input to Amituk Lake, about 80% of which occurred during spring freshet in June and early July. Spring freshet is the critical period of discharge from High Arctic watersheds because most total annual precipitation is deposited in the form of snow during the long polar winter (Woo 1983). Loseto et al. (2004a) observed that water concentrations of THg and MeHg in lakes on Cornwallis Island were highest in spring due to snowmelt inputs. Vertical profiles in the water column of Barren Lake on Cornwallis Island indicated that the surface layer had the highest THg and MeHg concentrations during the spring melt period (Fig. 5.6; Lean and Hui 2011). Reflecting the input of snowmelt, the surface water of the lake also had lower conductivities and colder temperatures than deeper layers. Surface water concentrations of THg in Barren Lake were much lower than those associated with snow during AMDEs—but similar to values in local snow just prior to spring melt—whereas MeHg concentrations were similar to those measured in local snow (Lahoutifard

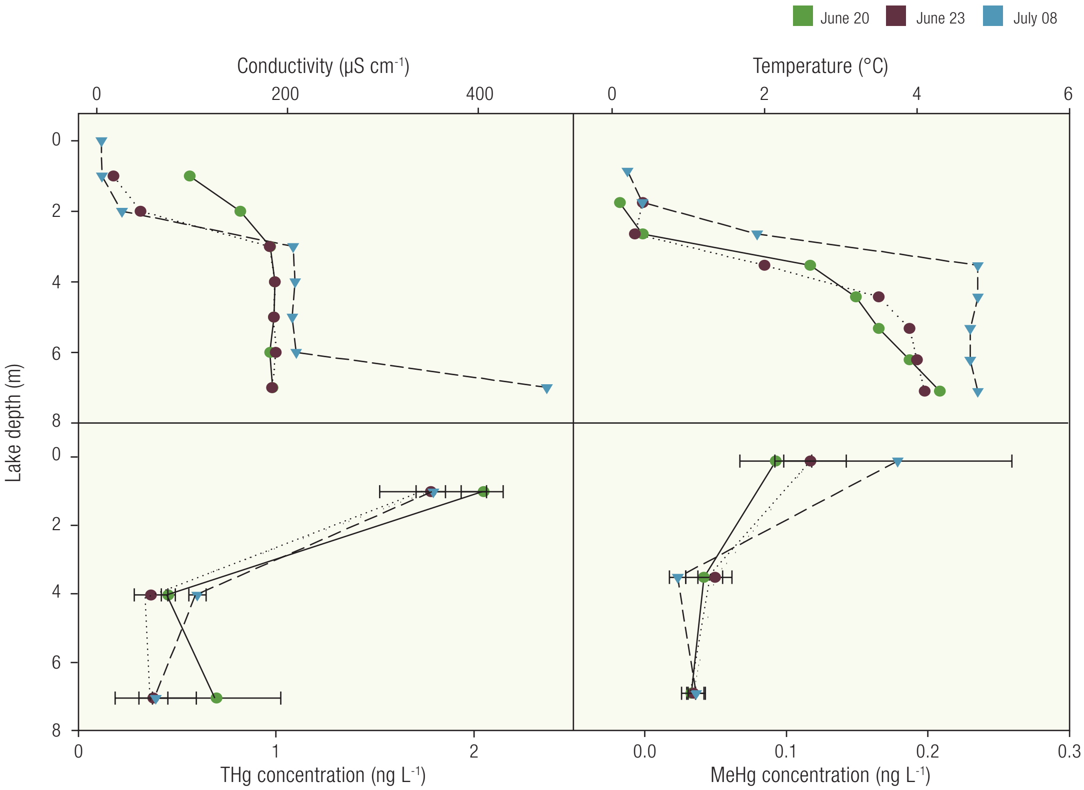

FIGURE 5.6

Vertical profiles of water temperature (°C), specific conductivity (μS cm^{-1}), and concentrations of THg and MeHg (ng L^{-1}) in the water column of Barren Lake, Cornwallis Island, during spring melt (Lean and Hui 2011).

et al. 2005). Less information is available on snowmelt delivery of Hg in other regions of the Canadian Arctic, although this spring-time source may be less important in the overall Hg budget of lower latitude Arctic lakes, such as in Alaska, because of differences in watershed characteristics and hydrology (Fitzgerald et al. 2005, Hammerschmidt et al. 2006).

5.4 Lake and stream sediments

5.4.1 Spatial analysis of Hg levels in bulk sediment from Arctic streams and lakes

Watershed-scale processes of Hg accumulation in the active layer of lake and stream sediments were investigated in the Canadian Arctic. This study was part of a larger Canada-wide analysis of sediment designed to identify how THg concentrations in lake and stream sediments relate to sediment attributes as well as the topography, climate, vegetation, and geology of water bodies and their upslope catchments (Nasr et al. 2011). Within this framework, a total of 36,310 sampling locations of Arctic stream and lake sediments were analysed using the National Geochemical Reconnaissance database (NGR) provided by the Geological Survey of Canada (GSC 2008) (Figs. 5.7 and 5.8). The following information and data sources were used in the analysis:

- THg (ng g^{-1}), loss on ignition (LOI, %), concentrations of cadmium, copper, zinc, manganese and antimony (ng g^{-1}) in bulk sediment (0–30 cm depth) on a dry weight (dw) basis (GSC 2008);
- national and provincial or territorial digital data for geological and lithological specifications (NRCan 2007);
- national and provincial or territorial digital data for land cover type (GeoBase);
- stream and lake morphometry (e.g., depth, stream width) (NGR);
- national and provincial or territorial digital elevation models to evaluate the topographic flow-accumulation at each of the GSC stream and lake sampling locations and to delineate upland or lowland sites (Murphy et al. 2009); and,
- GRAHM (Global/Regional Atmospheric Heavy Metals Model) estimates of atmospheric Hg deposition for 2005 at each sampling location (Dastoor and Moran 2010).

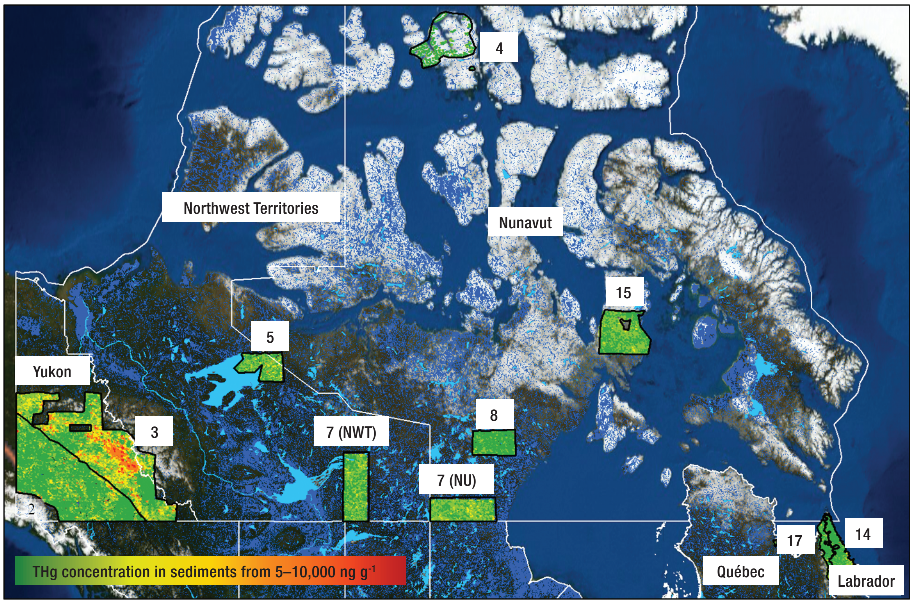

FIGURE 5.7

Map of numbered survey zones examined for THg concentrations in bulk sediment from the NGR database (GSC 2008). Concentrations inside these zones are categorized by colour, from low (green) to high (red). Lowlands (in dark blue) were identified using a national digital elevation model (300 m resolution).

Sediment THg concentrations in streams and lakes ranged from 5–5,950 ng g^{-1} with an overall mean concentration of 65 ng g^{-1}. The vast majority of sediment samples (99.6%) had values between 5–600 ng g^{-1}, 112 had values over 600 ng g^{-1}, and 20 had values over 1,500 ng g^{-1} (Fig. 5.9; Table 5.2). Most notable were the generally high values (mean: 110 ng g^{-1}) within the Selwyn Basin (Yukon), and the generally low values (mean: 25 ng g^{-1}) on Bathurst Island (Nunavut). For survey zones with both stream and lake data, THg was slightly but significantly lower in lake than in stream sediments in the northeastern Arctic but the opposite was found in the Yukon (Table 5.2) and southern locations in Canada (Nasr et al. 2011).

The differentiation of upland versus lowland sites by digital elevation modelling revealed that, overall, there was significantly higher sediment THg in upland locations (mean: 49 ng g^{-1} for lakes, 78 ng g^{-1} for streams) than in lowland locations (mean: 39 ng g^{-1} for lakes, 65 ng g^{-1} for streams). These upland versus lowland trends were also found for lakes within specific survey zones, namely Great Bear Lake (60 vs. 50 ng g^{-1}), Baker Lake (33 vs. 26 ng g^{-1}), the Northwest Territories (52 vs. 42 ng g^{-1}) and southern Nunavut (62 vs. 51 ng g^{-1}) (Table 5.2). Upland stream sites had significantly higher THg compared to lowland sites in the east Yukon (Selwyn Basin; 113 vs. 96 ng g^{-1}) and west Yukon (43 vs. 37 ng g^{-1}).

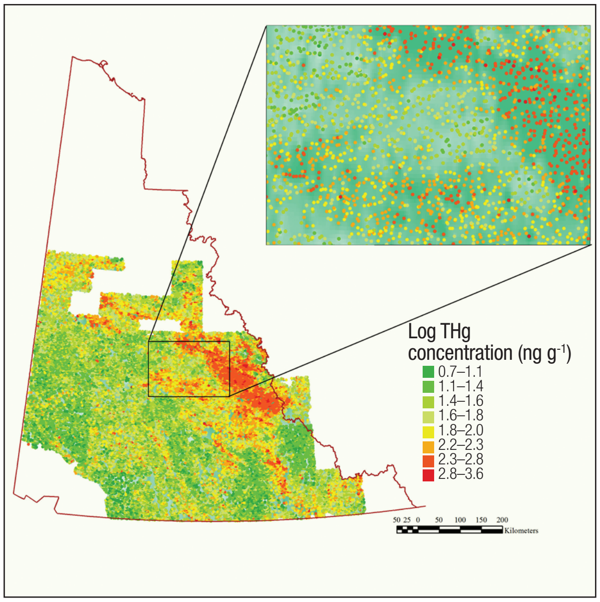

FIGURE 5.8

The Yukon survey zone, showing variation in sediment THg concentrations, with an enlarged view of 1,600 stream sites used to investigate basin-specific geologic and topographic controls on sediment Hg (GSC 2008). Concentrations are presented on a logarithmic scale.

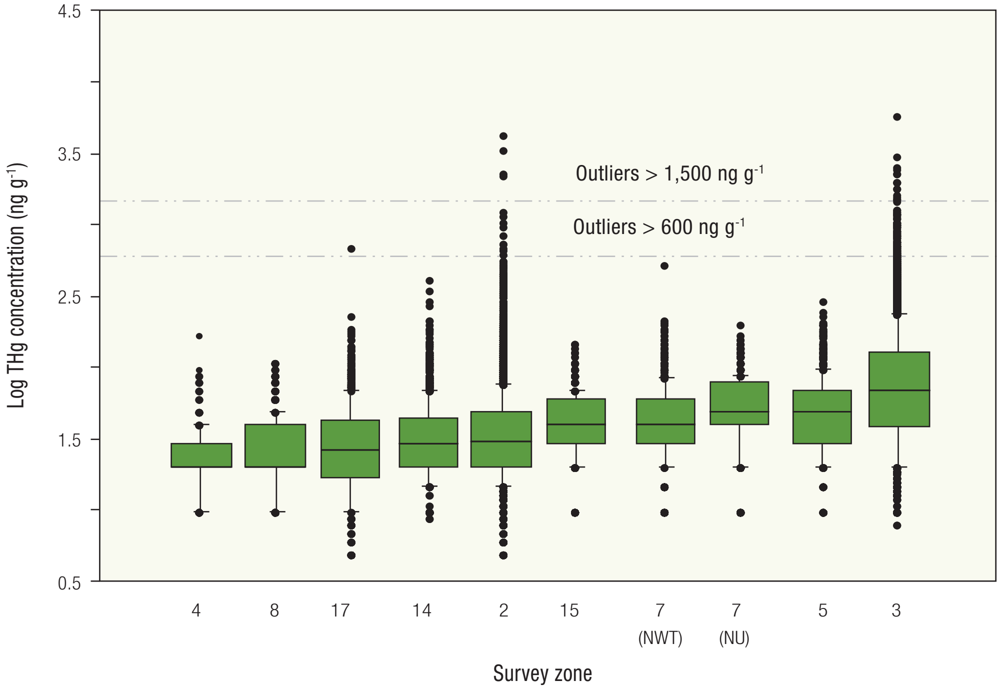

FIGURE 5.9

Box plots of THg concentration (on a logarithmic scale; ng g^{-1}) in stream and lake sediments across northern Canada by survey zone (see Fig. 5.7; GSC 2008). The line inside each box is the median, the upper and lower edges of the box are the 75th and 25th percentiles, and the upper and lower error bars are the 90th and 10th percentiles. Highly elevated outliers for the entire dataset are presented with dashed horizontal lines.

TABLE 5.2 Sediment concentrations of THg (GSC 2008) in lakes and streams as well as modelled rates of atmospheric Hg deposition (Dastoor and Moran 2010), by survey zone and by upland/lowland delineation.

Survey Zone	THg in Lake Sediments (ng g^{-1})						
	Terrain	*n*	Mean	Standard Deviation	Standard Error	Critical Difference	*P*-Value
Bathurst Island, Nunavut (4)	Upland	-	-	-	-	-	-
	Lowland	-	-	-	-	-	-
Baker Lake, Nunavut (8)	Upland	546	32.9	19.4	0.8	2	< 0.0001
	Lowland	686	25.9	16.7	0.6		
Quebec (17)	Upland	635	35.9	38.7	1.5	7.6	0.7555
	Lowland	113	37.1	33.1	3.1		
Labrador (14)	Upland	453	49.3	26.3	1.2	6.2	0.0654
	Lowland	78	43.4	23.7	2.7		
Yukon, West (2)	Upland	80	58.6	48.9	5.5	19.8	0.1922
	Lowland	28	45.5	34	6.4		
Melville Peninsula, Nunavut (15)	Upland	1,539	44.7	19.8	0.5	1.9	0.07
	Lowland	579	42.9	21.7	0.9		
Northwest Territories (7 NWT)	Upland	1,780	52.1	29.3	0.7	2.3	< 0.0001
	Lowland	993	42.3	30.4	1		
Nunauvut (7 NU)	Upland	1,300	61.9	27.2	0.8	3.6	< 0.0001
	Lowland	1,202	50.9	26.9	0.8		
Great Bear Lake, Northwest Territories (5)	Upland	906	59.7	36.7	1.2	4.2	< 0.0001
	Lowland	392	50	32.2	1.6		
Yukon, East (3)	Upland	72	123.2	111.3	13.1	47.8	0.47
	Lowland	24	105.8	66.7	13.6		
Zones with lake & stream data	Upland	605	59.3	39.4	3.2	26.4	0.2373
	Lowland	130	55.4	33.8	5.5		
TOTAL		**735**	**58.6**	**38.4**	**3.6**		
All zones	Upland	7,311	48.5	27.3	1	1.2	< 0.0001
	Lowland	4,095	38.8	23.4	1		
TOTAL		**11,406**	**45**	**25.9**	**1**		

Survey Zone	THg in Stream Sediments (ng g^{-1})						Lake Versus Streams		Atmospheric Hg deposition
	n	Mean	Standard Deviation	Standard Error	Critical Difference	*P*-Value	Critical Difference	*P*-Value	(µg m^{-2} y^{-1})
Bathurst Island, Nunavut (4)	311	25.7	16.4	0.9	1.6	0.421	-	-	7.4
	92	24.1	16.3	1.7					
Baker Lake, Nunavut (8)	-	-	-	-	-	-	-	-	9.1
	-	-	-	-					
Quebec (17)	-	-	-	-	-	-			12.3
	-	-	-	-					
Labrador (14)	997	32.1	32	1.01	5.5	0.0737	3	<0.0001	10
	133	27	17.8	1.55					
Yukon, West (2)	10,212	44.9	79.1	0.8	3.6	< 0.0001	17.1	0.1	8.3
	1,883	37.8	34.4	0.8			12.7	0.2	
Melville Peninsula, Nunavut (15)	-	-	-	-	-	-	-	-	9.8
	-	-	-	-			-	-	
Northwest Territories (7 NWT)	-	-	-	-	-	-	-	-	10.9
	-	-	-	-			-	-	
Nunauvut (7 NU)	-	-	-	-	-	-	-	-	9.9
	-	-	-	-			-	-	

Survey Zone	THg in Stream Sediments (ng g^{-1})						Lake Versus Streams		Atmospheric Hg deposition
	n	Mean	Standard Deviation	Standard Error	Critical Difference	*P*-Value	Critical Difference	*P*-Value	(μg m^{-2} y^{-1})
Great Bear Lake, Northwest Territories (5)	-	-	-	-	-	-	-	-	6.4
	-	-	-	-			-	-	
Yukon, East (3)	9,557	112.7	153.4	1.6	7.7	< 0.0001	35.5	0.56	8.3
	1,719	95.7	127.4	3.1			51.1	0.7	
Zones with lake & stream data	20,766	76.9	124.7	0.9	4.2	< 0.0001	1.9	< 0.0001	9.2
	3,735	64.4	95	1.6			2.9	< 0.0001	
TOTAL	**24,501**	**75**	**120.2**	**1**			**2.1**	**< 0.0001**	
All zones	20,130	77.9	114.5	0.9	4.1	< 0.0001	2.2	< 0.0001	9.2
	3,701	64.7	77.3	1.6					
TOTAL	**23,831**	**75.8**	**108.8**	**1**					

Survey Zone	Temp. July	Temp. January	Annual Precipitation	Ecozone	Dominant Land Cover
	(°C)	(°C)	(mm)		
Bathurst Island, Nunavut (4)	4	-34	-	North Arctic	Barren, none/sparsely vegetation, mostly covered by snow/ice
Baker Lake, Nunavut (8)	11	-32	290	South Arctic	Tundra, wetland, shrubs, barren, none/sparsely vegetation, bryoids
Quebec (17)	10	-23	595	Arctic Cordillera	Barren, none/sparsely vegetation, shrubs, open and sparse coniferous
Labrador (14)	10	-22	614	Arctic Cordillera	Barren, none/sparsely vegetation, shrubs
Yukon, West (2)	14	-23	317	Taiga Cordillera	Mixedwood, broadleaf, coniferous, shrub, herb, exposed land and rock, wetland (treed, herb, shrub), bryoids
Melville Peninsula, Nunavut (15)	7	-32	225	North Arctic	Barren, bare soil with cryptogam crust, none/sparsely vegetative colluvium till, sedge, tundra, shrub
Northwest Territories (7 NWT)	15	-27	328	Taiga Arctic	Mixedwood, broadleaf, coniferous, shrub, exposed land, wetland (treed, herb, shrub)
Nunauvut (7 NU)	13	-29	377	Taiga Shield	Bryoids, shrub, exposed rock with none/sparsely vegetation, dense coniferous
Great Bear Lake, Northwest Territories (5)	12	-28	260	Taiga Shield & Taiga plain	Bryoids, Mixedwood, broadleaf, coniferous, shrub, exposed land, wetland (treed, herb, shrub)
Yukon, East (3)	16	-25	321	Taiga Cordillera & Boreal cordillera	Mixedwood, broadleaf, coniferous, shrub, herb, exposed land and rock, wetland (treed, herb, shrub), bryoids
Zones with lake & stream data	11	-28	370		

Average atmospheric Hg deposition rates for survey zones based on GRAHM modelled estimates varied from 6–12 μg m^{-2} y^{-1} (Table 5.2). Within each survey zone, local concentrations of sediment THg did not correlate with the large-scale GRAHM projections (grid cells of 25 x 25 km). Among survey zones, the lowest rates of atmospheric deposition were for Great Bear Lake (6.4 μg m^{-2} y^{-1}) and Bathurst Island (7.4 μg m^{-2} y^{-1}). However, sediment Hg concentrations in the Great Bear Lake survey zone were higher than on Bathurst Island and similar to sediments from southern Nunavut where there was a higher deposition rate (Table 5.2). Unlike the Great Bear Lake survey zone, no significant difference was found for THg concentrations of upland and lowland locations on Bathurst Island (Table 5.2). The higher sediment THg at Great Bear Lake may be associated with greater vegetation cover (approximately 50%) and higher geogenic Hg sources. Lower water and sediment interactions in the cold and harsh climate of Bathurst Island restrict the geogenic release of Hg to sediments, while atmospheric Hg deposition enhances sediment Hg at a low rate (Givelet et al. 2004). It is also evident that despite higher atmospheric deposition in northern Quebec and Labrador, there was a minimal effect on Hg concentrations in stream and lake sediments there, perhaps due to the sparsely vegetated Arctic cordillera conditions (Table 5.2). The disconnection between atmospheric deposition and THg concentrations in bulk sediment may be due to the importance of watershed transport processes that occur following deposition.

Sediment THg concentrations in streams and lakes from the Yukon were correlated with landscape features and sediment characteristics. Higher sediment THg was found in streams from swamps, flood plains, hills, and mature mountainous terrain relative to streams in undulating terrain, lowlands, geologically young mountainous terrain, and peneplains (a low-relief plain). Terrains with a trellis stream flow pattern had significantly higher sediment THg than streams following rectangular, basinal, and herringbone flow patterns. Sediment THg was slightly but significantly higher in streams flowing through alluvial and organic soils than in streams flowing over bare rock and through outwash and till deposits. Dark-coloured sediments tended to have higher THg than light-coloured sediments. While bedrock and sediment types identified by the GSC were not significant factors, sediment THg concentrations were correlated with concentrations of other trace elements, specifically copper, cadmium, and zinc, and reflected processes of geogenic supply (Nasr et al. 2011). Stream attributes such as channel width, depth, stream order, and flow rate explained little of the variation in sediment THg. Within Yukon's Selwyn Basin, sediment THg was positively correlated with organic matter content, confirming the role of organic matter in capturing and retaining Hg. In this survey zone, sediment THg concentrations were also negatively correlated with the wet-area to catchment-area ratio. Wet area was defined as the area within each catchment where the cartographic depth-to-water index remains within 0.5 m from the soil surface. This correlation was consistent with the general trend of higher THg in sediments from upland, relative to lowland, locations in the Selwyn Basin.

In summary, the analysis of THg in lake and stream sediments across northern Canada indicated that bulk concentrations were affected by geogenic Hg sources and local transport processes. Factors pertaining to local climate, vegetation cover, and topography played an important role. Colder, generally frozen areas had lower sediment THg concentrations, while catchments with greater vegetation cover and water bodies with enhanced organic matter accumulation displayed a greater degree of Hg sequestration. The role of topography varied from exposing trace-metal bearing bedrock along steep slopes to trapping water-mobilized Hg through sedimentation processes in lakes and slow-flowing streams. These factors highlight the importance of: 1) upland-lowland transfer patterns for sediments and other Hg carrying substances such as particulate and dissolved organic matter; 2) flow paths of Hg transfer along upland/wetland transition zones and towards anoxic locations in wetlands and lake bottoms where incoming Hg may be further transformed to MeHg; and, 3) impacts from changes in vegetation cover and climate on sediment Hg in streams and lakes.

5.4.2 Mercury fluxes to Arctic lake sediments

Lake sediments have been shown to be reliable archives for estimating historical Hg fluxes to lakes (Lockhart et al. 2000, Biester et al. 2007, Lindberg et al. 2007). These studies involve the collection of cores from deep points in lakes, followed by analysis of extruded core slices. Dating of slices is generally accomplished using lead-210 isotope (^{210}Pb), a naturally occurring radioactive element that is part of the uranium radioactive decay series, and with cesium-137 (^{137}Cs), a product of nuclear weapons testing which was elevated in the atmosphere in the early 1960s when testing was most intense (Oldfield and Appleby 1984, Blais et al. 1995). The half-life of ^{210}Pb is 22.3 years, and therefore its usefulness as a geochronological tool is limited to the past 100–150 years. Radiocarbon dating (^{14}C) has been routinely applied to older sediment horizons in paleoliminology studies but rarely to study Hg deposition (Cooke et al. 2010).

The NCP supported much of the sediment work conducted in the 1990s on Hg deposition to lake sediments in the Canadian Arctic, which was summarized in Macdonald et al. (2000). Landers et al. (1998) assembled a large dataset of Hg fluxes to Arctic freshwater sediments in Canada and Alaska, as well as the European Arctic, which included fluxes from pre-industrial (before ~1850) and recent periods (~1950 to 1995) for 10 Canadian Arctic or sub-Arctic lakes north of 54° latitude. As part of *CACAR II*, Lockhart (2003) summarized studies published up to the year 2000. Since then, studies by Bindler et al. (2001), Fitzgerald et al. (2005), Outridge et al. (2007), Landers et al. (2008), Muir et al. (2009b), Stern et al. (2009), Carrie et al. (2010), and Cooke et al. (2010) have reported on Hg in sediments from 40 lakes in Alaska, northern Canada, and West Greenland. The following is an assessment of latitudinal and longitudinal trends in Hg fluxes to these 40 lakes, the majority (28) of which are located in northern Canada.

Spatial and temporal trends of Hg fluxes inferred from lake sediments

Sediment Hg fluxes (µg m^{-2} y^{-1}) were determined for each dated core slice by multiplying the measured THg concentration (µg g^{-1}) by the estimated sedimentation rate (g m^{-2} y^{-1}) for that slice. Anthropogenic Hg fluxes (ΔHgF, µg m^{-2} y^{-1}) were calculated as the difference in the core between the recent flux and the pre-industrial flux. For most cores, the Hg flux in the pre-industrial era was estimated with sediment horizons dated to the

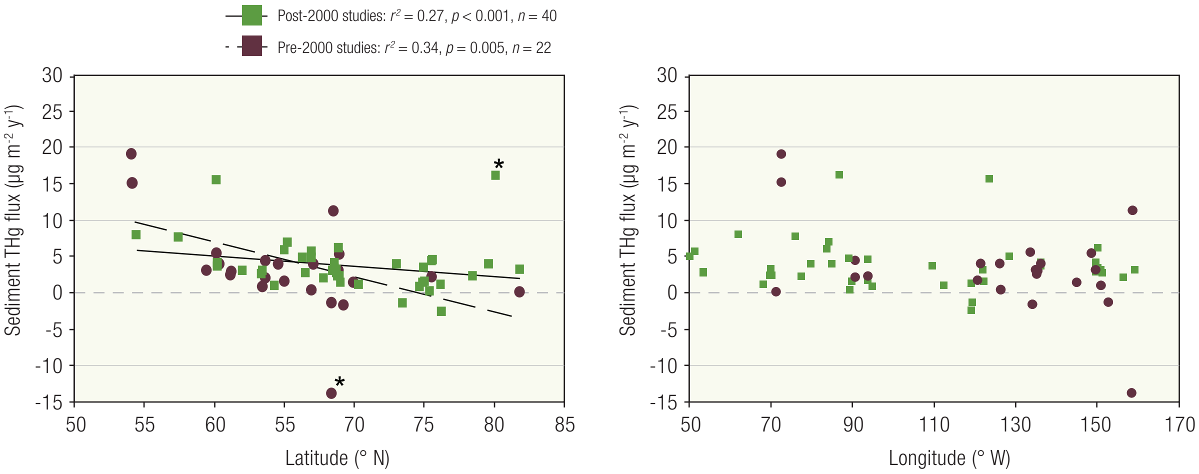

FIGURE 5.10

Anthropogenic fluxes of THg (ΔHgF) in dated sediment cores from Arctic and sub-Arctic lakes in Canada, Alaska, and West Greenland. Two results with asterisks were omitted from the regressions. Data are separated for cores reported (and in most cases sampled) post-2000 (references in text) and pre-2000 cores reported by Landers et al. (1998) and Lockhart et al. (1998).

1820 to 1870s and the recent flux to the period of 1990 to 2005, typically represented by the top 1–2 cm of most Arctic and sub-Arctic cores. Results were, in most cases, corrected for particle focussing, changing sedimentation rates, and/or erosional inputs (Fitzgerald et al. 2005, Muir et al. 2009b, Carrie et al. 2010). In post-2000 studies, ΔHgF ranged from -2.6 to 16 µg m^{-2} y^{-1} (geometric mean: 3.6 µg m^{-2} y^{-1}). The negative values reflect higher estimated Hg fluxes in pre-industrial horizons, which were observed in only 2 of 40 lakes. Latitudinal and longitudinal trends of ΔHgF for the 40 lakes are presented in Fig. 5.10. Omitting one very high flux for Lake AX-AJ (Muir et al. 2009b), ΔHgF declined weakly with latitude (r^2 = 0.27, p < 0.001, n = 40) but was not significantly correlated with longitude (Fig. 5.10).

In earlier studies, Landers et al. (1998) and Lockhart et al. (1998) reported similar fluxes and latitudinal trends for ΔHgF in a combined total of 23 Arctic and sub-Arctic lakes north of 54° in Canada and Alaska. They corrected for focussing but not for erosional inputs or increasing sedimentation. These pre-2000 results are also shown in Fig. 5.10 where ΔHgF ranged from -14 to 19 µg m^{-2} y^{-1} (geometric mean: 3.0 µg m^{-2} y^{-1}). A steeper decline in ΔHgF with latitude was found in the earlier studies after omitting one negative result (Feniak Lake; Landers et al. 1998) (r^2 = 0.34, p = 0.004, n = 23), which reflects higher fluxes in two sub-Arctic cores (Fig. 5.10). Overall, the combined results of earlier and more recent studies show quite uniform Hg fluxes across the Canadian Arctic above 65° N.

Mercury profiles in almost all dated sediment cores show increases in ΔHgF post-1900 as illustrated in Fig. 5.11 for 14 sub-Arctic and 18 Arctic lakes studied by Muir et al. (2009b). Fitzgerald et al. (2005) reported a similar trend in five lakes in northern Alaska. The increase, particularly during the first half of the 20th Century, coincides with other reports of increasing Hg in the Arctic such as trends in hard tissues of marine animals (Dietz et al. 2009). The sub-Arctic cores, as well as those from northern Alaska, show a levelling off of Hg fluxes starting in the 1980s. Lindberg et al. (2007) noted that while there was good evidence from lake sediment records for large (30–50%) declines in Hg deposition in urban areas in the Northern Hemisphere due to local reductions in Hg(II) emissions, reductions in anthropogenic inputs at remote locations would be much less pronounced because of a dominant input from the global Hg pool. This pattern is less apparent in the High Arctic cores presented here (Fig. 5.11) and in other studies at the same latitude by Cooke et al. (2010) and Outridge et al. (2007) which show continuing increases in Hg fluxes.

Agreement of Hg fluxes inferred from lake sediment with modelled deposition

Atmospheric deposition fluxes were estimated with the GRAHM model (section 3.6), which ranged from about 2.2–9.5 µg m^{-2} y^{-1} over the area from 60° N to 83° N in the Canadian Arctic (Muir et al. 2009b, Durnford et al. 2010). The Danish Eulerian Hemispheric model (DEHM) predicted annual Hg deposition,

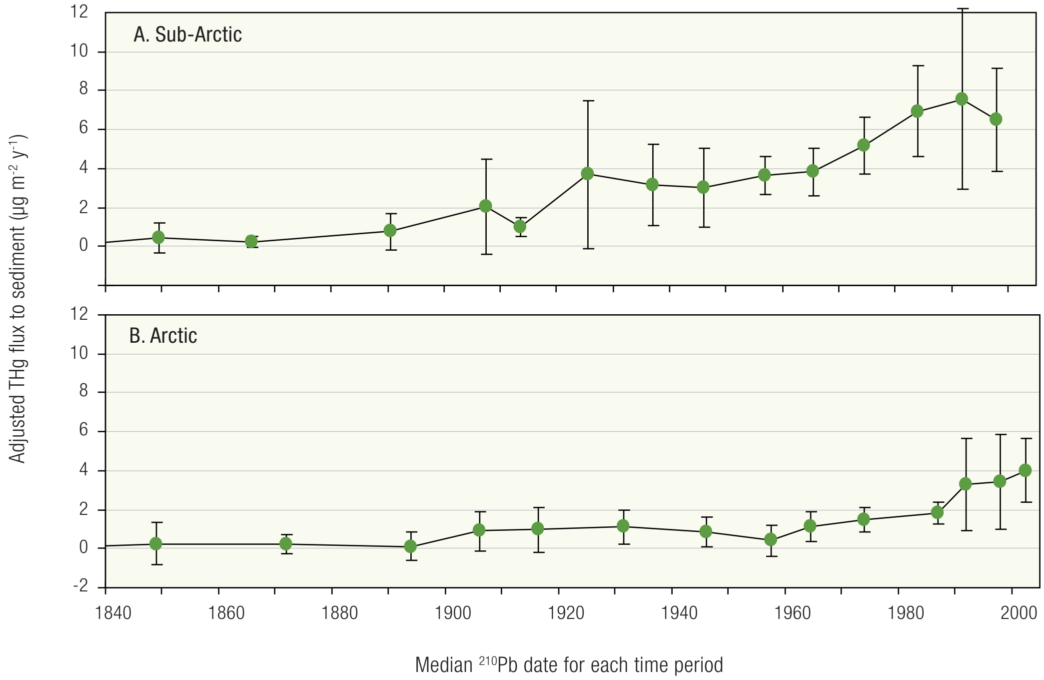

FIGURE 5.11

Average historical profiles of anthropogenic Hg deposition fluxes adjusted for changes in particle focussing and sedimentation (ΔHgF_{adj}, µg m^{-2} y^{-1}, ± 95% confidence limits) in sub-Arctic (*n* = 14, 51–64° N) and Arctic (*n* = 18, 65–83° N) sediment cores over time intervals of 5–20 years (data from Muir et al. 2009b).

including AMDEs, that ranged from 6–12 µg m^{-2} y^{-1} in the Canadian Arctic Archipelago (Christensen et al. 2004). Thus, there is relatively good agreement between modelled terrestrial Hg fluxes and measured anthropogenic fluxes to fresh waters.

Interpretation of Hg fluxes to lake sediments

While all evidence points to increasing Hg fluxes, especially in High Arctic lakes, there is much debate on whether this implies that Hg from anthropogenic sources, such as from recent increases in Asian Hg emissions, is entering the lakes in greater amounts or whether other factors such as increased sedimentation and higher primary productivity driven by climate change are attenuating Hg inputs from the atmosphere or the lake catchment. Recent studies have examined these various factors influencing Hg profiles in Arctic lake sediments.

Much focus has been on historical variations in sedimentation rates estimated from ^{210}Pb dating of the cores, which may be affected by erosion, aeolian inputs, or by artefacts related to the dating technique. Fitzgerald et al. (2005) measured pre-industrial (pre-1850) sedimentation rates that were one to five times lower than late 20th Century rates in five Alaskan Arctic lakes north of the treeline. They concluded that 11–64% of Hg in recent sediments was from soil erosion. In a core from Lake DV-09 (Devon Island, Nunavut), Outridge et al. (2005) found significant correlations between aluminum and zinc, as well as between aluminum and Hg, and attributed a significant fraction of Hg input to local geological sources via weathering and runoff from melting snow. Lindeberg et al. (2006) found that large fluctuations in Hg concentrations in pre-19th Century sediments of lakes in west Greenland were related to changes in the influx of material from regional aeolian activity. Muir et al. (2009b) also found higher recent sedimentation rates in 16 of 31 Arctic and sub-Arctic lakes. However, those authors concluded that the greater sedimentation did not appear to have a large erosional component because lithogenic elements—aluminum and zinc—were not correlated with sedimentation rate. Cooke et al. (2010) have argued that the ^{210}Pb dating method has overestimated sedimentation rates for horizons in the mid-19th Century and earlier. Using a composite age-depth model incorporating ^{210}Pb and ^{14}C dates, they estimated pre-industrial Hg fluxes were 0.25–0.30 µg m^{-2} y^{-1} in two High Arctic lakes, or about five times lower than most other estimates. The higher modern sedimentation rates could also be due to bioturbation of the ^{210}Pb activity profile near the sediment surface or to diagenetic dilution of ^{210}Pb from accumulation of iron oxides at the surface (Gubala et al. 1990). Finally, it is worth noting that ^{210}Pb activities in Arctic sediments are very low in high-latitude lake sediments. Frozen soil retards the release of the parent isotope radon-222, and lake-ice cover limits the efficiency with which atmospheric ^{210}Pb is transferred to lake sediments (Wolfe et al. 2004). These studies highlight the challenges in accurately estimating sedimentation rates.

The role of climate change in modifying Hg fluxes into lake sediments has recently been an area of active research (Outridge et al. 2005, 2007, Stern et al. 2009, Carrie et al. 2010). Large increases in algal productivity have occurred over recent decades in Arctic lakes (Gajewski et al. 1997, Michelutti et al. 2005). There is evidence that these increases may have markedly enhanced the rate of Hg scavenging by algae and transfer of Hg into lake sediments (Outridge et al. 2005, 2007), in a process analogous to the well-established phytoplankton *biological pump* for vertical Hg fluxes in oceans (Cossa et al. 2009, Sunderland et al. 2009). Strong relationships between fluxes or concentrations of Hg and algal carbon (with correlation r^2 values > 0.75) were found in four investigated Canadian Arctic lakes (Outridge et al. 2007, Stern et al. 2009, Carrie et al. 2010). For Kusawa Lake in the Yukon and Lake DV-09 on Devon Island (Nunavut), it was estimated that no more than 22–29% of the 20th Century increase in Hg concentrations could be attributed to increased anthropogenic Hg inputs because of this climate-related effect (Outridge et al. 2007, Stern et al. 2009). These recent findings, though requiring further investigation and testing, have implications if lake sediments are used to test the validity of model estimates for atmospheric deposition rates.

Evidence for the algal Hg scavenging hypothesis is based, in part, on a detailed characterization of organic carbon compounds and their profiles in lake sediment cores. Total organic carbon by itself may be a poor measure of the labile, thiol-rich algal organic matter that is believed to be involved in Hg scavenging (Sanei et al. 2010). Outridge et al. (2005, 2007) reported increasing total organic carbon in Amituk Lake and Lake DV-09, but the relative increase in algal-derived carbon, estimated using a kerogen carbon parameter (S2), was markedly greater (760% since 1854). These S2 carbon increases were highly correlated with diatom valve abundance, suggesting good preservation of the historical organic matter. Greater organic carbon could be partly due to increased production of autochthonus organic carbon in the littoral or pelagic zones of lakes (Michelutti et al. 2005, Smol et al. 2005). Another process that could generate a profile of increasing organic carbon in recent horizons is the progressive loss of carbon following burial, as shown by Gälman et al. (2008) for varved lake sediment in northern Sweden. However, based on careful characterization of the organic matter using organic geochemistry and petrographic techniques, as well as the agreement between trends in Hg and diatom abundance, decomposition is unlikely to explain the similar down-core profiles of algal carbon and Hg in the Arctic and sub-Arctic lakes studied by Outridge et al. (2005, 2007), Carrie et al. (2010), and Stern et al. (2009). Other algal productivity indicators, such as total diatom valve abundance and concentrations of total pigment and biogenic silica, corroborate the occurrence of profound, widespread productivity increases in Arctic lakes as a consequence of earlier melting and ice-out under warming conditions (Gajewski et al. 1997, Michelutti et al. 2005, Smol et al. 2005).

Further testing of the algal Hg scavenging hypothesis was recently conducted by comparing profiles of Hg with those for algal carbon and species composition in dated sediment cores from 14 lakes spanning latitudinal and longitudinal gradients across the Canadian Arctic (Kirk et al. 2011). Fluxes of THg to sediments increased during the Industrial Era (approximately post-1850) in 11 of the 14 lakes (post-industrial Hg fluxes corrected for particle focussing, ΔHgF = 2–24 μg m^{-2} y^{-1}) (Fig. 5.12; Kirk et al. 2011). To tease apart Hg inputs from catchment erosion and other sources—which include anthropogenic atmospheric deposition—flux values were adjusted for post-industrial changes in sedimentation rate (ΔHgF_{adj}) following the methods of Muir et al. (2009b). Fluxes of THg originating from catchment-independent factors such as atmospheric deposition increased since industrialization in all 14 lakes after correction for catchment contributions (ΔHgF_{adj} = 0.3–16 μg m^{-2} y^{-1}) (Kirk et al. 2011). Several of these lakes also showed post-industrial shifts in algal assemblages consistent with climate-induced changes (Fig. 5.12). For example, in three lakes where species-level diatom analysis was carried out, the benthic *Fragilaria* spp.—typical of ice-dominated, oligotrophic High Arctic lakes—was replaced by planktonic, epiphytic or other benthic species characteristic of longer ice-free seasons. In addition, greater species diversity (measured by Hill's N2 index; Hill 1973) was generally observed in more recent sediment layers. In 11 lakes, sediment profiles showed post-1850 increases in algal carbon flux (ΔS2, corrected for particle focussing), suggesting that lake primary productivity has recently increased at the majority of the sampled sites (ΔS2 = 0.1–4 g m^{-2} y^{-1}) (Kirk et al. 2011).

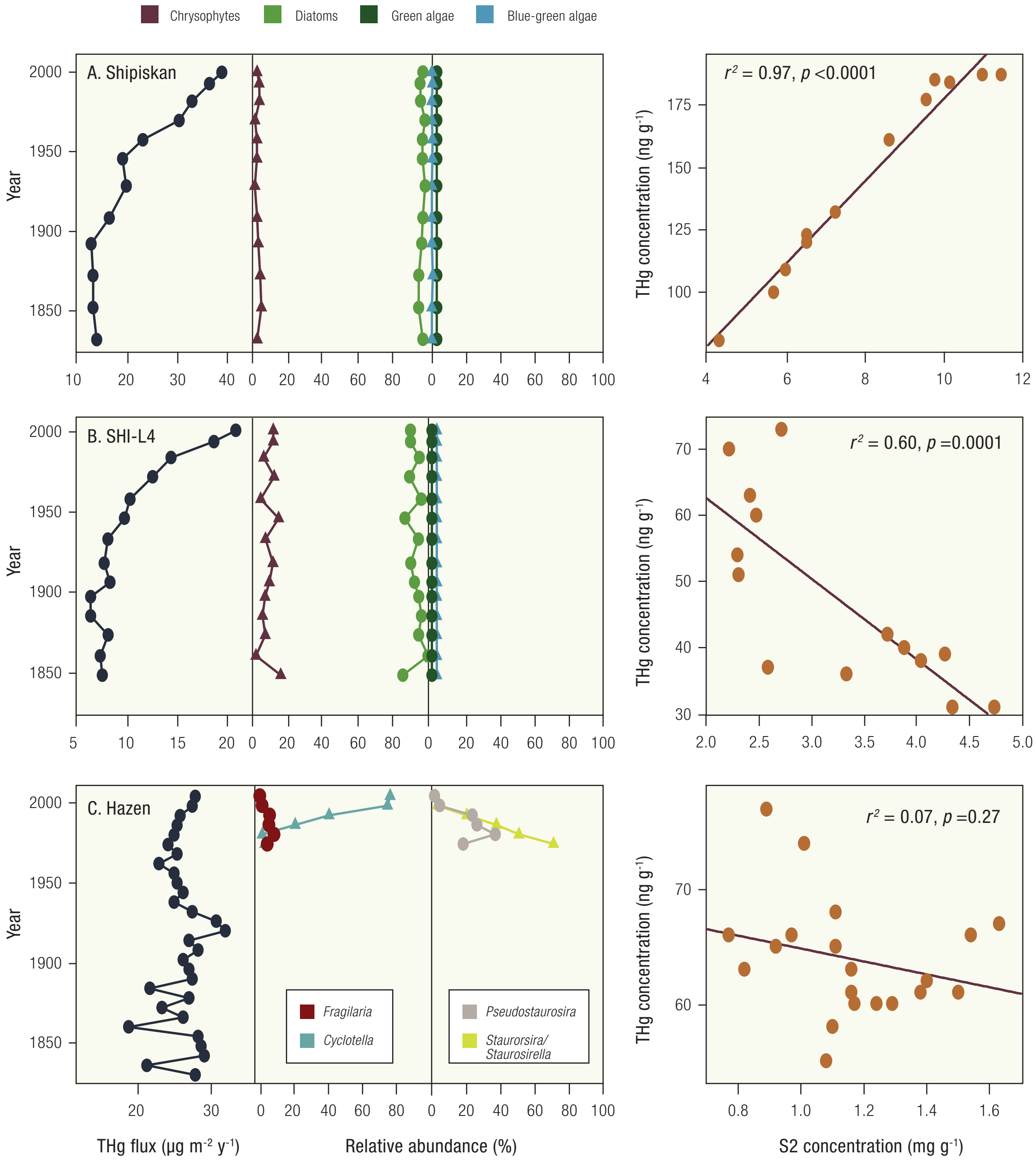

FIGURE 5.12

Examples of different Hg and algal profiles found in dated sediment cores from 3 of 14 Canadian Arctic lakes (Shipiskan, SHI-L4, Hazen). For each lake, sediment THg fluxes adjusted for particle focussing (ΔHgF), relative abundance (%) of microfossils—including either diatoms, chrysophytes, green algae and blue-green algae or predominant diatom species—and the relationship between THg and S2 carbon concentrations are presented. Reprinted with permission from Kirk et al. (2011). © American Chemical Society.

Eric Loring

Other studies (Outridge et al. 2007, Stern et al. 2009, Carrie et al. 2010) have interpreted significant relationships between THg and S2 concentrations in Arctic sediment as support for the algal scavenging hypothesis. However, in six of the 14 lakes surveyed by Kirk et al. (2011), no THg:S2 relationship was observed, and in one lake, a significant negative relationship was observed due to increased THg and decreased S2 carbon deposition during the Industrial Era (Fig. 5.12). In two of the seven lakes where a significant THg:S2 correlation was observed, the relationship was interpreted as an artefact of post-depositional S2 degradation. In six of the seven lakes where a significant positive THg:S2 correlation was observed, algal assemblages either did not change through time or the timing of the shifts did not correspond to changes in Hg deposition. Kirk et al. (2011) suggested that, although Arctic lakes are experiencing a myriad of changes including increased Hg and S2 deposition or shifting algal assemblages, increased lake primary productivity may not be driving changes in Hg fluxes to sediments.

In summary, the scientific literature is divided over the explanation for increased Hg fluxes in Arctic lake sediment cores. Using their revised pre-industrial sedimentation rates, Cooke et al. (2010) concluded that ratios of recent versus pre-industrial Hg flux are about 12 rather than factors of 1–3 that have been reported for most Arctic lakes (Landers et al. 1998, Fitzgerald et al. 2005, Muir et al. 2009b). This higher value is in line with observations for the Upper Freemont Glacier in Colorado (Schuster et al. 2002) and with increases in Hg in hard tissues from Arctic animals since around the 14th to 16th Century (Dietz et al. 2009). Other authors have argued that anthropogenic Hg inputs are overestimated by approximately four- to five-fold based on relationships of S2 carbon and Hg concentrations (Outridge et al. 2007, Stern et al. 2009). However, there is consensus that climate change, which is occurring rapidly in the Arctic, is having a major influence on sedimentation rates. Mean annual snowfall has increased in the central Canadian Archipelago where most of the High Arctic cores have been collected (Michelutti et al. 2003), suggesting more snowmelt runoff is leading to greater erosion inputs in some catchments. The disappearance of shallow ponds on islands of the Canadian Arctic Archipelago has been attributed to increased evaporation to precipitation ratios (Smol and Douglas 2007b), and more rapid drying of lake catchments could result in greater aeolian inputs for some lakes. There have been increases in primary production (Michelutti et al. 2005) and inorganic sedimentation (Outridge et al. 2005, Thomas and Briner 2009). Thus, additional studies are needed to explain increasing Hg fluxes in Arctic lakes.

5.4.3 Impact of permafrost thawing on Hg transport to Arctic lakes

Thawing permafrost is already changing the Arctic landscape and will likely accelerate in coming decades (ACIA 2005, Lantz and Kokelj 2008). Consequent to thaw, significant changes in hydrology, organic carbon pathways, and freshwater resources are also expected (ACIA 2005). Currently, nearly half of the land surface in Canada is underlain by some form of permafrost (Smith and Burgess 1999). The extent to which thawing permafrost will change the amount of Hg entering freshwaters in the Canadian Arctic is a significant gap in our knowledge of how ecosystems will respond to climate warming.

Recent studies show that the influx of slump material from degrading permafrost into freshwater systems will introduce a variety of materials that were previously trapped in the frozen ice and soil (Kokelj and Burn 2005, Kokelj et al. 2005). As the active layer deepens and more hydrological flow pathways develop in the permafrost, greater geochemical weathering from drainages is expected (Hobbie et al. 1999, Kokelj et al. 2005, Prowse et al. 2006). The inputs will result in changes to freshwater chemistry, including higher concentrations of major ions and decreases in DOC (Kokelj et al. 2005). Studies of permafrost peatlands in northern Sweden indicate that thawing and erosion of thermokarst can release significant amounts of Hg to sub-Arctic lakes (Klaminder et al. 2008, Rydberg et al. 2010).

The consequences of thawing permafrost on the Hg cycle are complex and may be linked to several environmental changes including the release of old soil organic carbon previously locked in permafrost, the alteration of plant growth and the movement of newly produced terrestrial organic carbon (e.g., switch from tundra to willows, and deepening of the active layer), and the alteration of aquatic productivity due to change in the light regime or hydrology (Outridge et al. 2007, Smol and Douglas 2007a). Connections with the Hg cycle will, accordingly, include the exposure and release of Hg presently archived in frozen ground, the alteration of methylating activity for Hg cycling within northern aquatic systems (Macdonald and Loseto 2010), and change in the burial efficiency of Hg in aquatic sediments (Outridge et al. 2007). Depending on how exactly the changes occur in a given location, these alterations could lead to increased exposure of food webs to MeHg (e.g., by creating new wetlands) or decreased exposure (e.g., by the drying out of shallow standing waters, by enhanced burial in sediments, or by growth dilution). Clearly, progress in understanding the risks Hg present to aquatic food webs under a changing climate also requires a detailed understanding of the changes occurring in the production and transport of organic matter.

Since 2008, the transport of Hg from thawing permafrost has been investigated in the Mackenzie Delta region near Inuvik, where thaw slumping is occurring on a large scale (Blais 2010). A series of lakes with catchments disturbed by permafrost melting were paired to undisturbed lakes of similar size in adjacent catchments. This paired-lake design allowed for a case-control analysis of lakes where retrogressive thaw slumps were present and absent. Preliminary results suggest that thaw slump development increases inorganic sedimentation rates in lakes while decreasing concentrations of organic carbon, THg, and MeHg in sediments. Sediment cores drawn from four lakes with permafrost thaw slump development on their shorelines had higher sedimentation rates (of largely inorganic material) and lower concentrations of THg, MeHg, and organic carbon in surface sediments compared to four lakes where thaw slumps were absent. Concentrations of THg and MeHg were positively correlated with total organic carbon and labile algal-derived (S2) carbon due to an association between the movement of organic matter and Hg accumulation in lake sediments.

A preliminary assessment of Hg in water and amphipods from these same lakes suggested that lakes with thaw slump development had lower concentrations of THg in water and amphipods, but higher Hg bioaccumulation factors in amphipods. Blais (2010) proposed that the higher bioaccumulation of THg in amphipods from lakes with thaw slump development may result from: 1) increased Hg bioavailability due to lower concentrations of large and hydrophilic Hg complexes with DOC; and, 2) less algal biodilution due to lower nutrients and lower phytoplankton biomass in lakes with thaw slump development. Further research is in progress to investigate links between permafrost melting, organic carbon cycling, and Hg bioaccumulation in food webs of Arctic lakes in the Mackenzie Delta region.

5.5 Biogeochemical transformations of Hg

5.5.1 Microbial transformations of Hg

Microbes actively alter Hg speciation in the environment. It is thought that two main reactions compete for the inorganic divalent Hg substrate: 1) reduction of Hg(II) to elemental Hg(0); and, 2) methylation of Hg(II) to MeHg. Methylmercury can also be demethylated via microbial pathways. Molecular tools allowing the quantification of intracellular Hg, such as luminescent biosensors hosted by bacteria, suggest that a fraction of the Hg deposited by AMDEs in the Arctic is bioavailable (Scott 2001, Lindberg et al. 2002, Larose et al. 2010, 2011). In this context, bioavailable Hg is the inorganic form accessible and internalized by microbes and potentially used as substrate for MeHg production. The link between bioavailable Hg deposited during AMDEs and its methylation remains unclear (Dommergue et al. 2009). Recent evidence suggests that wet deposition may contribute a higher proportion of bioavailable Hg to Arctic snow packs than AMDEs (Larose et al. 2011). Whether it is in snow, water, sediments, or in littoral or pelagic zones, the predominant location(s) in freshwater ecosystems where methylation occurs have not been identified.

In temperate environments, Hg methylation is mostly a microbial process driven by the activity of sulphate- and iron-reducing bacteria (Gilmour et al. 1992, Fleming et al. 2006). Currently for the Arctic, further investigation is required on both the chemical speciation of Hg and the physiology of psychrophilic microbes, which are adapted to cold environments. It may be misleading to assume that what we know about mesophilic (warm-adapted) microorganisms encountered in temperate locations directly applies to cold-adapted microbes or to mesophilic organisms metabolizing at low temperatures. Likewise, little information exists on which Hg species are potentially available for uptake in the cells of microbial methylators in the Arctic (i.e. Hg(II) complexes with inorganic and organic compounds; Schaefer and Morel 2009).

Measurements of MeHg production rates are not yet available for aquatic ecosystems in the Canadian Arctic. However, projects currently underway will provide first estimates in the near future. Laboratory incubations in Arctic wetland soils indicate that methylation can occur (section 4.5). Elevated MeHg concentrations in High Arctic pond waters and mass balance modelling suggest that shallow water bodies can be important methylation sites (section 5.6). Sediment methylation rates were measured in Alaskan lakes and found to be the primary source of MeHg to those ecosystems (Hammerschmidt et al. 2006). Sulphur profiles in lake sediments in the Canadian High Arctic indicate that sulphate reduction is occurring (Drevnick et al. 2010). Interestingly, the authors reported increased concentrations of reduced sulphur in sediment accumulated after about 1980. This suggests a recent acceleration of sulphate reduction with potential implications for Hg methylation in these systems (Drevnick et al. 2010).

Very little research has been conducted on microbial-mediated processes of Hg(II) oxidation and reduction in Arctic fresh waters. Poulain et al. (2007) showed that on Cornwallis Island, filtration of snowmelt samples through 0.2 μm porosity filters did not alter the rate of Hg reduction when exposed to sunlight or kept in the dark. This result was in stark contrast with what was observed in salt water (section 6.7.2), but is in agreement with work performed at lower latitudes that showed Hg(0) production was mostly photomediated in lake surface waters (Poulain et al. 2004). Further work is required to evaluate the contribution of microbes to the redox cycling of Hg in Arctic fresh waters.

5.5.2 Photochemical transformations of Hg

Photochemical transformations of Hg play a key role in modifying the mobility and toxicity of Hg in air, snow, seawater, and fresh water. These transformations are mediated through a variety of direct and indirect processes that can be grouped in two categories: processes affecting the formation of volatile Hg(0) (i.e. redox processes) and processes affecting the balance between methylation and demethylation. Most studies on the photochemistry of Hg have been conducted at field sites in temperate regions or in the laboratory, and some recent reviews provide a good overview of the results (Zhang 2006, Ariya et al. 2009).

Photoreduction and photooxidation

Species of Hg(II) are generally soluble and reactive in water, whereas elemental Hg(0) is volatile and not very reactive. As a result, photoreduction of Hg(II) species into Hg(0) will promote Hg evasion from Arctic freshwater ecosystems and Hg(0) oxidation leads to its retention. Only two studies have directly evaluated photochemical reaction rates for Hg reduction and oxidation in polar fresh waters (Table 5.3). In those studies by Amyot et al. (1997) and Poulain et al. (2007), water samples were incubated under solar radiation in transparent or opaque Teflon bottles. Some transparent bottles were wrapped in different light filters in order to isolate the effect of a given waveband. Elemental Hg(0) was not directly measured. Instead, all volatile Hg species dissolved in water were purged and analyzed. The resulting operationally defined fraction is referred to as dissolved gaseous Hg (DGM), which is often assumed to be mainly composed of Hg(0) in fresh water. In Amyot et al. (1997), formation rates of DGM were measured for three Arctic lakes and one wetland located on Cornwallis Island. The rate of DGM formation was shown to be controlled by: 1) the intensity of solar radiation, particularly in the ultraviolet (UV) range of the spectrum; and, by 2) the concentration of available photoreducible Hg(II) complexes. In Poulain et al. (2007), the authors measured *in situ* net reduction rates during spring and summer in a series of streams and ponds along a salinity gradient covering four orders of magnitude. Production of DGM, normalized for the amount of photons received, declined dramatically with increasing salinity. These authors further provided evidence that biogenic organic materials produced by algae favoured the oxidation of Hg(0) in marine lagoons, but not in freshwater ponds. Overall, the Arctic data from Poulain et al. (2007) indicate that aqueous Hg in inland fresh waters is more prone to photoreduction—and therefore to loss by evasion—than in nearby marine systems.

TABLE 5.3 Water concentrations and production rates of DGM in Arctic freshwater ecosystems.

Location	Date of Sampling	*n*	pH	DOC (mg L^{-1})	DGM Concentration (fM)	Production Rate of DGM (fM h^{-1})	Source
Lakes							
Cornwallis Island (Nunavut)	1994–1995	5					1
Amituk Lake	08/03/1994		8.2	1.6	158 ± 15	0	1
Amituk Lake	08/05/1994		8.2	1.6	203 ± 21	0	1
Merretta Lake	08/14/1994		8.0	2.3	303 ± 6	16	1
North Lake	08/15/1994		8.3	1.1	290 ± 28	17	1
North Lake	07/27/1994		8.3	1.1	617 ± 37	24	1
Toolik Field Station (Alaska)	2000	9	7.9–8.3	2.7–5.8	180		2
Toolik Field Station (Alaska)	2000–2002	27	8.0–8.3	2.7–4.3	198 ± 87		3
Wetlands							
Cornwallis Island (Nunavut)	1994–1995	1	8.1–8.2	1.8	238	28	1
Freshwater ponds, streams, snowmelt, and coastal water							
Cornwallis Island (Nunavut)	2004	12	7.5–8.5			250*	4

n = number of sites sampled, DOC = dissolved organic carbon, fM = femtomoles L^{-1}

* Units in fM $\text{mole}_{\text{photon}}^{-1}$ m^{-2}

Sources: 1 = Amyot et al. (1997); 2 = Tseng et al. (2004); 3 = Fitzgerald et al. (2005); 4 = Poulain et al. (2007).

In addition to these field experiments, freshwater concentrations of DGM have only been measured at two Arctic locations, specifically in lakes near the Toolik Field Station (Alaska, USA) and in lakes, ponds, and streams on Cornwallis Island (Table 5.3). In a set of ten lakes located on the tundra of Alaska's North Slope near Toolik, DGM concentrations ranged from 75–391 fM (i.e. 10^{-15} M) with a mean of 198 ± 87 fM (Tseng et al. 2004, Fitzgerald et al. 2005). These lakes exhibited supersaturation—exceeding 100% saturation—with Hg(0) ranging from 234–1220% under all conditions. Light attenuation and hence DOC were the best predictors of surface DGM levels. This suggests that DGM production was dominated by photochemical processes (Tseng et al. 2004). Surface DGM levels in lakes on Cornwallis Island ranged from 158–617 fM and exhibited supersaturation with values typically between 100–200% (Amyot et al. 1997). The lower supersaturation of Cornwallis lakes is partly due to the colder temperatures encountered in these systems (1–3°C) compared to Alaskan lakes (12–15°C).

Photodemethylation and photomethylation

Photodemethylation of MeHg is a significant, although often ignored, mechanism affecting MeHg pools in temperate lakes (Sellers et al. 1996, Lehnherr and St. Louis 2009). In polar regions, few studies have been published on *in situ* measurements of this phenomenon (although see section 5.6). Hammerschmidt and Fitzgerald (2006) conducted field experiments in Toolik Lake (Alaska, USA) during which they incubated filtered and unfiltered water samples amended with MeHg chloride (final concentration: 3 ng L^{-1}) under epilimnetic conditions. The decomposition of MeHg was shown to be exclusively abiotic and mediated by light. Photodecomposition rates were related to MeHg concentrations and to the intensity of photosynthetically active radiation (PAR). The estimated loss of MeHg to photodecomposition in this lake accounted for about 80% of the MeHg mobilized annually from *in situ* sedimentary production—the main source to Toolik Lake. The estimated photodecomposition flux of MeHg was 1.3 µg m^{-2} y^{-1} over an ice-free period of 100 days. In their flux calculations, Hammerschmidt and Fitzgerald (2006) assumed that PAR was responsible for photodemethylation. More recently, Lehnherr and St. Louis (2009) revisited this dataset and calculated a photodecomposition flux of 0.57 µg m^{-2} y^{-1}, assuming that the bulk of photodemethylation is caused by UV radiation rather than PAR. A lower estimate of photodecomposition flux was obtained by Lehnherr and St. Louis (2009) because UV light is more rapidly attenuated in the water column compared to PAR. Further work by

Hammerschmidt and Fitzgerald (2010) suggests that iron also plays an important role in the photochemical decomposition of MeHg in Alaskan lakes.

There is some evidence that MeHg can be formed in the water column by photomethylation of Hg(II) in certain lakes at temperate latitudes (Siciliano et al. 2005). This photoproduction is affected by the concentration and type of dissolved organic matter. However, no field study has been conducted on this topic in Arctic freshwater environments.

5.6 Mass balance modelling

5.6.1 Mass balance of MeHg in High Arctic ponds

Wetland and pond waters in the Canadian High Arctic tend to have higher concentrations of MeHg compared to larger lakes (St. Louis et al. 2005; section 5.3.1), and it has been suggested that MeHg is actively being produced in those systems (Loseto et al. 2004a, 2004b, St. Louis et al. 2005).

The production of MeHg was quantified by Lehnherr (2010a) in two wetland ponds in the Lake Hazen region using a mass-balance approach for a three-week period (July 6–23, 2005) during the ice-free season. The Lake Hazen region, located on northern Ellesmere Island within Quttinirpaaq National Park, experiences anomalously warm summer conditions for its latitude due to its continental location on the lee side of the Grant Land Mountains. The summer melt period, promoted by continuous daylight, extends for eight to ten weeks and results in a greater diversity and abundance of vegetation compared to surrounding areas (Soper and Powell 1985). However, the region receives very little precipitation, approximately 95 mm annually (Thompson 1994). Wetland sites were characterized by a wet sedge-grass meadow that surrounds a central pond containing aquatic mosses and emergent vegetation.

The two study sites (Ponds 1 and 2) lacked inflow and outflow streams. Their hydrological isolation allowed for the calculation of a simple mass balance budget of MeHg by quantifying MeHg inputs from wet atmospheric precipitation, MeHg removal by photodemethylation, and MeHg storage in the water column compartment (Fig. 5.13).

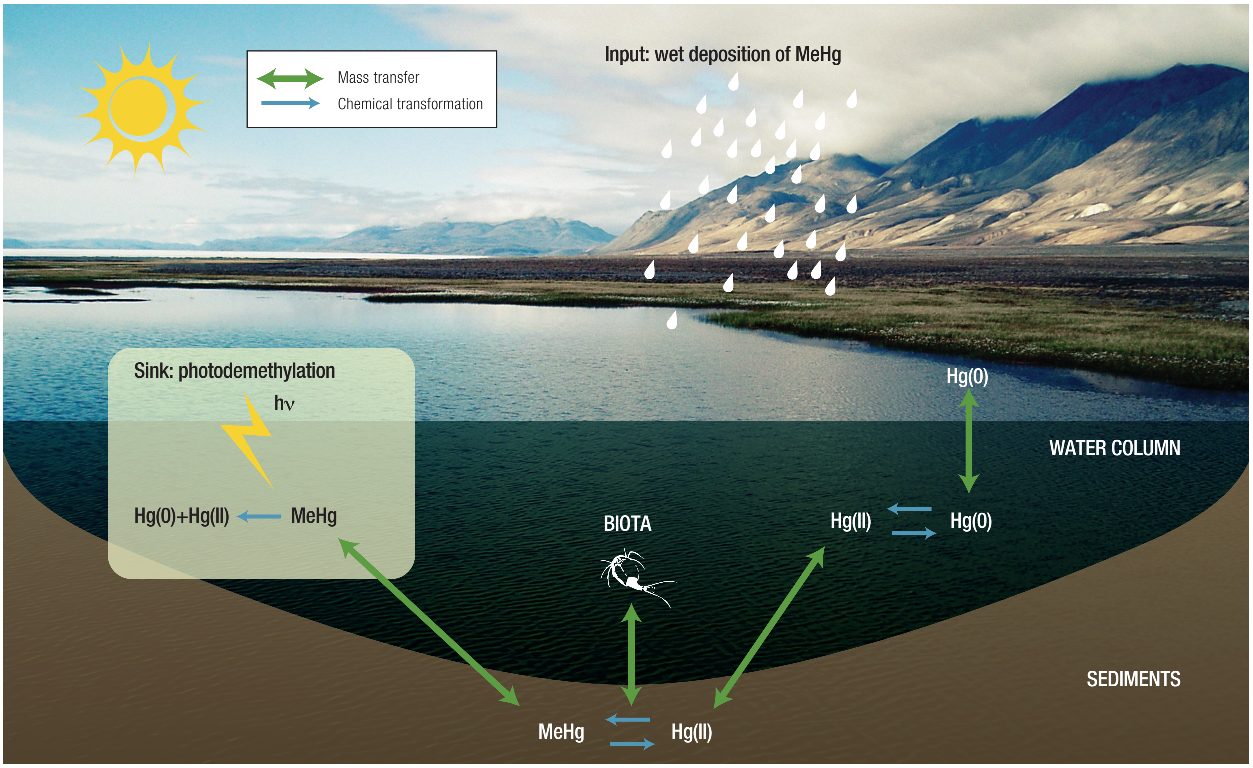

FIGURE 5.13

Schematic of the MeHg mass balance study design for two wetland ponds in Quttinirpaaq National Park on Ellesmere Island (Lehnherr 2010a).

TABLE 5.4 MeHg fluxes in two wetland ponds of Quttinirpaaq National Park on Ellesmere Island (Lehnherr 2010a).

		Δ MeHg	**Precipitation**	**Photodemethylation**	**Total MeHg Production**
Pond 1	µg	66 ± 18	0.3 ± 0.03	188 ± 53	254 ± 56
Pond 2		-4.1 ± 1.1	1.0 ± 0.1	94 ± 36	89 ± 36
Pond 1	$\mu g\ m^{-2}$	0.105 ± 0.03	$4.89 \times 10^{-4} \pm 5 \times 10^{-5}$	0.300 ± 0.08	0.404 ± 0.09
Pond 2		-0.002 ± 0.0005	$4.89 \times 10^{-4} \pm 5 \times 10^{-5}$	0.046 ± 0.017	0.043 ± 0.016

Concentrations of MeHg in unfiltered water ranged from 1.19–1.69 ng L^{-1} (n = 6) in Pond 1 and 0.13–0.18 ng L^{-1} (n = 6) in Pond 2, and were comparable to the range of MeHg concentrations reported in surveys of Ellesmere Island wetlands and ponds (Table 5.1; Loseto et al. 2004a, St. Louis et al. 2005). In both ponds, the proportion of THg in the MeHg form peaked in mid-July (July 12–16), reaching a remarkable 47% in Pond 1 and 16% in Pond 2. The storage of MeHg in the water column showed a net accumulation (ΔMeHg) of 0.105 µg m^{-2} in Pond 1, while Pond 2 showed a slight decrease of -0.002 µg m-2 over the three weeks of observation (Table 5.4). Methylmercury concentrations in precipitation ranged from below detection (< 0.015 ng L^{-1}) to 0.13 ng L^{-1}, and the 7.9 mm of rainfall during the study period represented an input of 4.9×10^{-4} µg MeHg m^{-2} (Table 5.4). No other estimates of MeHg inputs from rainfall currently exist for the Canadian Arctic. However, due mostly to low precipitation rates in the High Arctic, the average daily flux of 0.029 ng MeHg m^{-2} d^{-1}, quantified for the Lake Hazen region during July, was significantly lower than the long-term average of 0.11 ng m^{-2} d^{-1} reported for wet deposition in the remote boreal ecoregion of northwestern Ontario (Graydon et al. 2008).

Photodemethylation, or the degradation of MeHg by solar radiation, was quantified using a combination of incubation experiments and modelling. The experimentally determined rate constant of photodemethylation (k_{pd}) in surface waters, normalized to the incident photon flux of PAR, was $3.4 \pm 0.2 \times 10^{-3}$ m^2 E^{-1} (mean ± standard deviation). Additional incubation experiments conducted at different depths throughout the water column at Pond 2 indicated that rates of photodemethylation were driven principally by UV-A radiation. The experimentally determined *kpd* was used in a model based on the results of Lehnherr and St. Louis (2009) which incorporates variables such as aqueous MeHg concentration, incident solar radiation, attenuation of solar radiation with depth in the water column, and bathymetry to calculate whole-pond MeHg photodemethylation. Loss by photodemethylation was greater in Pond 1 (0.3 µg m^{-2}) than in Pond 2 (0.05 µg m^{-2}) because Pond 1 had higher concentrations of MeHg, which led to higher photodemethylation rates. And, because Pond 1 was shallower than Pond 2, on average, the water column was exposed to higher levels of solar radiation.

The in-pond production of MeHg—roughly equivalent to the net diffusion of MeHg to the water column from sediments where Hg(II) is methylated—was calculated as the quantity of MeHg required to balance the MeHg budget and took into account inputs and sinks as follows:

MeHg production = ΔMeHg – inputs (precipitation) + sinks (photodemethylation)

The production of MeHg was an order of magnitude higher in Pond 1 (0.4 µg MeHg m^{-2} or 0.024 µg MeHg m^{-2} d^{-1}) than in Pond 2 (0.04 µg MeHg m^{-2} or 0.0025 µg MeHg m^{-2} d^{-1}). At both sites, in-pond Hg(II) methylation was a much more important source of MeHg than precipitation (Table 5.4). Compared to sediment-water MeHg fluxes measured by Hammerschmidt et al. (2006) in Alaskan tundra lakes (0.0015–0.0045 µg m^{-2} d^{-1}), Pond 1 sediments were a greater source of MeHg while Pond 2 was similar to the Alaskan lakes. Furthermore, MeHg accumulation in the water column of Pond 1 (0.0062 µg m^{-2} d^{-1}) was similar to the net yield of MeHg from known important sites of MeHg production in temperate boreal wetlands (0.0005–0.006 µg m^{-2} d^{-1}) (St. Louis et al. 1996), demonstrating that High Arctic ponds can also be important sites. Methylmercury produced in these shallow ponds will bioaccumulate in local food webs because the sites provide habitat for various zooplankton species and waterfowl (loons, long-tailed ducks, and terns), as well as juvenile char when the ponds become connected to Lake Hazen at the end of summer during the seasonal peak in lake water levels.

5.7 Food webs

5.7.1 Trophic transfer of Hg in coastal Arctic lakes with and without anadromous Arctic char

On the west coast of North America, through a phenomenon termed biotransport, anadromous Pacific salmon transport nutrients and contaminants from the ocean into freshwater environments. Massive salmon deaths after spawning release marine-derived nutrients and contaminants into freshwater ecosystems from decomposing carcasses. These inputs affect aquatic productivity, fish growth, and contaminant loading in spawning grounds used by anadromous salmon (Naiman et al. 2002, Krümmel et al. 2003, Stockner 2003, Gregory-Eaves et al. 2007). Anadromous populations of Arctic char have also been identified as a potential biotransport vector for contaminants in the Canadian Arctic (Blais et al. 2007). Unlike Pacific salmon, anadromous Arctic char may not be very efficient biotransporters because they do not die immediately in massive numbers after spawning. Until recently, however, their impact had not been evaluated.

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The effect of anadromous Arctic char on Hg biomagnification through lake food webs was investigated in six Arctic lakes in the West Kitikmeot region of Nunavut from 2006 to 2008 (Swanson and Kidd 2010). Three lakes contained anadromous populations of Arctic char whereas the other three lakes contained no Arctic char. All lakes contained lake trout as a top predator in addition to a similar suite of forage fishes. Lake trout had THg concentrations ranging from 0.01–1.40 $\mu g\ g^{-1}$ wet weight (ww) (Swanson and Kidd 2010). In an analysis that considered fish size as a covariate, THg concentrations in lake trout were significantly lower in lakes that contained anadromous Arctic char. This difference was not due to reduced biomagnification of Hg because there was no significant variation in the slopes of THg versus nitrogen stable isotope ratios ($\delta^{15}N$; an estimate of trophic position) between lakes with and without anadromous char. Instead, lower THg in lake trout from lakes with anadromous Arctic char resulted from increased lake trout condition. Fish Hg concentrations are often negatively related to fish condition (an index of how heavy a fish is at a given length) (Greenfield et al. 2001, Dittman and Driscoll 2009), and lake trout were in significantly better condition in lakes where anadromous Arctic char were present (Swanson et al. 2011). This effect could be due to greater prey availability for lake trout in lakes with anadromous Arctic char, but further research is required. Overall, it appeared that either the mass of Hg biotransported by anadromous Arctic char to lakes was negligible, or that the effect of increased Hg loading via anadromous char was counteracted by increased fish growth in lakes where anadromous char were present (Swanson and Kidd 2010).

5.7.2 Trophic transfer of Hg in lakes with landlocked Arctic char

Lakes in the Canadian Arctic Archipelago are very unproductive and have simple food chains. Arctic char are often the only fish species present and are thus the apex predator. Pelagic zooplankton communities typically consist of few species at low abundance. The benthic environment of lakes is typically dominated by chironomids, although central and sub-Arctic systems may host a more diverse invertebrate community. Several recent studies have investigated the bioaccumulation of Hg in these lake food webs containing landlocked Arctic char. These studies either included investigations of the entire food web including Arctic char (Gantner et al. 2010a, 2010b), or focussed on Hg uptake in aquatic invertebrates such as zooplankton and chironomids (Chételat et al. 2008, Chételat and Amyot 2009).

A food web study in 18 lakes along a latitudinal gradient from 61–82° N included sampling of Arctic char, pelagic zooplankton, benthic invertebrates and, where present, ninespine stickleback (Gantner et al. 2010b). In addition, sediment and surface water samples were collected. Lake area and watershed area were determined from topographic maps for each site. Biota, sediment, and water samples were analysed for MeHg and THg. Trophic relationships such as food web length and trophic magnification factors (TMFs) for Hg were determined using nitrogen and carbon stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and MeHg concentrations. Benthic invertebrates were typically higher in Hg than pelagic zooplankton. Trophic characterization using $\delta^{15}N$ and $\delta^{13}C$ showed that benthic invertebrates—mainly chironomid larvae and pupae—were the main source of nutrients and thus, also of Hg for landlocked Arctic char. Stomach content analysis in combination with stable isotope results indicated that zooplankton are a less important prey for char (Gantner et al. 2010a, 2010b). Further, cannibalism commonly occurs in landlocked populations of Arctic char and results in higher concentrations of Hg in cannibalistic fish (Muir et al. 2005, Gantner et al. 2010a). Trophic magnification factors, which were estimated by the slope of MeHg concentration versus trophic level for all food web organisms in a lake, did not explain among-lake variation of Hg concentrations in adult Arctic char (Fig. 5.14).

Detailed investigations of the benthic invertebrate communities in lakes and fishless ponds indicated that larvae, pupae, and adult chironomids provide variable amounts of Hg to Arctic char, as MeHg differs among these life stages (Chételat et al. 2008; section 5.7.3). Cannibalism also contributes greatly to variability of Hg (and $\delta^{15}N$ ratios) in char (Hobson and Welch 1995, Guiguer et al. 2002, Gantner et al. 2009b). The relative importance of differential feeding behaviour of chironomid life stages for Hg bioaccumulation in char (Chételat et al. 2008) versus the effects of opportunistic cannibalism remains unknown. Seasonal effects on Hg concentrations and trophic signatures are also possible as, depending on availability, char feed on different chironomid life stages throughout the year. However, since $\delta^{15}N$ ratios and Hg concentrations in muscle tissue reflect long-term dietary consumption, such effects may be integrated in muscle over this period. The winter diet of landlocked char remains unknown in most systems, but could also affect their Hg content and $\delta^{15}N$ ratios.

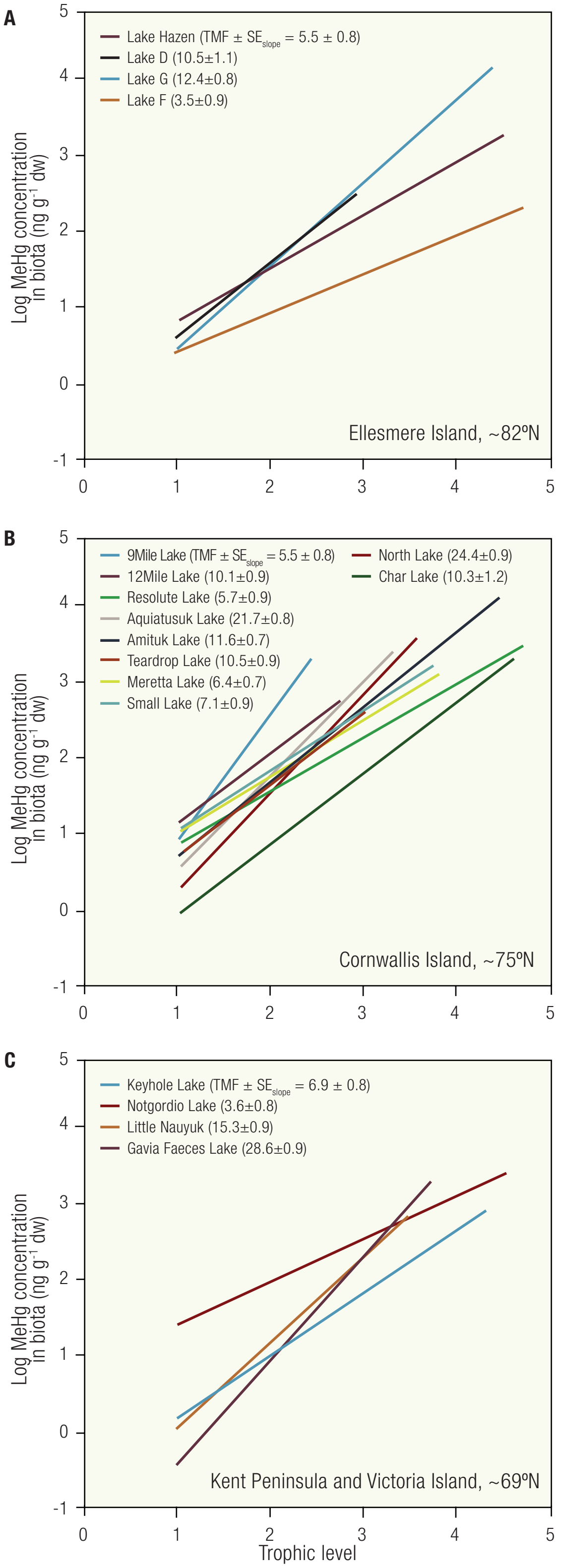

FIGURE 5.14

Trophic magnification factors of MeHg in lake food webs with landlocked Arctic char. Regression lines of the relationship between log MeHg concentration versus trophic level and TMFs (slope ± standard error) are for lake food webs from Ellesmere Island (A), Cornwallis Island (B) and the Kent Peninsula and Victoria Island (C) (Gantner et al. 2010b).

It has been a scientific challenge to differentiate the sources of Hg to Arctic lakes and aquatic biota. Unless Arctic char feed on zooplankton, newly deposited Hg from the atmosphere—which may be available for uptake by plankton in the water column—is less likely to be directly reflected in char Hg concentrations. Food web responses to inputs from the surrounding environment may be delayed until Hg is taken up by chironomids from lake sediments and then by Arctic char from chironomids. Gantner et al. (2010a) reported lower Hg concentrations in Arctic char at more southern, productive sites (approximately 69° N) with more diverse food webs, compared to less productive northern lakes (approximately 75° N). Among-lake differences in Arctic char THg concentrations were not related to THg in surface water and sediment or to latitude and longitude. Catchment-to-lake area ratios were correlated with THg concentrations in landlocked Arctic char, which emphasizes the importance of loadings from the surrounding landscape on Hg cycling in lakes.

5.7.3 Factors affecting Hg bioaccumulation in Arctic invertebrates

Aquatic invertebrates have been recently investigated in several Arctic regions for Hg bioaccumulation (Fig. 5.15, Annex Table A2.3). Order-of-magnitude variation in MeHg concentrations was reported for invertebrate groups, particularly chironomids and zooplankton. Interestingly, maximum concentrations for several invertebrate types—zooplankton, chironomids, fairy shrimp, and mysids—were highest in lakes and ponds on Cornwallis and Devon islands compared to other study areas in the Canadian Arctic. Limited data are also available for *Daphnia*, isopods, and caddisflies (Fig. 5.15). The concentration of Hg in aquatic invertebrates is affected by their trophic level, Hg supply to the food web, taxonomic variation, and developmental effects (Tremblay et al. 1996, Watras et al. 1998, Chételat et al. 2008, Cremona et al. 2008).

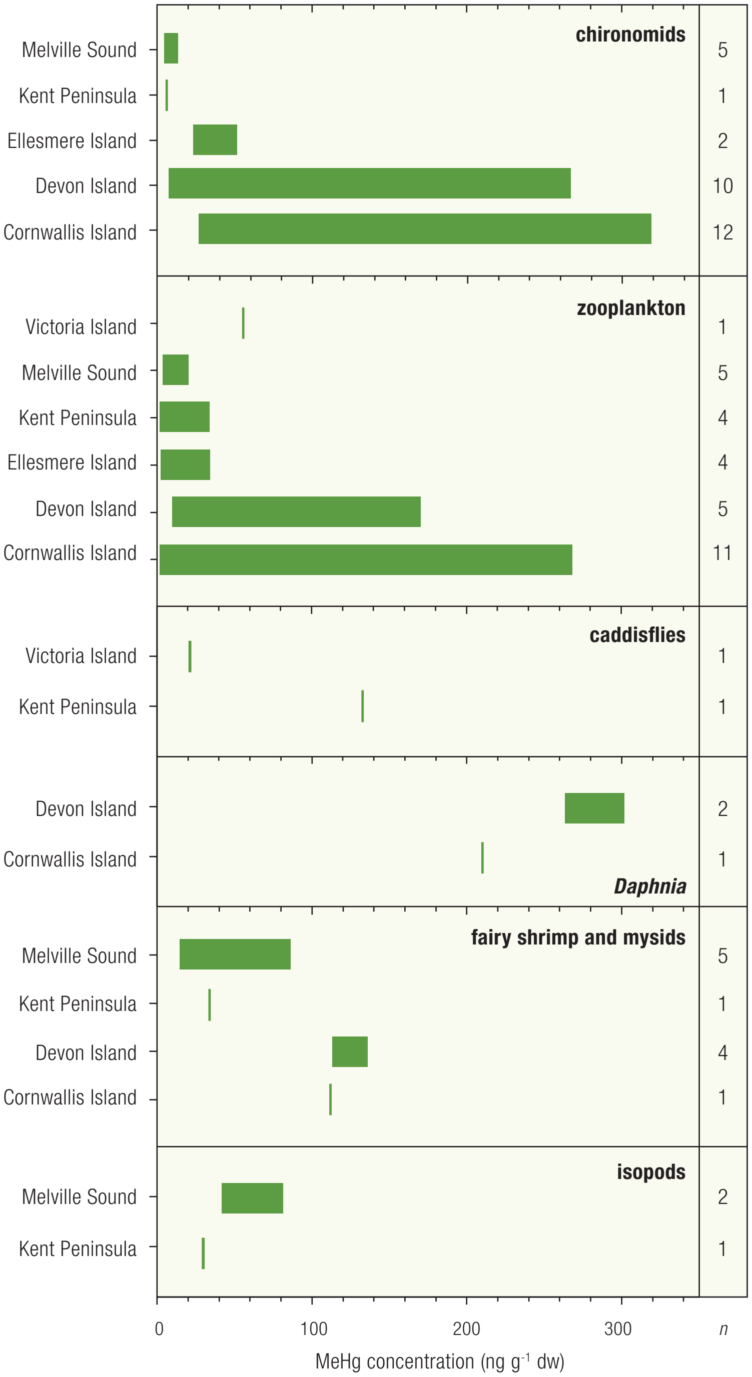

FIGURE 5.15

Ranges of MeHg concentrations in invertebrates from lakes and ponds in different regions of the Canadian Arctic. The number of water bodies (*n*) sampled per region is identified in the right-hand column. Data are from Chételat et al. (2008, 2009), Gantner et al. (2010b) and Swanson and Kidd (2010).

In the Canadian High Arctic, taxonomic composition is an important determinant of MeHg bioaccumulation in zooplankton communities, and different species can vary several-fold in their MeHg concentrations despite low levels of Hg in the water (Chételat and Amyot 2009). In a study of 16 lakes and ponds, zooplankton communities containing *Daphnia* (mainly *D. middendorffiana*) had, on average, five times the MeHg content of copepod-dominated communities. Copepods are common zooplankton in High Arctic lakes whereas *Daphnia* are generally more abundant in ponds. The occurrence of *Daphnia* in Arctic lakes is positively related to the amount of organic carbon in the water column, and climate-driven increases in aquatic productivity during the 21st Century could extend the geographic distribution of this Hg-rich zooplankter (Chételat and Amyot 2009). Further research should focus on how climate-driven shifts in invertebrate community composition affect the transfer of Hg in Arctic lake food webs.

Metamorphosis is an important factor affecting Hg bioaccumulation in chironomids (Chételat et al. 2008), which is the main invertebrate consumed by landlocked Arctic char in many high-latitude lakes. Passing through four developmental stages (eggs, larvae, pupae, adults), these benthic invertebrates undergo complete metamorphosis and have a life cycle of at least two to three years in the Arctic (Welch 1976). A study of 22 ponds and lakes in the Arctic Archipelago showed that MeHg concentrations in chironomids increased during development from larvae to pupae to adults (Chételat et al. 2008). Adult chironomids had 1.7–2.9 times higher MeHg than immature stages. Adults had higher MeHg concentrations than pupae because of a reduction in body weight during emergence and mating and no associated loss in MeHg burden. Larvae were least concentrated with MeHg because they were still growing and had the lowest burdens. The increase in MeHg content during chironomid growth may affect Hg transfer to Arctic char because of differences in diet between juvenile and adult fish. Juvenile char consume primarily larval chironomids which are lower in MeHg. Adult char, on the other hand, consume more pupae and adults (Rigét et al. 1986, Parker and Johnson 1991), which are more concentrated in MeHg.

5.7.4 Tracing the source(s) of Hg in lake food webs using Hg stable isotopes

Investigations of Hg stable isotopes in environmental samples have pursued the question of whether their ratios may be specific to a source (e.g., coal combustion) or a process (e.g., methylation, photochemical reduction), with the intention of using the ratios as an isotopic tracer similar to other elements such as carbon, nitrogen, oxygen, and lead. If a unique source *signature* could be identified, this information would be particularly useful in the Arctic, which has few local pollution sources and receives anthropogenic Hg via atmospheric transport from multiple regions (Durnford et al. 2010). Recent studies have shown that there can be significant variations in Hg isotope composition in environmental samples (Ridley and Stetson 2006, Bergquist and Blum 2009). However, while much progress has been made, a full understanding of Hg isotope fractionation and its application to trace sources and processes has not been achieved.

Mercury's seven stable isotopes (196, 198, 199, 200, 201, 202 and 204 atomic mass units) span a relative mass difference of 4%. Isotopes ^{200}Hg and ^{202}Hg are the most abundant (23.1% and 29.7%). Since about 2000, routine measurements of Hg isotope ratios have become possible using cold vapour multi-collector inductively coupled plasma mass spectrometry (CV–MC–ICP–MS) in which Hg is introduced to the plasma as a gas after chemical reduction and separation using a gas–liquid separator. The multi-collector technology detects all major isotopes (198–202) simultaneously with high precision (standard deviations $< 10^{-4}$ for ^{202}Hg) and at low nanogram sensitivity (Ridley and Stetson 2006).

Mass-dependent fractionation (MDF) of Hg, or fractionation as a function of isotope mass, can occur during processes such as volatilization from water, microbial reduction, and bioaccumulation (Bergquist and Blum 2009). Mass-dependent fractionation is reported in delta notation (δ), which is the per mil (‰) deviation from a reference material and usually reported relative to ^{198}Hg (Blum and Bergquist 2007). A wide range of MDF values have been reported (Fig. 5.16A). Mercury isotopes can also undergo mass-independent fractionation (MIF). Mass-independent fractionation is the difference between the measured δxxxHg ratio and the theoretically predicted δ^{xxx}Hg ratio and denoted by a Δ^{xxx}Hg notation usually relative to the most abundant ^{202}Hg isotope. The range of MIFs in geological media is low but large values (> 0.3‰) have been observed for odd-numbered Hg isotopes during photochemical reduction (Bergquist and Blum 2007), and a wide range of values has been reported in Arctic media (Fig. 5.16B). The processes that cause MIF are not fully understood but are thought to involve slight differences in reactivity of odd and even Hg isotopes due to their nuclear structure (Jackson et al. 2008, Bergquist and Blum 2009). Although there is still debate, most investigators now accept that MIF is mainly induced by photochemical reactions of Hg and MeHg, and that biochemical MIF is unlikely. This implies that Hg MIF variations may provide information about the source of Hg to aquatic food webs.

Sherman et al. (2010) found that Hg deposited on the surface of Arctic snow at Barrow (Alaska) underwent MIF from sunlight-induced reactions during AMDEs, wherein the odd-mass number isotopes of Hg were preferentially reduced and emitted. This depletion resulted in negative values of Δ^{201}Hg (Fig. 5.16B). This distinctive MIF for Hg in snow was also found in Arctic lake sediments that had similar negative MIF values (Fig. 5.16B; Gantner et al. 2009a). As of 2010, the values of Δ^{201}Hg reported by Gantner et al. (2009a) are the most negative MIFs ever reported for lake sediments. Jackson et al. (2004) also noted more negative MDF values of δ^{202}Hg in more recent horizons of a core from Romulus Lake near Eureka on Ellesmere Island—see Ridley and Stetson (2006) for replotting of Jackson's original data. Jackson et al. (2008) found similar negative δ^{202}Hg values in Cli Lake in the Northwest Territories and Shipiskan Lake in Labrador. It is not known whether this negative Hg signature represents a distinct long-range transport signal or is due to isotopic fractionation related to biological activity within the lake such as Hg methylation in sediments (Ridley and Stetson 2006).

The negative Δ^{201}Hg isotope signature in lake sediments also appeared in benthic chironomids from the same lakes (Gantner et al. 2009a) thereby suggesting transfer of this signature to sediment-dwelling organisms. However, this negative MIF source was not evident in pelagic organisms in Arctic lakes. Zooplankton and Arctic char samples had much different values of Δ^{201}Hg compared to chironomids. Results for Arctic char in Hazen and Resolute lakes ranged from -0.02 to 1.7‰ for Δ^{201}Hg (Fig. 5.16B) and char from most other lakes analysed by Gantner et al. (2009a) were also within this range. These results are in agreement with those for lake trout from sub-Arctic lakes (Cli and Shipiskan), which showed large positive Δ^{201}Hg values (Jackson et al. 2008). However char from Amituk Lake differed by having lower Δ^{201}Hg values (-0.11–0.16‰) while Pingualuk char had the highest values (3.67–3.89‰). Pingualuk Lake serves as an interesting site to investigate atmospheric inputs of Hg because it has no inflow, no outflow, minimal catchment area, and it receives its water mainly through direct precipitation. Although no zooplankton were analysed for Hg isotopes in Pingualuk Lake, the large positive Δ^{201}Hg is consistent with char feeding on zooplankton. The low Δ^{201}Hg for char in Amituk Lake may be due to the large catchment area of Amituk which could contribute to an isotope signature resembling that of snow Hg which has undergone a greater degree of photochemical transformation.

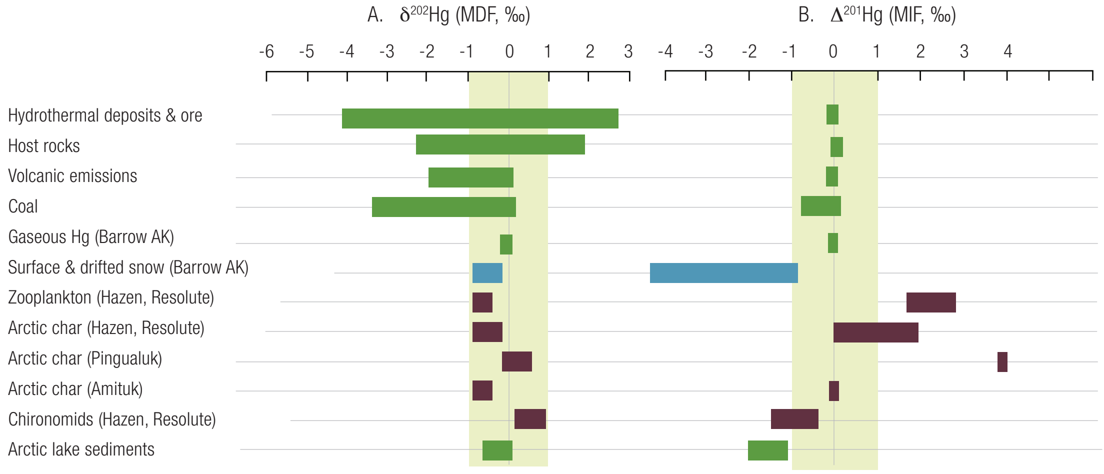

FIGURE 5.16

Mercury isotope results for selected geological sources and for samples from the Arctic. The horizontal bars represent the range in (A) mass-dependent fractionation (MDF) expressed as δ^{202}Hg, and (B) mass-independent fractionation (MIF) expressed as Δ^{201}Hg. The gray vertical bars represent estimates of crust- and mantle-derived Hg isotope ratios. Geological data are from Bergquist and Blum (2009), results for snow and gaseous Hg are from Sherman et al. (2010), and biota and sediments are from Gantner et al. (2009a).

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A study of Hg isotope ratios in eggs of thick-billed murres collected from colonies in the North Pacific Ocean, the Bering Sea, and the Chukchi Sea found that Δ^{201}Hg declined with latitude from about 1.0 to 0.5‰ (Point et al. 2011). Similar to other Arctic biota, this Hg MIF signature did not resemble the highly negative Hg MIF in snow from Barrow, Alaska (Sherman et al. 2010). The Δ^{201}Hg in murre eggs was negatively correlated with sea-ice cover but not with the trophic level of the birds. Therefore, the authors concluded that the effect of sea-ice cover was to impede the photochemical breakdown of MeHg in surface waters, which implies that greater open water will result in more MeHg transformation (Point et al. 2011). Gantner et al. (2009a) found a negative correlation of δ^{202}Hg in Arctic char with latitude, which may be related to degree of MDF of Hg during atmospheric transport to the Arctic from various source regions or because of differences in terrestrial Hg sources. Those authors did not observe a latitudinal gradient in Δ^{201}Hg, but found it was related to the size of the lake catchment.

5.8 Freshwater fish

5.8.1 Geographic and species variation in fish Hg concentrations

Broad-scale patterns of Hg concentrations in freshwater fish species common to the Canadian Arctic were examined using the National Fish Mercury Database (Depew 2011), an initiative of the Canadian Mercury Science Assessment (section 1.6). Fish THg concentrations for a total of 85 Arctic sites were obtained from NCP and other government monitoring programs as well as private-sector environmental monitoring reports (see Annex Table A2.2 for data and data sources). Data for six species of fish—lake trout, lake-dwelling Arctic char, northern pike, burbot, lake whitefish, and round whitefish—were available from multiple lakes and rivers sampled between 2002 and 2009. Older data on Hg in Arctic fish are summarized elsewhere (Fisk et al. 2003, Lockhart et al. 2005). Fish muscle concentrations of THg were length-adjusted using the mean length of all individuals for each species and sorted into three concentration categories: < 0.2 µg g^{-1} ww, 0.2–0.5 µg g^{-1} ww, and > 0.5 µg g^{-1} ww. The Health Canada consumption guideline for commercial sale of fish is 0.5 µg g^{-1} ww (Health Canada 2011). Lake trout was the most widely sampled species (46 sites), followed by whitefish (round or lake whitefish in 32 sites), Arctic char (25 sites), northern pike (9 sites) and burbot (7 sites).

Average length-adjusted THg concentrations in lake trout were mainly between 0.2–0.5 µg g^{-1} (52% of sites) or less than 0.2 µg g^{-1} (39%) (Fig. 5.17). The only lakes that contained lake trout with mean length-adjusted THg concentrations greater than 0.5 µg g^{-1} were Cli and Kelly lakes in the Mackenzie River basin, Fox Lake in Yukon, and Kodiak Lake, a monitored site downstream of the Ekati diamond mine in the central Arctic. Average THg concentrations in lake-dwelling Arctic char were predominately less than 0.2 µg g^{-1} (68%) or between 0.2–0.5 µg g^{-1} (28%). The only site where Arctic char had average THg greater than 0.5 µg g^{-1} was Amituk Lake on Cornwallis Island. At the majority of sites (91%), whitefish had average THg concentrations of less than 0.2 µg g^{-1} (Fig. 5.18). Narrow Lake, northeast of Yellowknife, was one site where lake whitefish had THg levels above 0.5 µg g^{-1}. Few data were available for northern pike or burbot, but average Hg concentrations for these species were less than 0.5 µg g^{-1} at most sites (Fig. 5.18).

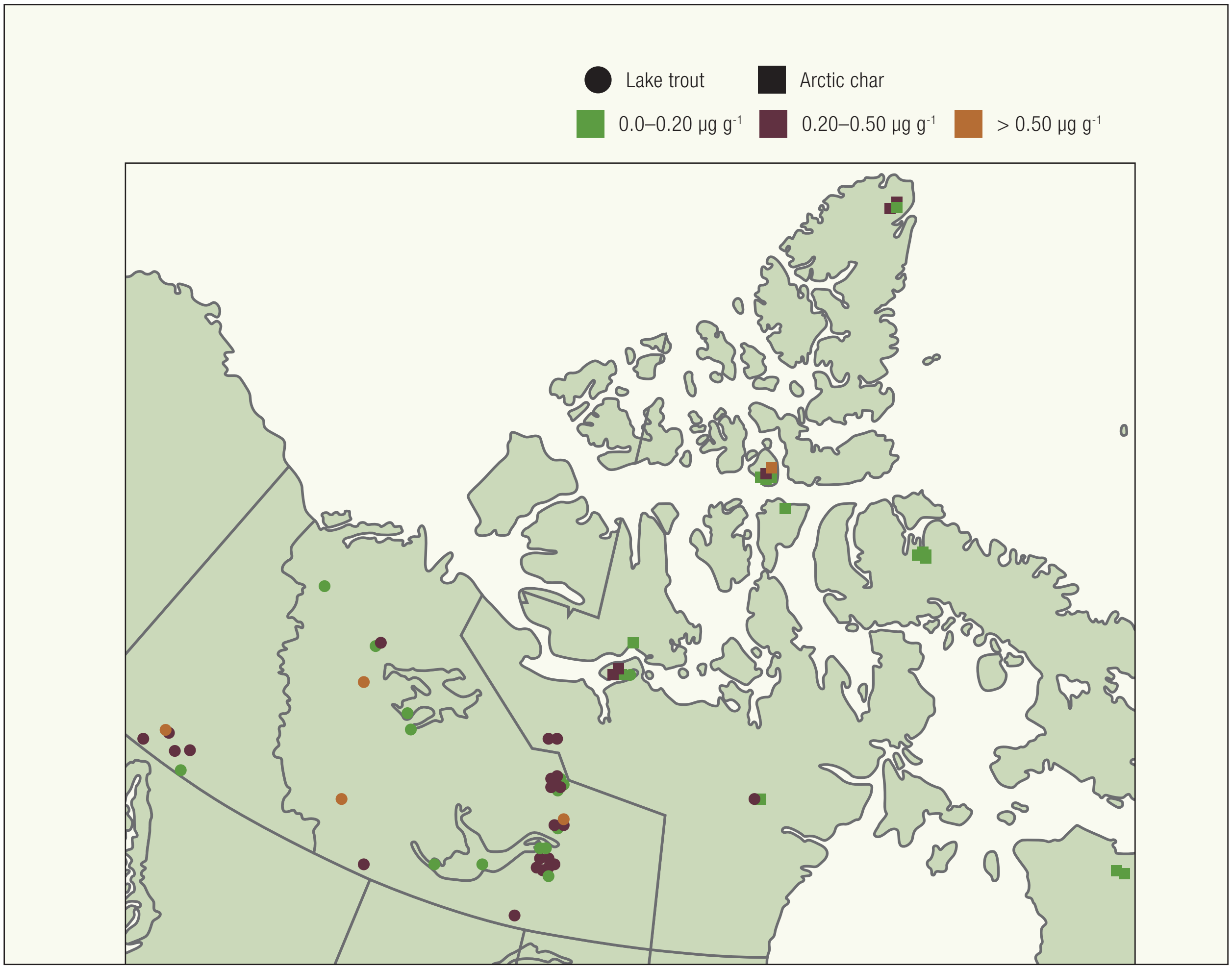

FIGURE 5.17

Length-adjusted THg concentrations in lake trout and Arctic char muscle measured between 2002 and 2009 in freshwater ecosystems in the Canadian Arctic (Depew 2011). Site-average concentrations are sorted into three categories with the highest concentration category (> 0.5 µg g^{-1} ww) representing the Health Canada consumption guideline for commercial sale of fish (see Annex Table A2.2 for a list of the data and sources). Concentrations were standardized to lengths of 544 mm and 377 mm for lake trout and Arctic char, respectively.

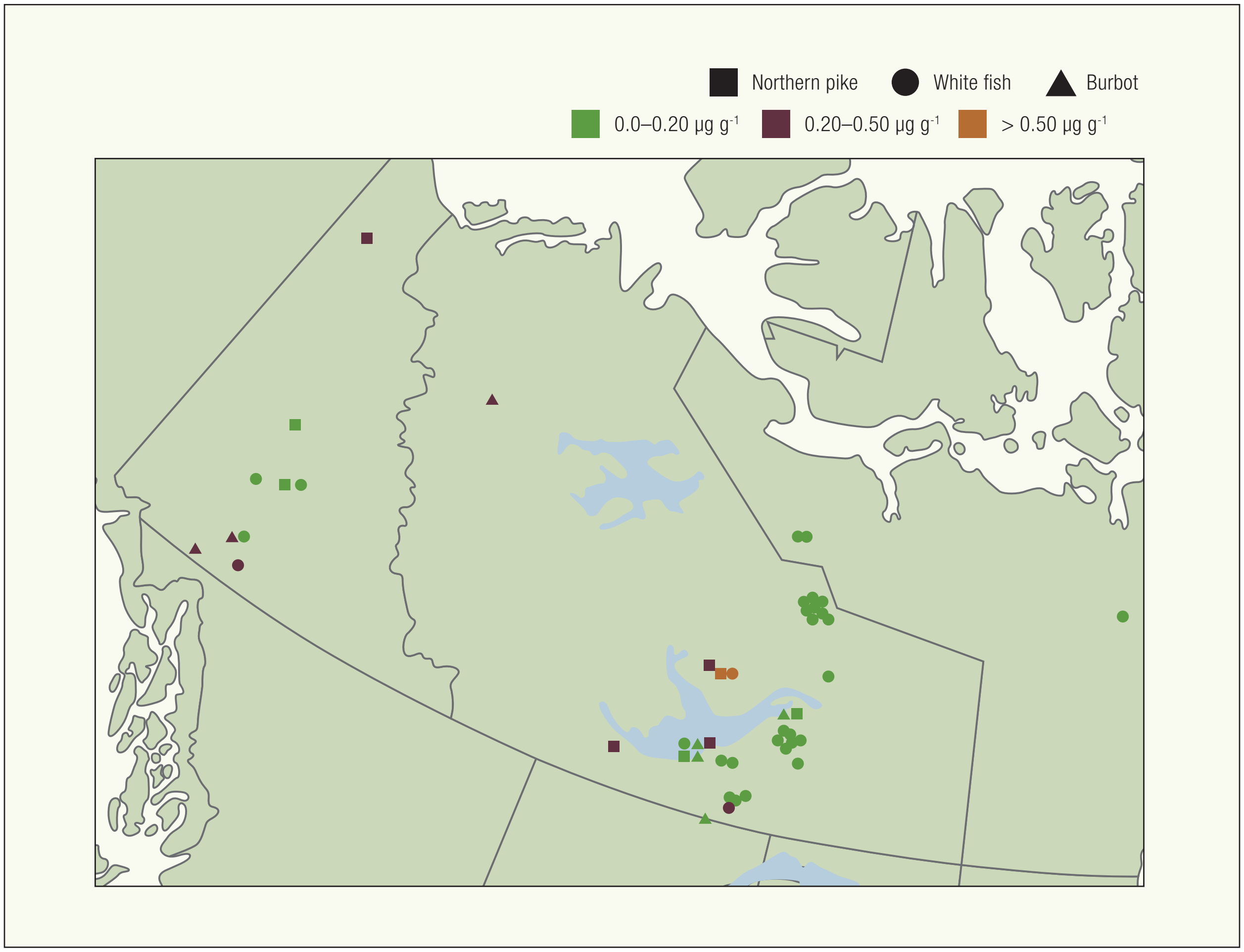

FIGURE 5.18

Length-adjusted THg concentrations in northern pike, burbot and lake or round whitefish measured between 2002 and 2009 in freshwater ecosystems in the Canadian Arctic (Depew 2011). Site-average concentrations are sorted into three categories with the highest concentration category (> 0.5 µg g^{-1} ww) representing the Health Canada consumption guideline for commercial sale of fish (see Annex Table A2.2 for a list of the data and sources). Concentrations were standardized to a length of 599 mm for pike, 595 mm for burbot, 327 mm for round whitefish, and 395 mm for lake whitefish.

No geographic patterns or regional hotspots for Hg were evident for freshwater fish in the 2002 to 2009 dataset. This finding is consistent with Lockhart et al. (2005) who also found no strong geographic patterns in the Canadian Arctic using an older, larger dataset of fish collected over 30 years since 1971. Intensive lake surveys in the Mackenzie River basin and the High Arctic suggest that differences in fish Hg concentrations among lakes are related to lake-specific characteristics such as lake and watershed size or DOC concentration that influence local transport and/or methylation of Hg (Evans et al. 2005, Gantner et al. 2010a). Although the majority of water bodies in the 2002 to 2009 dataset were undisturbed, a few monitoring sites were also included from the impounded Talston River south of Great Slave Lake as well as waters adjacent to mines where human activities could have affected fish Hg concentrations.

Regional differences in sampling effort were evident for Hg measurements on Arctic fish. Over the last decade, most work has been conducted in the Northwest Territories and Yukon, and few recent data exist for lake trout or lake-dwelling Arctic char within their distribution ranges in the Kivalliq region (south-central Nunavut) and Nunavik (Fig 5.17). Considerable progress has been made in measuring Hg levels in lake-dwelling char from several areas in the High Arctic, although larger islands such as Baffin and Victoria appear to be under-sampled. Likewise, other predatory fish species that can attain high Hg concentrations such as burbot, northern pike, and walleye received little attention within their ranges in the Northwest Territories and Yukon. Although the National Fish Mercury Database does not contain all existing data for Arctic fish, it is nevertheless considered sufficiently comprehensive to evaluate regional sampling effort. Given recent increases in fish Hg concentrations for some sites in the Northwest Territories (sections 5.8.3 and 5.8.4), renewed investigation of fish Hg bioaccumulation in the central and eastern Arctic is warranted.

5.8.2 Arctic char

Comparison of Hg bioaccumulation in sea-run and lake-dwelling Arctic char

Previous investigations have noted that sea-run Arctic char have much less Hg than lake-dwelling populations (Muir et al. 1997, Lockhart et al. 2005). It is unclear why Hg levels are so much lower given that sea-run char spend only a few weeks feeding in the ocean, generally on an alternate year basis. An ongoing project is currently examining this question by comparing THg concentrations in sea-run and lake-dwelling populations of Arctic char along a latitudinal gradient in the eastern Arctic running from Lake Heintzelman on Ellesmere Island to Nain in Labrador (Table 5.5). The study includes comparisons of Hg levels in char as well as in marine and lake food webs.

Preliminary results confirm that lake-dwelling char are distinctly smaller than sea-run char and have substantially higher THg concentrations (Fig. 5.19). Similar differences in fish size and THg concentrations were observed between sea-run and landlocked Arctic char in the West Kitikmeot region (Swanson et al. 2011). In that study, the authors found that sea-run and resident (with access to the sea, but not migrating) Arctic char had significantly lower THg (0.04 µg g^{-1} ww) than landlocked char (0.19 µg g^{-1} ww) at a standardized fork length of 500 mm. At all latitudes in the eastern Arctic, THg levels were higher in lake-dwelling than in sea-run char, and latitudinal gradients were not obvious (Fig. 5.20). Similarly, Gantner et al. (2010a) found no evidence of a latitudinal gradient in THg in landlocked char once concentrations were adjusted for length.

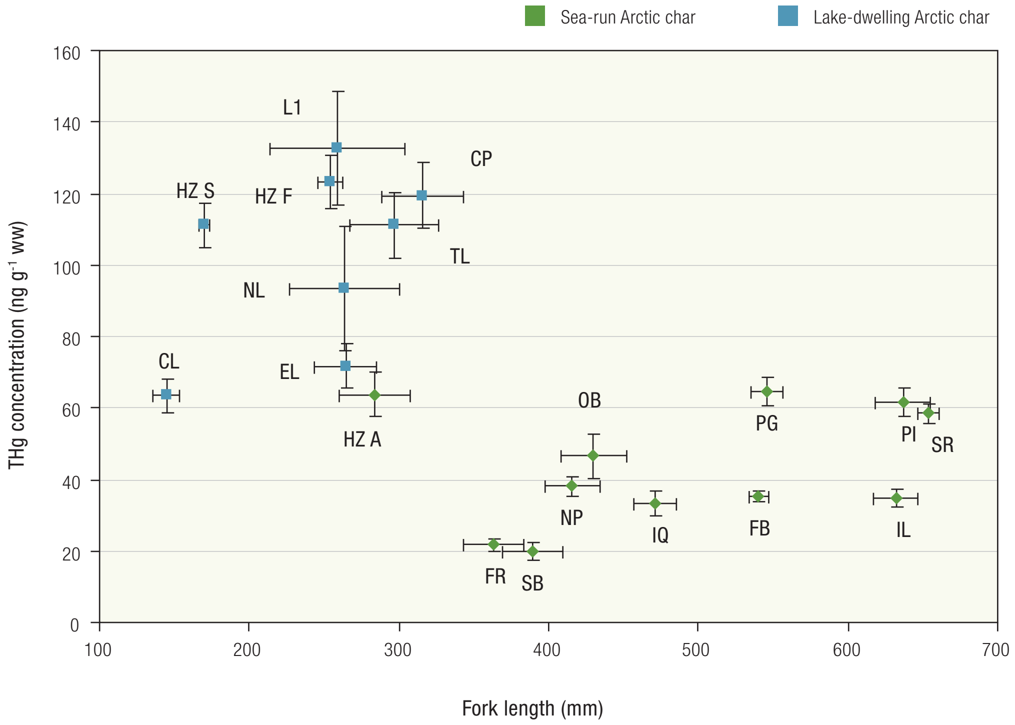

FIGURE 5.19

Relationship between mean (± standard error) fork length and THg concentration in sea-run and non-piscivorous, lake-dwelling populations of Arctic char from the eastern Canadian Arctic (Dorn and Power 2010). Labels identify the site as defined in Table 5.5.

TABLE 5.5 Sampling sites in the eastern Canadian Arctic where sea-run and lake-dwelling Arctic char were captured (Dorn and Power 2010). Figure ID identifies the site labels used in Fig. 5.19 and 5.20. Type S = sea-run Arctic char, type L = non-piscivorous lake-dwelling Arctic char.

Sample Site	Figure ID	Location	Latitude	Type	*n*
Heintzelman Lake	HZ A	Ellesmere Is.	81°42' N	S	7
Fraser River, Nain Bay	FR	Labrador	56°38' N	S	20
North Arm, Saglek	SB	Labrador	58°33' N	S	22
Okak Bay	OB	Labrador	57°33' N	S	20
Iqalugaarjuit Lake	IL	Baffin Is.	66°34' N	S	20
Pangnirtung Fiord	PG	Baffin Is.	66°08' N	S	20
Iqaluit, bay	IQ	Baffin Is.	63°45' N	S	10
Salmon River mouth	PI	Baffin Is.	72°41' N	S	40
Nepihjee River	NP	Ungava Bay	58°32' N	S	33
Frobisher Bay	FB	Baffin Is.	63°41' N	S	20
Salmon River	SR	Baffin Is.	72°41' N	S	30
Heintzelman Lake (fast-growing)	HZ F	Ellesmere Is.	81°42' N	L	56
Heintzelman Lake (slow-growing)	HZ S	Ellesmere Is.	81°42' N	L	56
Coady's Pond 2	CP	Labrador	56°39' N	L	20
Upper Nakvak Lake	NL	Labrador	58°40' N	L	18
Esker Lake	EL	Labrador	57°09' N	L	19
Tasiapik Lake	TL	Ungava Bay	58°31' N	L	31
Crazy Lake	CL	Baffin Is.	63°52' N	L	40
Unnamed lake 1	L1	Baffin Is.	72°36' N	L	16

In the eastern Canadian Arctic, differences in THg concentrations between sea-run and lake-dwelling char may be related to variation between populations in growth rates, trophic position, or prey Hg content. Preliminary evidence from two study locations at Nain (Labrador) suggest that growth rates and trophic position cannot explain lower THg levels in sea-run char of the Fraser River compared to a nearby lake-dwelling population in Coady's Pond (Dorn et al. 2010). A comparison of Hg levels in the prey of these two populations is currently in progress. Consistent with the results from Labrador, Swanson et al. (2011) concluded that trophic position could not explain differences in THg concentrations among life history types of Arctic char in the West Kitikmeot. In contrast though, fish age and carbon to nitrogen ratios—an estimate of lipid content—were statistically related to higher THg concentrations in landlocked relative to sea-run char. Those findings suggest that slower growth rates and poorer body condition in landlocked char resulted in greater Hg bioaccumulation in the West Kitikmeot.

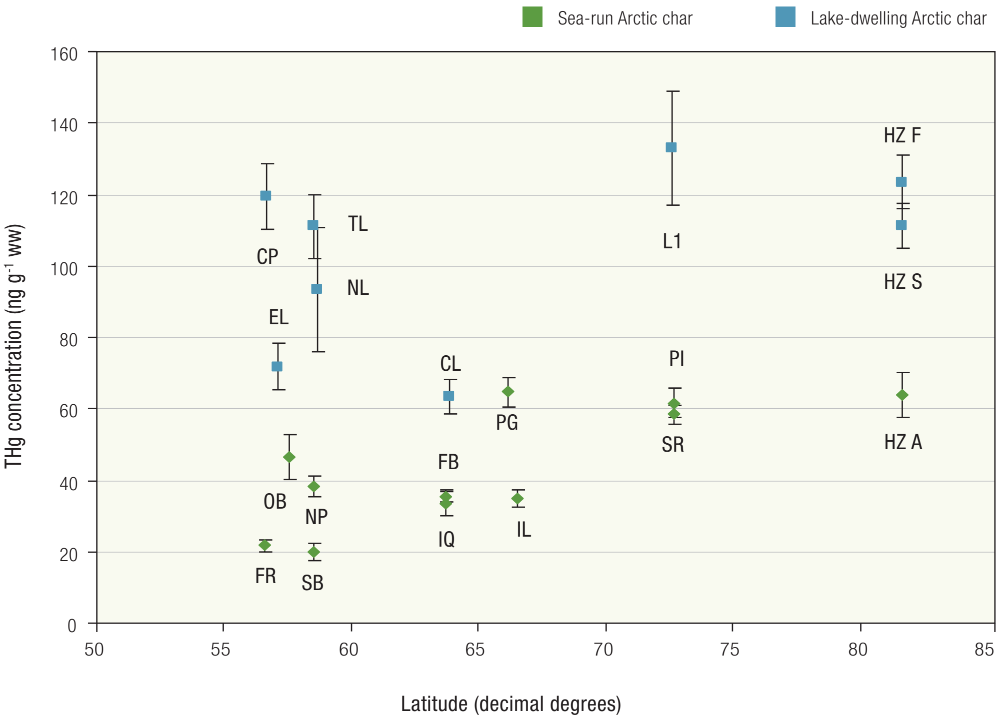

FIGURE 5.20

Relationship between mean (± standard error) THg concentration and latitude in sea-run and non-piscivorous, lake-dwelling populations of Arctic char from the eastern Canadian Arctic (Dorn and Power 2010). Labels identify the site as defined in Table 5.5.

Temporal trends of Hg in landlocked char

As the only top predator in High Arctic lakes, landlocked Arctic char are considered good indicators of Hg inputs (Köck et al. 2004). Temporal trends of THg concentrations in Arctic char are ultimately driven by inputs from geogenic and atmospheric sources, including long-range atmospheric transport and deposition to the lake surface and catchment (Fitzgerald et al. 2005, Muir et al. 2009b). Local catchments are typically the primary source of water for Arctic lakes (Wrona et al. 2005). An intense three-week runoff period in spring is also the main pathway for Hg delivery to these ecosystems (Semkin et al. 2005). The sedimentary records for Hg in 18 High Arctic lakes showed overall increasing trends since the 1950s and recent anthropogenic fluxes are in agreement with modelled estimates (Muir et al. 2009b; section 5.4).

During NCP III, annual sampling of landlocked Arctic char was conducted at Resolute, Char, and Amituk lakes on Cornwallis Island, as well as at Lake Hazen in Quttinirpaaq National Park on Ellesmere Island. For landlocked char, annual collections have been successfully carried out in Resolute Lake since 1997 (Köck et al. 2004, Muir et al. 2005). However, fish collection is more difficult in Char and Amituk lakes due to a low population size in Char and weather dependent access by helicopter to Amituk. Char collection in Lake Hazen is also a challenge mainly due to the high cost of flights to the lake. As a result, collections from Char, Amituk, and Hazen have not been made consistently on an annual basis. Collection numbers have ranged from 7–25 adult fish (> 200 g) per lake except in Char Lake where the range has been 3–10 fish annually.

BOX 5.1 Community involvement in NCP environmental monitoring on Cornwallis Island

The small Hamlet of Resolute Bay on Cornwallis Island is the second most northern community in Canada and home to about 250 residents. Since the beginning of the NCP in the early 1990s, the Iqaluk family in Resolute has participated in scientific studies of lakes. Debbie Iqaluk, her son Pilipoosie (Harry), and other members of the family have been, and continue to be, actively involved in the freshwater monitoring of trends in contaminant bioaccumulation in Arctic char as well as investigations on contaminant cycling.

Debbie Iqaluk is an avid fisher and very good at catching lake-dwelling Arctic char, which can be difficult to capture in sufficient numbers for the long-term monitoring program. She and her family have also taught southern students and researchers how to travel on the rugged land by ATV, move on lake ice safely, and be aware of other dangers such as polar bears. Their land skills facilitate access to remote lakes that otherwise might not be sampled. Debbie's contributions to the monitoring programs have earned her several co-authorships on peer-reviewed publications. Pilipoosie has attended two scientific conferences in southern Canada to learn more about how the work he is involved in is put into a broader scientific context. He was also able to meet other scientists and Northerners when attending the meetings, which allowed him to make additional contacts in the Arctic research community.

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The Iqaluks are not the only family in Resolute Bay that works with scientists from different research programs and institutions. Nancy Amarualik, Peter Amarualik, and the local Hunters and Trappers Organization in Resolute have facilitated the collection of marine mammal tissue (mainly from harvested seals) by local hunters for NCP studies. The invaluable contributions of Resolute Bay community members to the NCP are an excellent example of successful partnerships between local stakeholders and scientists in a small Arctic community.

Time trends of THg in char muscle were analysed with PIA software (Bignert 2007) using length-adjusted log-transformed geometric means for each sampling year (Table 5.6). No statistically significant trends were found in any of the four lakes. Among the four lakes, the dataset for Resolute Lake, with 14 sampling years, had the highest power (65%) to detect a log-linear trend of 5% change in THg concentration (Table 5.6). It was also the closest to achieving the desired goal of detecting a 5% change with a power of 80% (at α = 5%) with 16 years of monitoring. The three-year running average, as shown in Fig. 5.21, suggests that concentrations have been declining in all four lakes since about 2005. However, as of 2010, the number of sampling years is insufficient to determine whether this decline is statistically significant.

For some contaminants, changes in inputs are reflected relatively quickly in Arctic char. For example, the use of man-made flame retardants (polybrominated diphenyl ethers) increased in consumer products by 200–300% in North America and Europe in the 1990s (Alcock et al. 2003), and, subsequently, concentrations of polybrominated diphenyl ethers increased by 59% in char muscle from Lake Hazen over the period from 2001 to 2008 (Muir et al. 2009a). However, atmospheric emissions of Hg are changing relatively slowly. That is, emissions are estimated to have increased by close to 50% in Asia from 1990 to 2005 while declining in North America and Europe (section 3.6.5; AMAP/UNEP 2008, Pacyna et al. 2010). Small changes in Hg inputs could be obscured by year-to-year variation in biological and environmental factors influencing the availability of Hg.

TABLE 5.6 PIA analysis of time series for THg concentrations in landlocked char muscle (Muir 2011).

Lake	Sampling Period	Number of Years in Dataset	Overall Trend	Years Required to Detect a 5% Change[a]	Current Power to Detect a Log-Linear trend of 5%[b]	Lowest Detectable Change for a 10-Year Period[c]
Amituk	1989–2009	10	-0.70% (NS)	19	17%	14%
Char	1993–2010	9	0.6% (NS)	13	34%	7.6%
Hazen	1990–2010	10	1.2% (NS)	23	11%	20%
Resolute	1993–2010	14	-2.2% (NS)	16	65%	11%

NS = not statistically significant, $p > 0.20$.

[a] With a power of 80% at $\alpha = 5\%$.

[b] With the number of years in the current time series.

[c] With the current between-year variation at a power of 80%.

The temporal trends in the Canadian Arctic contrast with changes in THg observed in a single population of landlocked char from a small lake in southwest Greenland (Rigét et al. 2010) where length-adjusted concentrations increased significantly over the period from 1994 to 2008. This trend was tentatively linked to increasing mean monthly air temperatures during May to August although correlations were not statistically significant due to the small number of sampling years (n = 6).

5.8.3 Lake trout

Anadromous lake trout in the West Kitikmeot

Although lake trout are generally thought to be a wholly freshwater fish species, recent research in the West Kitikmeot region has shown that there are actually three life history types: 1) landlocked (no access to the sea, wholly freshwater); 2) resident (access to the sea, exist in sympatry with anadromous individuals but wholly freshwater); and, 3) anadromous (Swanson et al. 2010). The geographic extent of anadromous lake trout is not currently known. However, it is possible that anadromous populations exist across the entire mainland Arctic coast (Swanson et al. 2010).

Life history was found to have a significant effect on Hg bioaccumulation in lake trout from the West Kitikmeot (Swanson and Kidd 2010, Swanson et al. 2011). In four coastal lakes where both anadromous and resident lake trout were present, anadromous fish had significantly lower THg concentrations (0.19 μg g^{-1} ww) than resident fish (0.31 μg g^{-1} ww) at a standardized fork length of 650 mm (Swanson and Kidd 2010). Landlocked trout in two nearby inland lakes had an average, length-adjusted THg concentration similar to the coastal, resident trout (Swanson et al. 2010). Differences among life history types appear to be related to muscle lipid content, which can result from varying energy costs for foraging or diet type and abundance (Swanson et al. 2010). Other studies have reported less Hg in fish with higher lipid content (Wiener et al. 2003), which may reflect growth dilution or a dilution effect by the lipid.

Lake trout in the Mackenzie River basin

Concentrations of THg in lake trout of the Northwest Territories have been summarized by Lockhart et al. (2005). Lake trout can have THg concentrations that exceed Health Canada's 0.5 μg g^{-1} ww guideline for commercial sale of fish (Lockhart et al. 2005). The study by Lockhart et al. (2005) was based on the synthesis of many years of fish inspection records with more recent measurements made during stock assessments in lakes along the Mackenzie River (Stewart et al. 2003a, 2003b, Lockhart et al. 2005). Spatial patterns in THg concentrations of trout from lakes along the Mackenzie River were related to lake and watershed size—higher THg concentrations were more commonly associated with smaller lakes having larger watersheds (Evans et al. 2005). Fish age was also important in affecting high THg concentrations, with trout beginning to exceed the 0.5 μg g^{-1} guideline once they reached the age of about eight to ten years (Evans et al. 2005).

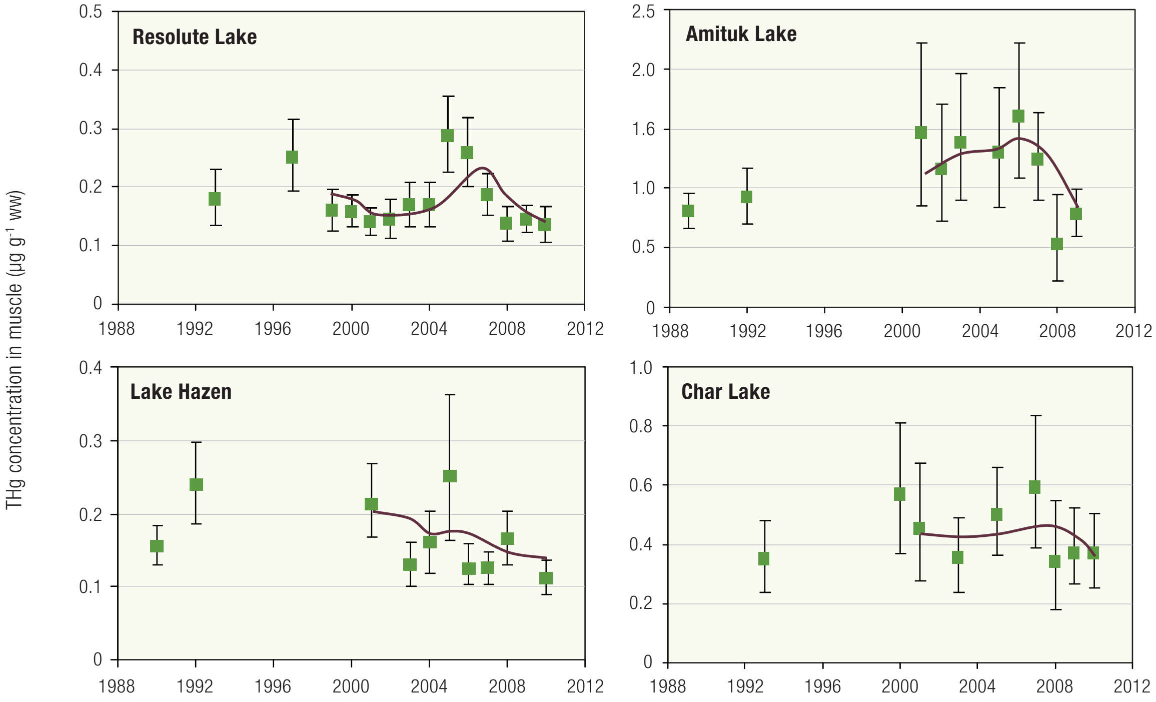

FIGURE 5.21

Temporal trends of length-adjusted THg concentrations (geometric means ± 95% confidence intervals) in muscle of landlocked char from Amituk, Char, Hazen, and Resolute lakes (Muir 2011). The smoothing line represents a three-year running average.

Since 2005, contaminant trend monitoring of lake trout under the NCP has focused on Great Slave Lake in the East Arm near Lutsel K'e and the West Basin near Hay River (see section 5.8.5). In addition, from 2004 to 2009, lake trout were investigated in two lakes per year in a series of lakes along the Mackenzie River for which historic records of their Hg levels exist (Evans et al. 2005, Lockhart et al. 2005, Evans and Muir 2010b). The purpose of these periodic assessments was to investigate temporal Hg trends in these smaller lakes.

Periodic assessments conducted in lakes of the Mackenzie River basin revealed that THg concentrations in lake trout follow the same general patterns reported in Evans et al. (2005). Highest concentrations were observed in Cli and Kelly lakes, and Lac Ste. Therese, the three smallest lakes with surface areas of 43, 116, and 127 km^2, respectively (Fig. 5.22). Concentrations were also high in Stark Lake with a surface area of 177 km^2. Mean concentrations in other lakes were below 0.5 µg g^{-1} of THg with particularly low levels in the West Basin and East Arm of Great Slave Lake.

Slightly higher THg concentrations in Great Bear Lake were associated with the older ages of trout in this lake (Evans et al. 2005). These studies demonstrate that lake trout inhabiting smaller lake ecosystems are more likely to have high Hg levels than fish living in large lake ecosystems. This difference is thought to be due to a combination of higher temperatures, lake productivity, and wetland and littoral zone influences on Hg methylation rates in small rather than large lake ecosystems (Evans et al. 2005). Furthermore, small lakes have less dilution capacity than large lakes.

Temporal variation of Hg in lake trout collected during periodic lake assessments

The investigation of THg temporal trends in lake trout from lakes along the Mackenzie River is limited by the fact that fish were sampled only periodically. Nevertheless, trends of THg increase were observed, particularly in the smaller lakes. Colville Lake (surface area 448 km^2) was investigated in 1999, 2005, and 2009 with length-adjusted THg concentrations

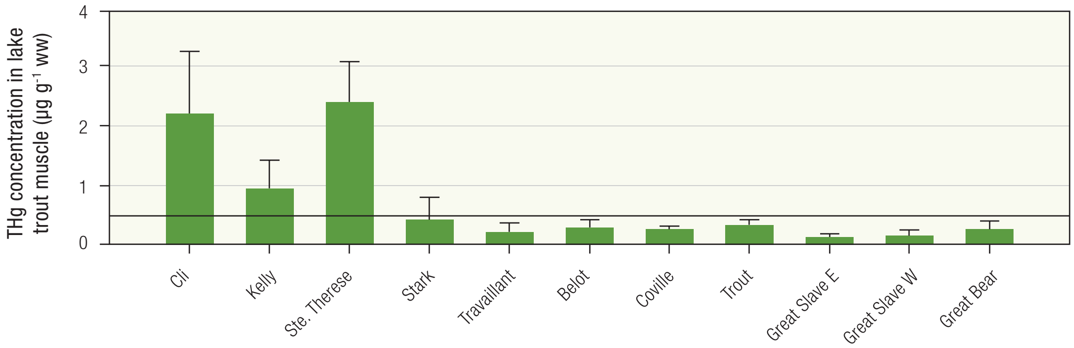

FIGURE 5.22

Mean THg concentrations (± 1 standard deviation) in lake trout muscle from various lakes in the Mackenzie River basin (Evans and Muir 2011). The black line is the Health Canada consumption guideline for commercial sale of fish (0.5 µg Hg g^{-1} ww).

showing only a slight increase (Fig. 5.23). Similarly, Lac Belot (surface area 304 km^2) was investigated in 1999 and 2005 with a slight increase observed between the two sampling years (Evans and Muir 2010b).

In the Norman Wells area, Kelly Lake was investigated in 1998 and 2007, and THg levels in lake trout were significantly higher in 2007 (Fig. 5.23). In Lac Ste. Therese, THg concentrations more than doubled between 2003 and 2008. In contrast, lake trout sampled from Great Bear Lake (31,328 km^2) in 1979, 2001, and 2007, showed only a small but significant increase in THg concentrations. In this same area, Carrie et al. (2010) reported increases in THg concentrations in burbot caught from the Mackenzie River at Fort Good Hope and related these increases to a long-term warming trend (section 5.8.4).

Further south, in the Fort Simpson area, Cli Lake was sampled in 1996, 2006, and 2009. Concentrations of THg in lake trout did not differ significantly between years although the levels were consistently very high (Fig. 5.23) probably because the fish were old (commonly 20 years and over at 600 mm). Trout Lake (507 km^2), south of Fort Simpson, is believed to only have limited deep water habitat to support its lake trout population. Concentrations in lake trout increased significantly between 1991, 2003, and 2008, exceeding 0.5 µg g^{-1} of THg in 2008.

Based on these lake surveys, Hg advisories were issued for Cli, Trout, and Kelly lakes, and Lac Ste. Therese in the summer of 2010. Given that strong increases were associated with smaller and/or shallower lakes, it is probable that other small- to medium-size lakes in

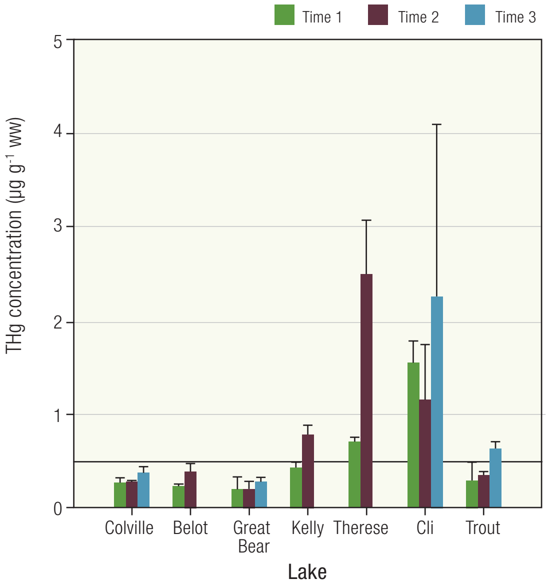

FIGURE 5.23

Temporal changes in length-adjusted THg concentrations in lake trout muscle collected from lakes along the Mackenzie River. The sampling years for each lake are identified in the text. Length adjustments are based on fish of 555 mm (Trout), 600 mm (Kelly, Lac Ste. Therese) or 650 mm (Colville, Great Bear, Cli), depending on the size range of fish caught in each lake (Evans and Muir 2010b). The black line is the Health Canada consumption guideline for commercial sale of fish (0.5 µg Hg g^{-1} ww)

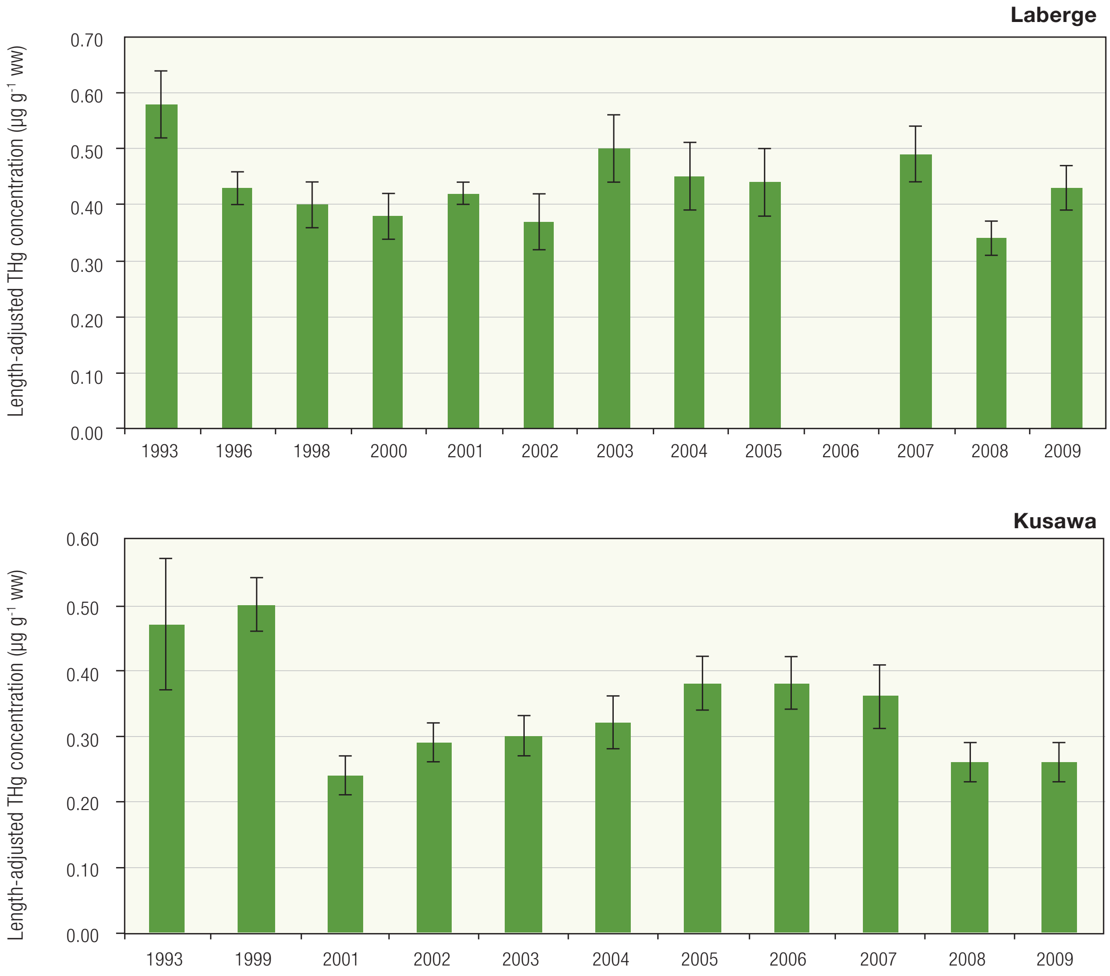

FIGURE 5.24

Length-adjusted THg concentrations (mean ± standard deviation) in lake trout muscle from Lake Laberge (1993 to 2009) and Kusawa Lake (1993 to 2009) (Stern 2010b). For Kusawa Lake, only trout less than 700 mm in length were included in the analysis.

the Northwest Territories are showing similar Hg increases. Larger lakes located in the far north such as Colville Lake and Lac Belot and great lakes such as Great Bear and Great Slave have lake trout with lower THg levels overall and showed less of an increase. It is unclear what environmental factors are driving these recent trends in Hg bioaccumulation, and additional study should examine the role of climate change in the region as well as potential changes in deposition patterns of atmospheric Hg.

Temporal trends of Hg in lake trout from the Yukon

Concentrations of Hg in lake trout were monitored in Laberge and Kusawa lakes in the Yukon from 1993 to 2009 with 12 and 11 yearly measurements, respectively (Fig. 5.24). Mean THg concentrations in muscle during that period were 0.47 ± 0.22 µg g^{-1} ww for Lake Laberge (*n* = 123) and 0.38 ± 0.25 µg g^{-1} ww for Kusawa Lake (*n* = 104). Mean concentrations in both lakes were just below the 0.50 µg g^{-1} consumption guideline for commercial sale. After adjusting concentrations for the effect of fish length, no significant temporal trend in THg was observed in Lake Laberge lake trout over the last 16 years (Fig. 5.24). In Kusuwa Lake, length-adjusted THg concentrations did vary significantly between years. After a significant drop in THg in 2001, concentrations in lake trout increased until 2007 followed by a second significant decline measured in both 2008 and 2009. It is unclear what factors are driving the temporal variation in Kusawa Lake.

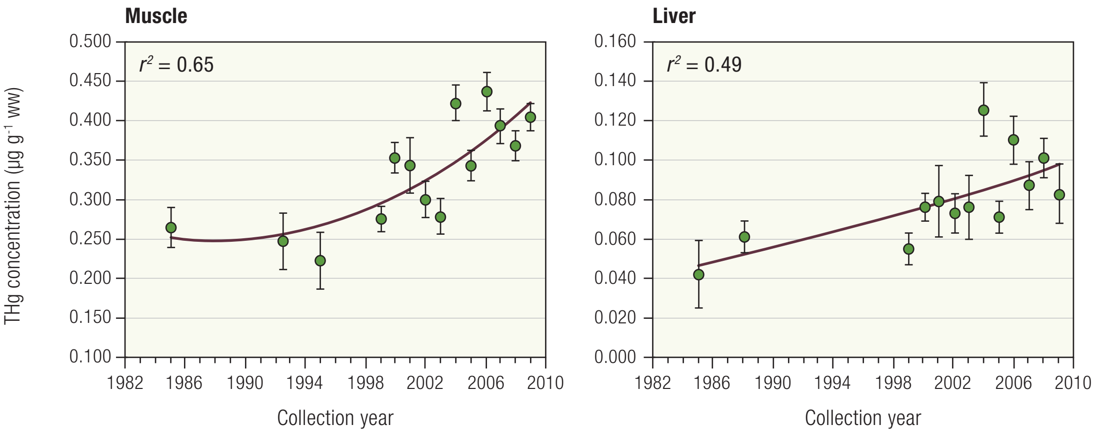

FIGURE 5.25

Temporal trends in mean unadjusted THg concentrations in muscle (left) and liver (right) from Fort Good Hope burbot (Stern 2010b).

5.8.4 Burbot

Temporal trends of Hg in burbot from Fort Good Hope

Long-term monitoring of Hg in burbot has been ongoing at the Rampart Rapids of the Mackenzie River near Fort Good Hope since 1985 (Stern 2010b). Burbot is a top predator species and an important staple for communities in northern Canada. For the monitoring program at Fort Good Hope, burbot were collected in December or January by local residents and analyzed for age (using otoliths), sex, size, THg in muscle and liver, and stable isotopes ($\delta^{13}C$, $\delta^{15}N$) in muscle (Carrie et al. 2010).

Over a 25-year period from 1985 to 2009, concentrations of THg in both muscle and liver increased approximately two-fold (Fig. 5.25). This temporal increase cannot be explained by differing physiological characteristics or changes in feeding behaviour (Carrie et al. 2010). There were no significant changes in $\delta^{15}N$ and $\delta^{13}C$ ratios between 1995 and 2005, implying that burbot had not altered their diet over time. Nor were there significant differences in $\delta^{15}N$ between years of lower THg concentration (1999: 0.24 ± 0.06 $\mu g\ g^{-1}$; mean ± standard deviation) and higher THg concentration (2004: 0.40 ± 0.09 $\mu g\ g^{-1}$). Additionally, no correlation was found between muscle THg concentration and $\delta^{15}N$ or $\delta^{13}C$ ratios. Muscle THg concentrations were not related to fish weight or length, although there was a weak, albeit statistically significant, correlation with age. This last finding is contrary to observations for other fish species (Evans et al. 2005, Gantner et al. 2009b) and may be due to a relatively short half-life of Hg in burbot muscle and liver, as suggested by Carrie et al. (2010). Those authors also concluded that changes in atmospheric deposition could not account for the increasing trend in burbot THg concentration because atmospheric Hg concentrations have been stable or falling in the Arctic over the last 25 years. However, other environmental factors in addition to atmospheric concentration can determine the net deposition of Hg to terrestrial surfaces (Dastoor et al. 2008).

To examine historical trends in sediment Hg fluxes in the watershed, a dated sediment core was collected from Hare Indian Lake, a tributary lake near the Mackenzie River sampling site. Carrie et al. (2010) showed a striking temporal correlation between THg concentrations in burbot muscle and in sediment core slices when plotted against year of fish collection and median year of sediment deposition, respectively (Fig. 5.26). Furthermore, the sediment core showed strong co-variation over the last 150 years between concentrations of THg and labile, algal-derived organic matter (measured as S2 carbon by Rock-Eval pyrolysis; Sanei and Goodarzi 2006) (Fig. 5.26). Concentrations of THg in the sediment core were normalized against the conservative element titanium (Ti) to correct for potential geogenic inputs of Hg.

The association between THg and algal organic matter may reflect Hg scavenging by primary producers, similar to what has been reported in other Arctic lakes (section 5.4.2). The increase in sediment accumulation of algal-derived organic matter since the 1980s is consistent with the climate record of the Mackenzie River basin. The mean annual air temperature for nearby

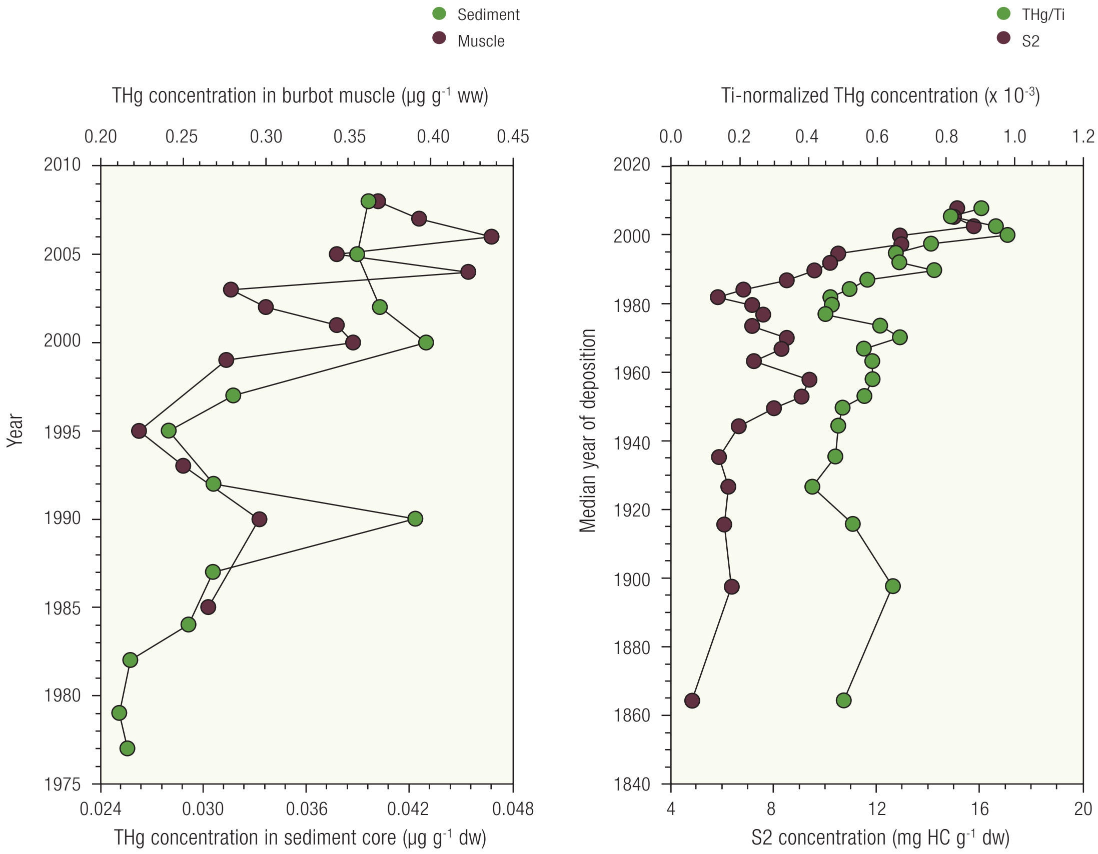

FIGURE 5.26

Chronological variation of THg concentrations in burbot from the Mackenzie River and in a sediment core from nearby Hare Indian Lake (left panel) as well as a comparison between THg concentrations—normalized for geogenic inputs using Ti—and algal (S2) carbon concentrations in the same sediment core (right panel). Reprinted with permission from Carrie et al. (2010). © American Chemical Society.

Norman Wells, for instance, has increased significantly by approximately 1.9°C since the early 1970s. This region has also seen a significant decrease in snow cover over the same time period. Reduction in the period of ice cover and consequent improvement in the light regime for algal growth is thus a plausible explanation for the influence of rising air temperature on algal productivity. The striking increase in burbot THg concentration, coincident with the temporal trends in lake algal productivity and sediment Hg fluxes, suggest that climate warming may mobilize Hg, which, in turn, results in increased exposure for Arctic fish.

5.8.5 A multi-species assessment of temporal Hg trends in Great Slave Lake

Beginning in 1999, a long-term monitoring program of contaminants in fish was initiated for Great Slave Lake as part of the NCP. Three top predator species are currently being monitored: lake trout, burbot, and northern pike. Monitoring in Great Slave Lake is being conducted in two general regions of the lake: 1) the West Basin which is more shallow (mean depth = 41 m, maximum depth = 163 m), warm and productive, and strongly influenced by the Slave River inflow; and,

2) the East Arm, including Christie Bay, which is deeper (mean depth = 199 m, maximum depth = 614 m), colder and less productive (Rawson 1955, Fee et al. 1985, Evans 2000). Data collected before 1999 also exist from other NCP studies in Great Slave Lake and periodic monitoring of the commercial fishery which operates out of Hay River in the West Basin. Only length and weight were measured in earlier fish collections and the sample number was often small. More recent sampling under the NCP includes measurements of age, carbon and nitrogen stable isotopes, and percent lipid. Studies of the three species and the two ecological regions of Great Slave Lake are focussing on factors that affect fish Hg levels, which can then be generalized more broadly to other Arctic regions.

Burbot caught in the West Basin from 1992 to 2008 show a trend of THg increase (p = 0.046), although muscle concentrations remained well below the 0.5 μg g^{-1} guideline for commercial sale (Fig. 5.27). The monitoring record for burbot in the East Arm is shorter and more sporadic. Nevertheless, this more limited record also shows a significant trend of THg increase from 1999 to 2009 (p = 0.03). Lower THg concentrations in burbot from the East Arm compared to the West Basin are due to the collection of younger and smaller fish in the East Arm. These trends persisted when general linear model analyses were conducted using the full suite of available independent variables.

A longer time record exists for lake trout in the West Basin with commercial fish records going back to the late 1970s (Fig. 5.27). Again, these data show that THg concentrations in muscle are significantly increasing either when analyses are based on 1979 to 2009 data (p = 0.008) or are limited to NCP data from 1993 to 2009 and are adjusted for the full suite of available independent variables. The actual trend of increase is less steep than for burbot, possibly because lake trout inhabit more offshore and colder waters and are less exposed to near-shore factors which could be driving the increase. While THg concentrations in lake trout in the East Arm appear to have increased, this trend was barely significant (p = 0.053) and was not increasing when all independent variables were considered in a general linear model. Given that lake trout is more pelagic than burbot and the East Arm is colder and deeper than the West Basin, the more muted increase in THg concentrations in lake trout in the East Arm may be explained by lake size and depth, and fish biology.

There is a more limited record for Hg levels in northern pike in the West Basin, consisting of measurements on fish caught from the domestic fisheries in the 1980s and early 1990s as well as periodic sampling for NCP studies. While THg concentrations appear to be slightly increasing (Fig. 5.27), this trend is not statistically significant even when fish length is considered. The reason for a lack of a trend is unclear, particularly since pike was anticipated to be the most responsive to changes in Hg inputs and transformations in the nearshore environment. While it is possible that the dataset is too small and insensitive to detect trends, other factors may be affecting the Hg trends in pike. For example, growth rates may have increased over the time period due to a warming trend, leading to growth dilution of Hg. Since northern pike is a warm water species, its growth may respond more to a warming trend than species such as lake trout that reside in cool to cold waters.

Historical Hg trends in sediments of Great Slave Lake

Sediment cores were collected from three locations in Great Slave Lake in 2009 to assess historical trends in Hg fluxes. The two locations in the West Basin were offshore of Hay River (site 12) and Fort Resolution (site 19). A third core was collected in the East Arm near Lutsel K'e. Sediment THg concentrations in the two West Basin cores averaged 83.2 ng g^{-1} and 60.2 ng g^{-1}, respectively, and there were no obvious temporal trends in concentrations (Fig. 5.28). In the East Arm, sediment THg concentrations were substantially lower, averaging 31.5 ng g^{-1}, and again no temporal trend in concentration was apparent.

Although THg concentrations were about 1.9–2.6 times lower in the East Arm core than in the West Basin cores, muscle THg concentrations of lake trout and burbot showed considerably smaller differences between the two regions of the lake (Evans and Muir 2010b). This indicates that THg concentrations in sediments were not strongly linked with Hg concentrations in fish. Rates of THg flux to sediments showed a long-term trend of increase in the East Arm while the trend was more variable for the West Basin sites (Fig. 5.28). Trends in THg flux largely reflected trends in sedimentation rate (data not shown). Algal (S2) carbon in sediments showed a pronounced trend of increasing concentration since the 1990s. However, this trend did not mirror trends in sedimentation rates

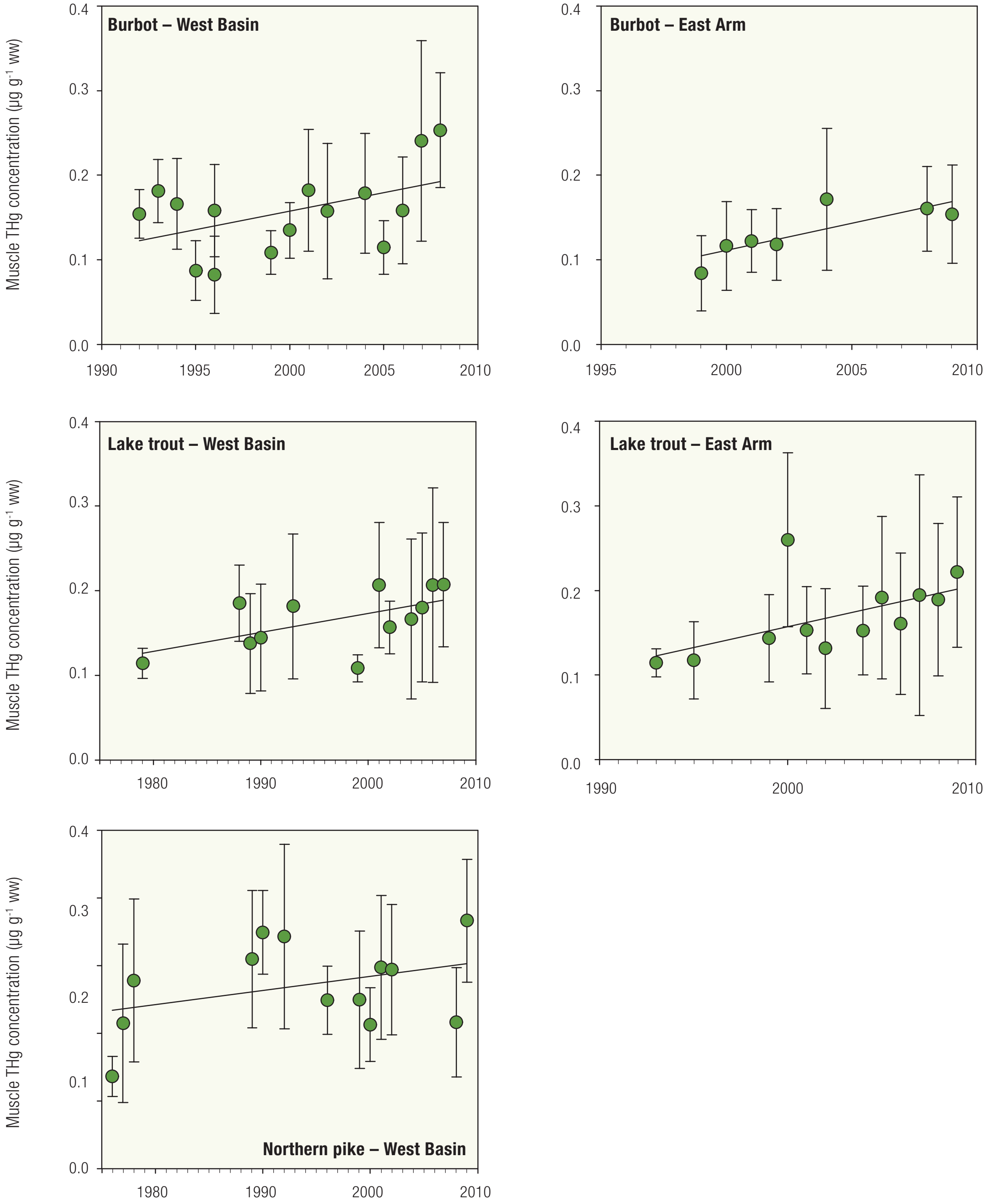

FIGURE 5.27

Temporal trends of THg in the muscle of burbot, lake trout and northern pike collected from the East Arm and/or West Basin of Great Slave Lake. Values are mean unadjusted concentrations (± 1 standard deviation), including data from NCP monitoring programs and commercial records for the West Basin (Evans and Muir 2010a, b).

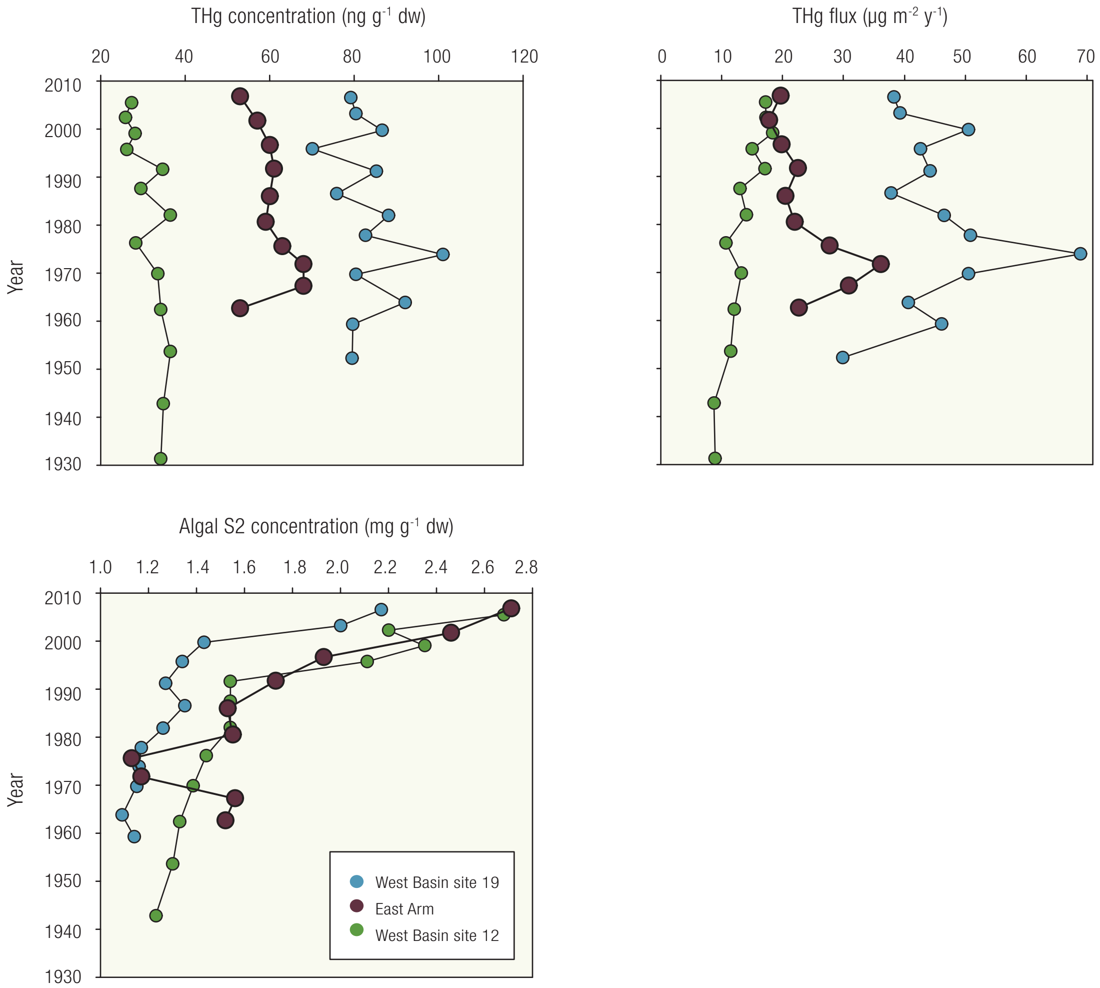

FIGURE 5.28

Temporal trends in THg concentration and flux as well as algal (S2) carbon concentration in two sediment cores from the West Basin (sites 19 and 12) and one from the East Arm of Great Slave Lake (Evans and Muir 2011).

and sediment THg concentrations. Algal S2 concentrations were higher in East Arm sediments possibly because of lower sedimentation rates which resulted in less dilution of algal carbon and/or higher productivity. This coring site is located in a fairly shallow inlet where vertical mixing would be less than in the more offshore sites of the West Basin. Littoral zone algae and biofilms could also be a significant component of this carbon. Upstream of Great Slave Lake in the Slave River at Fort Fitzgerald, total and dissolved phosphorus concentrations showed a linear increase from 1989 to 2006 but not during the more recent period from 1996 to 2006 (Glozier et al. 2009). Thus, the increase in THg concentrations over the past decade in lake trout and burbot from the West Basin, which receives the waters of the Slave River, cannot be explained by greater productivity as a result of increased phosphorus loading to the basin. Overall, the temporal and inter-basin patterns of Hg accumulation in sediments were not reflected in the THg trends for fish in Great Slave Lake.

5.9 Wildlife in freshwater ecosystems

5.9.1 Waterfowl

Waterfowl harvested by northern hunters, specifically ducks and geese, generally have low body burdens of Hg. From the late 1980s to mid-1990s, an extensive survey of contaminants in waterfowl was conducted across Canada that measured THg in the muscle of waterfowl captured at 28 locations in the Arctic (Muir et al. 1997, Braune and Malone 2006a). Bird species that feed mainly on terrestrial grasses and aquatic plants such as geese and dabbling ducks had muscle THg concentrations $< 0.35\ \mu g\ g^{-1}$ ww (Braune and Malone 2006a). Diving ducks, which also feed extensively on aquatic invertebrates, had similarly low THg concentrations (Braune and Malone 2006a). Piscivorous mergansers had higher muscle concentrations of THg up to $1.5\ \mu g\ g^{-1}$ ww due to their higher trophic level (Braune and Malone 2006a). Liver concentrations of THg in dabbling and diving duck species were also measured and were $< 1.5\ \mu g\ g^{-1}$ ww (Braune et al. 2005, Braune and Malone 2006b). Waterfowl from freshwater ecosystems have not been monitored in the Arctic for Hg since the national survey finished in 1995.

5.9.2 Small mammals

Two recent studies were conducted in the Yukon on muskrat and mink, which feed in freshwater ecosystems. Muskrat muscle tissue from the Old Crow flats averaged 1 ng THg g^{-1} dw (Humphries 2010). This low level is typical of an Arctic herbivore that feeds on vascular plants (section 4.8). Reflecting their carnivorous diet of fish and small mammals, mink sampled in the Yukon had hepatic THg concentrations of $0.92 \pm 0.90\ \mu g\ g^{-1}$ ww, similar to those found in Arctic fox (Hoekstra et al. 2003, Gamberg et al. 2005). Female mink had higher concentrations of THg and MeHg in all tissues than their male counterparts, likely because they ingest proportionally more food (and more Hg) due to their smaller body size and their greater energy requirements for reproduction. Mink had higher concentrations of THg in their liver than in their kidney, while the reverse pattern has been observed for terrestrial wildlife (section 4.8). Mink appear to have an efficient demethylation mechanism for MeHg that functions most strongly in the liver, less so in the

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kidney, and least in the brain (Gamberg et al. 2005). Interestingly, the highest THg concentrations in Yukon mink were measured in the same geographical area (Carcross) as the highest concentrations measured in Yukon woodland caribou (section 4.8.2; Fig. 4.8), suggesting a potential local source of Hg for these animals.

5.10 Summary

Since the previous *CACAR*, information on water Hg concentrations has vastly improved as a result of extensive sampling of freshwater ecosystems in the Canadian Arctic. Lakes, ponds, and wetlands were found to have relatively low concentrations of THg, whereas rivers often had considerably higher levels, largely associated with particulate matter. Most water bodies—for which data were available—had low MeHg concentrations, although higher levels were found in some fresh waters where conditions are likely more suitable for *in situ* methylation—shallow lakes, ponds, a river site downstream of wetlands. The export of Hg from large rivers differed among Arctic regions and varied seasonally due to a strong coupling with discharge. In the High Arctic, snowmelt was found to be an important source of THg and MeHg to the water column of lakes.

Bulk concentrations of THg in lake and stream sediments across northern Canada were related to geogenic Hg sources as well as local transport processes, which are affected by climate, vegetation cover, and topography. Colder, generally frozen areas had lower Hg concentrations, while catchments with greater vegetation cover and water bodies with enhanced organic matter accumulation displayed a greater degree of Hg sequestration.

Mercury fluxes to lake sediments have increased several-fold since the onset of the Industrial Era at a large number of sub-Arctic and Arctic sites. While all evidence points to increasing Hg fluxes, there is much debate on whether Hg from anthropogenic sources is entering the lakes in greater amounts or whether other factors such as increased sedimentation and higher algal productivity are attenuating Hg inputs from the atmosphere and lake catchment. The role of climate change has recently been an area of active research, specifically to determine if increased algal productivity in Arctic lakes during the 20th Century has enhanced the rate of Hg scavenging by algae and its transfer to lake sediments. It remains unclear to what extent Hg archives in Arctic lake sediments accurately reflect the deposition history of atmospheric Hg. Thawing permafrost in catchments near the Mackenzie River delta increased sedimentation rates of inorganic material in lakes, while decreasing concentrations of organic carbon, THg, and MeHg in sediments. Preliminary findings also indicated that lakes with thaw slump development had lower concentrations of THg in water and amphipods, but higher Hg bioaccumulation factors for amphipods. Further research is in progress to investigate links between permafrost melting, organic carbon cycling, and Hg bioaccumulation in food webs of Arctic lakes.

The biogeochemical cycling of Hg in Arctic fresh waters remains poorly resolved. Measurements of MeHg production rates are not yet available for aquatic ecosystems in the Canadian Arctic, although projects currently under way will provide first estimates in the near future. The predominant location(s) where methylation occurs have not been identified. That is, whether it is in snow, soil, sediments or in shallow or deep waters. In ponds on Ellesmere Island, elevated water concentrations of MeHg and mass balance modelling suggest that shallow water bodies can be important methylation sites. This mass balance model, which is the first available for MeHg in the Canadian Arctic, also determined that photodemethylation in the water column was a key loss process while summer precipitation contributed only minor inputs of MeHg. Other aspects of the Arctic mercury cycle have received equally little study, including Hg redox transformations and characterization of the microbial community involved in some of these processes.

Several studies highlighted the importance of both Hg supply from the catchment and food web processes in the transfer of Hg to Arctic fish. Cannibalism commonly occurs in landlocked populations of Arctic char, which results in higher concentrations of THg in cannibalistic fish. Landlocked Arctic char were also found to have greater Hg in lakes with larger catchment-to-lake area ratios but concentrations were not related to latitude or longitude. In the West Kitikmeot, the presence of sea-run char was shown to affect Hg transfer to lake trout at locations where the two species co-exist. Resident lake trout had significantly lower THg in lakes that also contained sea-run Arctic char, perhaps due to better growth from greater prey availability. Mercury bioaccumulation in aquatic invertebrates, which can be important prey for trout and char, was influenced by taxonomic composition for zooplankton and by ontogenetic development for chironomids.

The use of stable isotopes of Hg as a tracer of Hg sources and biogeochemical transformations is an emerging field of research. In recent Arctic studies, similar mass-independent fractionation of Δ^{201}Hg was found in snow, lake sediment, and benthic chironomids, whereas different ratios were found in Arctic char and zooplankton. Although further research is needed, this preliminary work suggests that Hg stable isotopes may have the potential to differentiate catchment snowmelt and direct precipitation sources of Hg.

Based on a spatial analysis of common fish species captured between 2002 and 2009, no geographic patterns or regional hotspots were evident for THg in freshwater fish across the Canadian Arctic. Differences in THg concentrations among species were consistent with their general trophic position in aquatic food webs, a pattern that results from biomagnification. Fish muscle concentrations of THg, adjusted for length, did not exceed 0.5 µg g^{-1} ww at the majority of the 85 sites surveyed. However, regional differences in sampling effort were evident, and additional surveys of under-studied regions such as the Kivalliq and eastern Arctic are warranted.

Lake-dwelling char are distinctly smaller than sea-run char and have substantially higher THg concentrations. Preliminary evidence from two study locations near Nain in Labrador suggests that growth rates and trophic position cannot explain lower THg levels in sea-run char compared to lake-dwelling char. Consistent with the study in Labrador, data from the West Kitikmeot indicated that trophic position does not explain differences in THg concentrations among life history types of Arctic char. However, higher THg concentrations in landlocked char relative to sea-run char from the West Kitikmeot were related to slower growth rates and poorer body condition. Differences in THg concentrations among life history types of lake trout were also recently identified in the West Kitikmeot.

Time trends of THg in fish muscle differed among species and locations in the Canadian Arctic. No statistically significant trends were found for landlocked Arctic char in four High Arctic lakes and for lake trout at Lake Laberge in the Yukon. A significant decline in THg recently occurred in lake trout from Kusawa Lake, also in the Yukon. Periodic sampling of lakes in the Mackenzie River basin provided evidence that THg concentrations in lake trout populations have increased markedly in recent years in small- to medium-size lakes. A nearly two-fold increase in THg concentration occurred in burbot muscle from the Mackenzie River near Fort Good Hope between 1985 and 2009. Algal productivity and THg fluxes to sediment in a nearby lake have increased in recent decades, which suggest that climate warming may mobilize Hg and lead to increased exposure to fish. Mercury concentrations also increased in burbot and lake trout from Great Slave Lake over the last two decades, although at a slower rate, possibly due to the muting effects of lake size, depth, and water temperature. There was no THg trend in northern pike in Great Slave Lake. Mercury fluxes to sediment and increased algal productivity did not explain the THg trends for fish in Great Slave Lake. Overall, the different Hg trends reported for Arctic fish indicate that the drivers of such change may be regional or habitat-specific. Further research is required to identify the underlying processes leading to changes in fish Hg concentration over time, particularly at sites where increases were observed.

Wildlife in Arctic freshwater ecosystems have received little attention in recent years. An extensive survey of waterfowl in the 1990s indicated that various species of waterfowl typically harvested by northern hunters contained low amounts of THg. Limited sampling of mink and muskrat showed THg levels that would be expected for their dietary habits.

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Marine Environment

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6.1 Introduction

The marine environment in the Canadian Arctic is truly vast and diverse. It includes deep basins of the Arctic Ocean, many fjords, channels and straits in the Arctic Archipelago, Hudson Bay (the world's largest northern inland sea) and large, productive polynyas such as the North Water in Baffin Bay. Over the last two decades, investigations of Hg in these marine ecosystems have largely focussed on marine mammals and birds because of their dietary and cultural importance for northern Aboriginal peoples and the presence of elevated Hg levels in some animals. Marine mammals such as seals, beluga, and polar bears generally have more Hg than terrestrial mammals and freshwater fish, and this is related (at least in part) to their higher trophic position in marine food webs (Fisk et al. 2003, Muir et al. 1997). Previous studies under the Northern Contaminants Program (NCP) have identified geographic variation in Hg bioaccumulation among populations of marine species, although the factors driving these differences remain poorly resolved. Temporal trend studies have shown recent increases in Hg concentrations in some Arctic animals but not in others. The reasons for these varying trends among Arctic species and populations are also unclear.

Several important advances in our understanding of the marine Hg cycle have been made, particularly for the abiotic environment, since the last *Canadian Arctic Contaminants Assessment Report* (*CACAR II*). Significant amounts of new data have been collected on Hg concentrations in marine waters, snow and ice pack, providing information that was virtually absent in 2002

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(see Bidleman et al. 2003). Data on Hg accumulation in sediments of Hudson Bay are now available, and mass balance models for the Arctic Ocean and Hudson Bay provide quantitative estimates of Hg fluxes into and out of these ecosystems. The first studies on microbial transformations of Hg were conducted in marine waters of the Canadian Arctic. Biomagnification of Hg in the Beaufort Sea food web was studied intensively in response to the dramatic temporal and spatial trends of Hg in beluga that were reported in the previous *CACAR*. New data suggest climate change may be affecting marine food webs with consequences for Hg bioaccumulation in seabirds and seals. Finally, annual monitoring of key marine species during Phase III of the NCP has further strengthened the temporal trend datasets for the marine environment.

6.2 Overview of the Hg cycle in the Arctic marine environment

The movement of Hg through the marine environment is complex because of the sheer size and open-nature of the system, the multiple pathways and transformations that deliver and export Hg, and the diverse habitats—such as continental shelves, deep ocean basins, and sea ice—with different propensities for Hg processing. In addition, it is likely that Hg cycling in the Arctic is strongly affected by environmental characteristics such as seasonality in primary production and light (ranging from 24-hour darkness to 24-hour sunlight), extended ocean ice cover, atmospheric mercury depletion events (AMDEs), large river inputs, and extensive continental shelves (Douglas et al. 2011). There is currently limited empirical information on Hg cycling in the Arctic marine environment (Macdonald and Loseto 2010), and the conceptual model outlined as follows is largely based on information from other oceans (e.g., Morel et al. 1998, Fitzgerald et al. 2007, Sunderland and Mason 2007, Cossa et al. 2011).

Atmospheric, terrestrial, and oceanic pathways deliver Hg to Arctic marine waters, primarily as divalent inorganic Hg [Hg(II)] (Fig. 6.1). Processes of atmospheric deposition (sections 3.2, 3.3 and 3.6), river export (section 5.3), and oceanic currents (section 2.5) are described earlier in the report. Model estimates of Hg

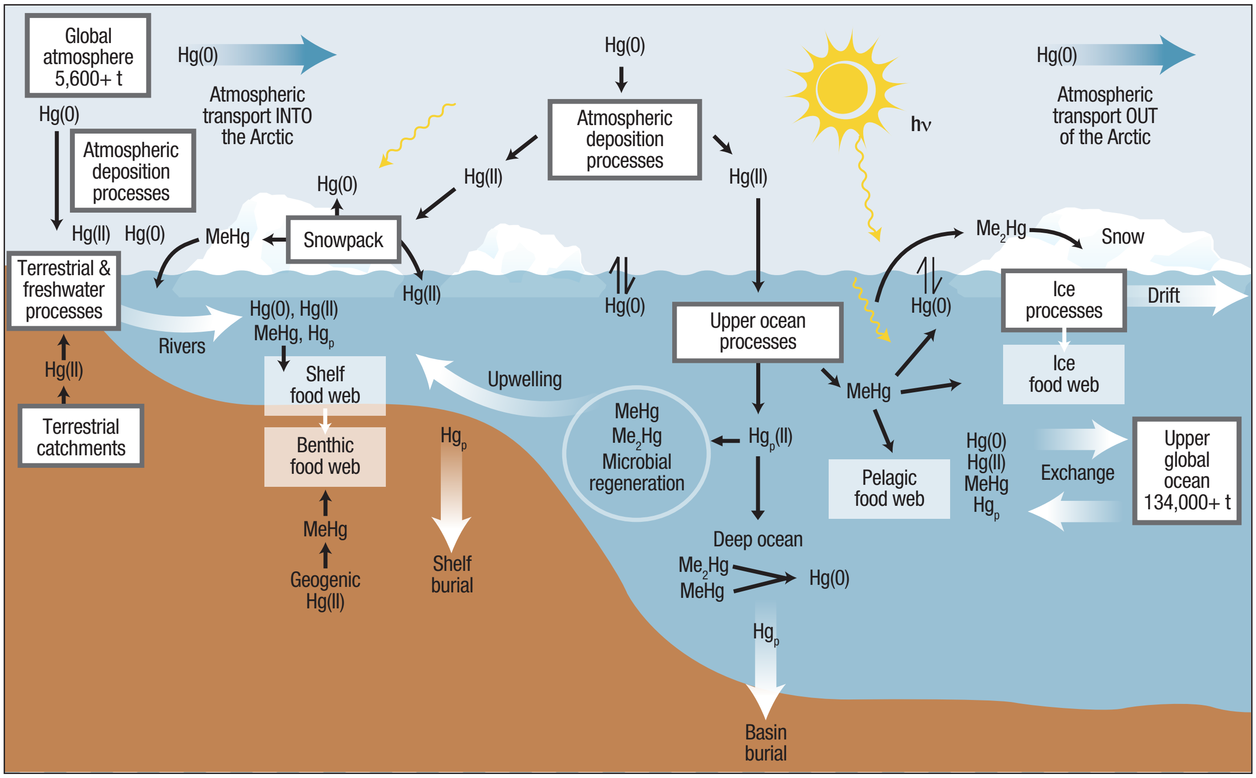


FIGURE 6.1

Schematic of major pathways and transformations in the Hg cycle for Arctic marine ecosystems. Large shaded arrows refer to atmospheric or oceanic transport pathways that exchange Hg with the global environment and move Hg between reservoirs within the Arctic Ocean. Small black arrows refer to chemical processes that transform Hg into different species (source: AMAP 2011).

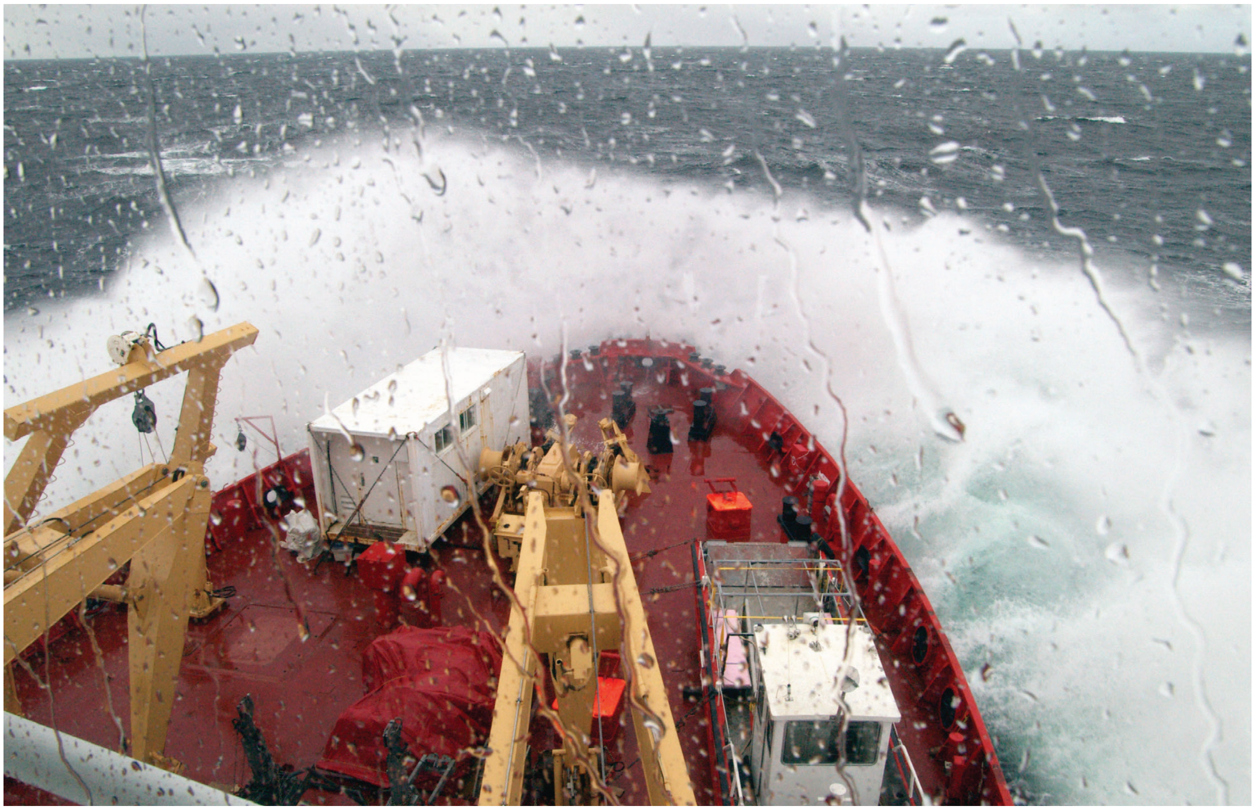
fluxes to the Arctic Ocean and Hudson Bay from each of these pathways are presented later in this chapter (section 6.6). In marine waters, Hg(II) is sequestered, transformed, or removed from the system by: 1) reduction to elemental Hg [Hg(0)] and evasion to the atmosphere; 2) sedimentation of particulate-bound Hg; 3) transformation to monomethylmercury (MeHg) and dimethylmercury (Me_2Hg); and, 4) hydrological removal via ocean currents. The production of MeHg occurs in anoxic sediments and likely also in the water column through either Hg methylation or the breakdown of Me_2Hg (Fitzgerald et al. 2007, Cossa et al. 2011). Aqueous MeHg is broken down to Hg(II) by photodemethylation in the water column or by microbial demethylation (Lehnherr et al. 2011). Although it is impeded by ice cover during the long Arctic winter, evasion of Hg(0) from surface waters to the atmosphere is an important flux driven by microbial or light-induced processes (Fitzgerald et al. 2007, Sunderland and Mason 2007). It has been proposed that sea ice may be important habitat for Hg cycling in the Arctic marine environment (Poulain et al. 2007c, Macdonald and Loseto 2010).

Algae and bacteria are the main entry points for uptake of aqueous MeHg in marine food webs (Atwell et al. 1998, Campbell et al. 2005). These microorganisms transfer MeHg to zooplankton in the water column, to benthic invertebrates on sediment and rock substrates, and to crustaceans such as amphipods and copepods that are associated with sea ice. Methylmercury is then further biomagnified to higher trophic level consumers, namely fish, marine mammals, and seabirds. Some marine mammals have specialized diets while others are generalists that forage on diverse prey in different habitats—including the water column, sea floor, and sea ice (Bluhm and Gradinger 2008)—with potential consequences for trophic transfer of MeHg.

6.3 Ocean waters

6.3.1 Speciation of Hg in Arctic marine waters

An extensive survey was recently conducted of water-column concentrations of total Hg (THg), gaseous elemental Hg (GEM), and methylated Hg (both MeHg and Me_2Hg) throughout the Canadian Arctic Archipelago and Hudson Bay to examine potential zones of production and/or destruction of different Hg species (Kirk et al. 2008). A total of 24 locations were sampled at multiple depths: five in the North Water Polynya, six in the Northwest Passage routes, three in the Hudson Strait, and the remaining ten in Hudson Bay.



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Concentrations of THg were low throughout the water column in both the Arctic Archipelago and Hudson Bay (mean ± standard deviation: 0.39 ± 0.40 and 0.42 ± 0.53 ng L^{-1}, respectively) (see Annex Table A2.4 for concentrations of Hg species at each site). Concentrations of GEM were quite low in surface waters and varied little among sites, averaging 25.4 ± 10.2 pg L^{-1} in the Arctic Archipelago and 29.6 ± 5.0 pg L^{-1} in Hudson Bay. In deeper regions of the water column, concentrations of GEM were variable, ranging from below the method detection limit to 133 pg L^{-1}, and averaging 34.3 ± 29.7 pg L^{-1} in the Arctic Archipelago and 26.5 ± 24.5 pg L^{-1} in Hudson Bay. In addition, concentrations of GEM at different depths within the same site often varied greatly. Therefore, biotic and/or abiotic processes responsible for net GEM production varied both geographically and with depth in the water column. Although reduction of Hg(II) is likely the dominant source of GEM in marine waters, photodegradation of MeHg (Chen et al. 2003) and Me_2Hg at the surface as well as biotic and/or abiotic reductive demethylation may also produce GEM deep in the water column (Mason et al. 1998).

Throughout both the Arctic Archipelago and Hudson Bay, concentrations of methylated Hg and Me_2Hg were generally low in surface waters (23.8 ± 9.9 and 4.7 ± 4.4 pg L^{-1}, respectively) but increased with depth (maximum: 178 and 170 pg L^{-1}, respectively; mean: 70.3 ± 37.3 and 56.8 ± 37.8 pg L^{-1}, respectively). This suggests that methylated Hg species are produced in deep Arctic marine waters and/or sediments (Fig. 6.2). In addition, the percent of THg in a methylated form was extremely high (maximum: 66%; mean: 28 ± 16%) and was associated with extremely low concentrations of THg at mid and bottom depths. Similarly high percentages of THg as MeHg were observed in deep waters of the Southern Ocean (Cossa et al. 2011). For the Arctic Ocean, oceanographic processes, such as water regeneration and vertical mixing, were shown to affect Hg distribution in marine waters (Kirk et al. 2008). Deep-water production may be an important source of MeHg to Arctic organisms feeding in the surface mixed layer, where primary production is generally greatest (Michel et al. 2006), and to surrounding landmasses through Me_2Hg evasion to the atmosphere and subsequent decomposition by sunlight (Niki et al. 1983a, Niki et al. 1983b, St. Louis et al. 2007).

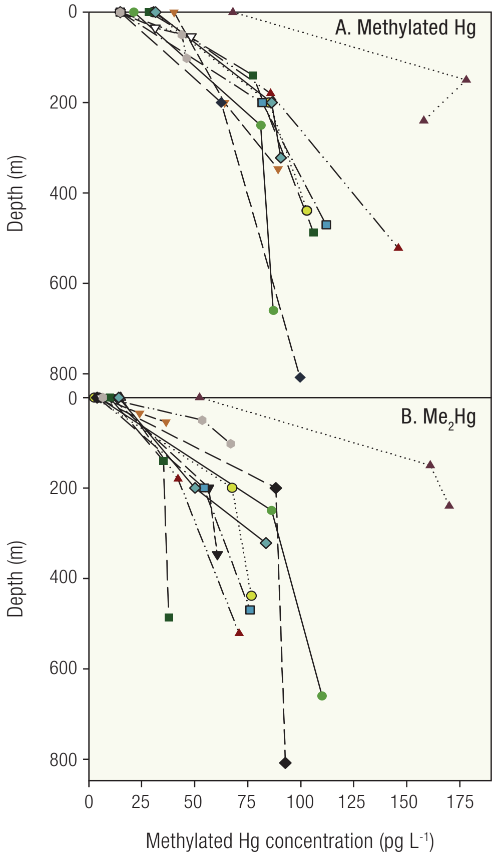

FIGURE 6.2

Water concentrations of methylated Hg (A = both MeHg and Me_2Hg; B = Me_2Hg) (pg L^{-1}) versus depth at sites located in the Canadian Arctic Archipelago. Reprinted with permission from Kirk et al. (2008). © American Chemical Society.

Instantaneous fluxes of gaseous Hg species from the ocean to the atmosphere were calculated to determine if Arctic marine waters are a source for atmospheric Hg. Ocean-atmosphere fluxes of both GEM and Me_2Hg were high (range: 2.6–388 and 0.2–176 ng m^{-2} day^{-1}, respectively; average: 130 ± 138 and 27.3 ± 47.8 ng m^{-2} day^{-1}, respectively), demonstrating that marine waters of the Arctic Archipelago and Hudson Bay region are a substantial source of gaseous Hg to the atmosphere during the ice-free season (Kirk et al. 2008). Interestingly, average ocean-atmosphere GEM fluxes in these regions of the Arctic were higher than those calculated previously for the North Atlantic, Mediterranean, and Baltic waters (2.8–94.7 ng m^{-2} day^{-1}) (Cossa et al. 1997, Mason et al. 2001, Kuss and Schneider 2007). Average ocean-atmosphere Me_2Hg fluxes for the Arctic Archipelago and Hudson Bay were also higher than previous estimates for the Arctic, Antarctic, and Atlantic Ocean (2.8–4.9 ng m^{-2} day^{-1}) (Pongratz and Heumann 1998). Further, the Arctic ocean-atmosphere fluxes of GEM and Me_2Hg were determined during the open-water season and are likely lower than fluxes directly after ice-out when substantial degassing may occur of gaseous Hg species trapped under the sea ice throughout winter (St. Louis et al. 2007). In fact, concentrations of Me_2Hg (11.1 ± 4.1 pg L^{-1}) and GEM (129 ± 36 pg L^{-1}) measured by St. Louis et al. (2007) under the sea ice near Resolute Bay (Nunavut) were higher than those measured in ice-free surface waters of the Arctic Archipelago and Hudson Bay.

6.4 Sea ice and snowpack

6.4.1 Concentrations of Hg in Arctic marine snow

Recent investigations of snow on sea ice and in Arctic coastal areas suggest that inorganic Hg accumulation is enhanced in marine snow relative to the terrestrial environment (Poulain et al. 2007a, St. Louis et al. 2007). Concentrations of THg in excess of 50 ng L^{-1} have been commonly reported for snow on sea ice (Lu et al. 2001, Kirk et al. 2006, St. Louis et al. 2007) but are rare for inland snowpack and have not been observed in glacier ice or snow in the Canadian Arctic (section 4.3). Higher snow THg concentrations may occur in the marine environment because AMDEs are a result of atmospheric photochemical reactions involving marine halogens (Lindberg et al. 2002, Simpson et al. 2007). Marine halogens, particularly chloride, may also affect Hg transformations in the snowpack by limiting Hg evasion through the formation of chloride-Hg complexes that are less susceptible to photoreduction (Poulain et al. 2007b, St. Louis et al. 2007, Bartels-Rausch et al. 2011) or by favouring the photooxidation of GEM (Amyot et al. 2003, Lalonde et al. 2004). In an extensive snow survey in the Canadian High Arctic, St. Louis et al. (2007) found a strong positive correlation between snow chloride and THg concentrations consistent with experimental observations of a chloride effect on Hg(II) retention. Interestingly, MeHg concentrations in snowpacks were not related to chloride concentration (St. Louis et al. 2007).

While THg is often most concentrated in the surface layer, the middle stratum and depth hoar—a layer of large crystals at the base of the snowpack—can also be dominant Hg reservoirs in marine snow (Table 6.1). High THg in lower snowpack layers may be due to Hg(II) deposited during AMDEs that penetrates into snowpacks (St. Louis et al. 2005). Strong winds may

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also cause wind pumping and enhance the transfer of reactive gaseous Hg (RGM) and particulate Hg (PHg) into deeper snow layers. Burial through snow accumulation, sublimation, condensation, and ice-layer formation are other processes that promote Hg retention in the snowpack (Douglas et al. 2008). In contrast with THg, average MeHg concentrations were generally low (< 0.1 ng L^{-1}) in both surface and deeper layers of marine snowpack on Hudson Bay and in the Canadian High Arctic (Kirk et al. 2006, St. Louis et al. 2007).

Areal loads of THg and MeHg in snow were estimated to determine their importance as a source of Hg to Arctic marine waters during spring melt. In the High Arctic, median loads in snowpacks were estimated at 5.2 mg THg ha^{-1} and 0.03 mg MeHg ha^{-1} (St. Louis et al. 2007), while in Hudson Bay, the average snow THg loading was 2.1 mg ha^{-1} (Kirk et al. 2006). These areal loads from snow were relatively low in comparison to annual wet deposition rates at lower North American latitudes which range from 22–200 mg ha^{-1} (Mercury Deposition Network data, as cited in St. Louis et al. 2007). These authors concluded that, overall, snow likely contributes low amounts to the marine pools of Hg in the Arctic Archipelago and Hudson Bay (Kirk et al. 2006, St. Louis et al. 2007).

6.4.2 Rapid re-emission of Hg from snow following AMDEs at Churchill (Manitoba)

In the spring of 2003 and 2004, surface snow was collected and analyzed for concentrations of THg and MeHg every 1–2 days from a site 1 km offshore on Hudson Bay sea ice near Churchill, Manitoba (Kirk et al. 2006). In addition, concentrations of THg were measured in snow samples from different layers of snowpacks at sites located 2.5, 5, and 7 km from the shoreline on three occasions in the spring of 2004. Due to the deposition and subsequent photoreduction of inorganic Hg(II) in snowpacks during AMDEs, average concentrations of THg in surface snow were quite high and variable throughout the spring of both years (in 2003: 111.4 ± 161.2 ng L^{-1}, $n = 23$; in 2004: 40.7 ± 69.7 ng L^{-1}, $n = 59$) (Fig. 6.3; see Fig. 3.4 for AMDEs at the site).

In the spring of 2004, when atmospheric Hg species were continuously monitored at this site, concentrations of THg in snow averaged 67.7 ± 97.7 ng L^{-1} during AMDEs. However, following the end of each AMDE, average concentrations of THg in surface snow declined dramatically and, by four or more days after AMDEs, were only 4.25 ± 1.85 ng L^{-1} (Fig. 6.4). Finally, THg concentrations in melt water collected

TABLE 6.1 Comparison of THg concentrations in different layers of the snowpack (surface [S], middle stratum [M], depth hoar [D]) at various sea ice locations in the Canadian Arctic. Observations are presented in order of increasing concentrations (mean ± 1 standard deviation, *n* in parentheses) in the surface layer. Modified from AMAP (2011).

Location	Date	Snow THg Concentration (ng L^{-1})			Rank of Snow Strata	Reference
		Surface	Middle Stratum	Depth Hoar		
Resolute, NU	June 9, 2004	1.8 ± 0.6 (3)	4.9 ± 0.5 (3)	45.2 ± 13.1 (3)	D > M > S	Poulain et al. (2007a)
Allman Bay, NU	May 10, 2004	3.6 (1)	0.3 (1)	0.6 (1)	S > D > M	St. Louis et al. (2007)
Talbot Inlet, NU	May 10, 2004	4.5 (1)	0.4 (1)	7.9 (1)	D > S > M	St. Louis et al. (2007)
Alert, NU	April 12, 2002	5.7 (1)	0.4 (1)	2.5 (1)	S > D > M	St. Louis et al. (2005)
Buchannan Bay, NU	May 10, 2004	6.1 (1)	1.3 (1)	5.4 (1)	S > D > M	St. Louis et al. (2007)
Norwegian Bay, NU	May 16, 2004	8.0 (1)	8.1 (1)	1.2 (1)	M > S > D	St. Louis et al. (2007)
Alert, NU	April 22, 2002	11.1 (1)	21.1 (1)	1.3 (1)	M > S > D	St. Louis et al. (2005)
Eureka Sound, NU	May 16, 2004	15.9 (1)	1.4 (1)	9.8 (1)	S > D > M	St. Louis et al. (2007)
Bay Fiord, NU	May 16, 2004	19.8 (1)	18.1 (1)	48.6 (1)	D > S > M	St. Louis et al. (2007)
Churchill, MB	March 31, April 16, May 22–23, 2004	21.4 ± 27.2 (9)	15.2 ± 13.8 (9)	10.6 ± 9.6 (9)	S > M > D	Kirk et al. (2006)
Wellington Channel, NU	May 16, 2004	66.4 (1)	3.3 (1)	2.3 (1)	S > M > D	St. Louis et al. (2007)
Jones Sound, NU	May 11, 2004	78.2 (1)	8.0 (1)	17.1 (1)	S > D > M	St. Louis et al. (2007)
Makinson Inlet, NU	May 10, 2004	150 (1)	253 (1)	281 (1)	D > M > S	St. Louis et al. (2007)

Note: Concentrations in each layer were measured in the same snowpack on a single sampling date except observations at Churchill which are means of multiple sampling dates and/or snowpacks.

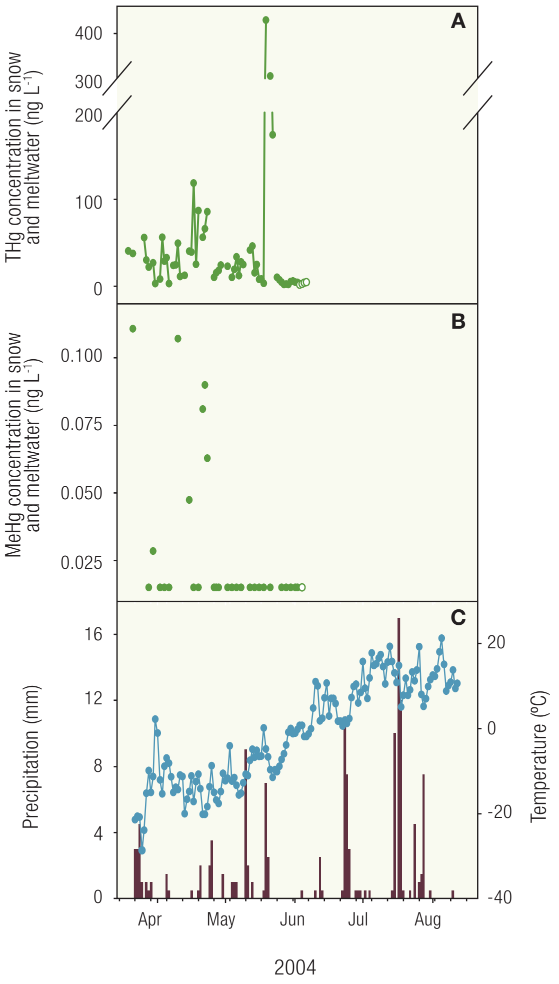

FIGURE 6.3

Concentrations of THg (A) and MeHg (B) in surface snow (closed circles) and melt water (open circles) collected throughout the spring of 2004 on Hudson Bay sea ice near Churchill (Manitoba). Samples with concentrations of MeHg below analytical detection were assigned the detection limit concentration (0.015 ng L^{-1}). (C) Daily precipitation (bars) and temperature (closed circles) were measured by Environment Canada at the Churchill airport. Reprinted with permission from Kirk et al. (2006). © American Chemical Society.

at spring snowmelt were low in both years. In 2003, meltwater THg concentrations collected on May 12th and 14th were only 7.54 and 3.49 ng L^{-1} respectively ($n = 2$). In 2004, snowmelt did not begin until the first week of June. However, average THg concentrations in melt water of 3.31 ± 1.35 ng L^{-1} ($n = 4$) were similar to those observed the previous year. Although AMDEs resulted in deposition of Hg(II) into the upper layers of the snowpack, they did not alter MeHg concentrations in snow because GEM is not oxidized directly to MeHg.

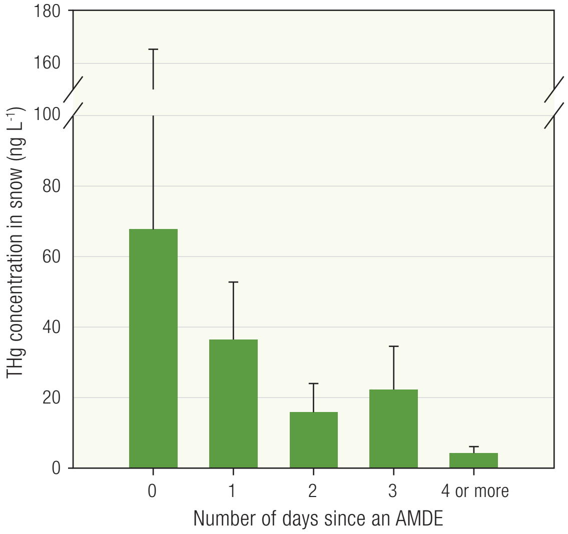

FIGURE 6.4

Average THg concentrations (± 1 standard deviation) in surface snow collected on Hudson Bay in 2004 during AMDEs, and one to four or more days following the end of individual AMDEs if additional AMDEs did not occur. Reprinted with permission from Kirk et al. (2006). © American Chemical Society.

In fact, MeHg concentrations in snow were often below the method detection limit of 0.015 ng L^{-1}. On several days following snowfall events, wet precipitation appeared to increase MeHg concentrations in surface snow to between approximately 0.06–0.11 ng L^{-1} ($n = 30$) (Fig. 6.3).

6.4.3 Distribution of Hg in Arctic sea ice

A detailed study of Hg dynamics in Arctic sea ice was recently conducted in the Beaufort Sea, mainly at sites offshore from Banks Island (Chaulk et al. 2011). A range of ice types were sampled: newly formed ice, first year ice (both drifting and landfast), and multiyear ice. Despite the variation of ice types, ice characteristics (thickness, stage of ice formation, extent of snow cover) and ambient air temperatures, THg in ice was consistently low (0.5–4 ng L^{-1}), with the highest concentrations found in the surface layer. Local AMDEs observed at the time of sampling did not appear to strongly impact THg concentrations in new ice or surface seawater. However, brine in the sea ice was substantially more concentrated in THg (2.6–71.2 ng L^{-1}), with higher levels found in more saline brines. The formation of brine pockets and channels was found to be an important process for Hg movement within the ice pack.

Sea-ice accumulation of Hg may potentially be driven by three major processes: 1) freeze rejection of Hg from seawater; 2) scavenging of atmospheric Hg by exposed ice surfaces; or, 3) leaching of Hg from overlying snow (Chaulk et al. 2011). Evidence collected from the ice survey in the Beaufort Sea suggested that freeze rejection from seawater and, in some cases, leaching from the overlying snow contributed to the observed Hg concentrations. Surface enrichment of sea ice was a ubiquitous phenomenon, probably due to the enrichment of brine and particles in surface frazil ice. No evidence was found for efficient scavenging of atmospheric Hg in newly formed frazel ice, although Douglas et al. (2008) observed very high THg concentrations in ice crystal formations (surface hoar, frost flowers, diamond dust) on the Alaskan coast. Brine pockets and channels, which concentrate Hg, are also important microbial niches in sea ice. It has been proposed that these Hg-rich environments may be key sites for microbial transformation and uptake of Hg into the pelagic, ice-based food web (Poulain et al. 2007c, Macdonald and Loseto 2010, Chaulk et al. 2011).

6.5 Marine sediments

6.5.1 Sediment Hg profiles in Hudson Bay

Little information exists on Hg concentrations in marine sediments from the Canadian Arctic, and the largest dataset currently available is for Hudson Bay. The first sediment investigations in Hudson Bay were conducted by Lockhart et al. (1998) and Stewart and Lockhart (2005) who reported THg concentrations in three cores taken from the southeastern region in 1992 and 1993. An additional 13 cores were collected during an ArcticNet study conducted in 2005 by Hare et al. (2008, 2010). The recent cores were collected at 11 sites across Hudson Bay, one site at the connection with Hudson Strait, and one site in the western section of Hudson Strait (Fig. 6.5).

Sediment THg concentrations were low in Hudson Bay compared to other marine regions in the circumpolar Arctic. In Hudson Bay, sediment THg ranged from 8–58 ng g^{-1} dry weight (dw) with a mean of 26 ng g^{-1} (n = 325) (Hare et al. 2008, Hare et al. 2010, Lockhart et al. 1998, Stewart and Lockhart 2005). These values were at the low end of concentrations observed in other sediment studies on the Arctic Ocean, the Beaufort Shelf, and along the Greenland coast (Fig. 6.6). No published measurements of THg concentrations are currently available for marine sediments of the Arctic Archipelago. The Hudson Bay values were also low compared to THg concentrations in freshwater sediments observed across the Canadian Arctic (section 5.4.2). Such low levels of sediment Hg in Hudson Bay likely reflect low geological inputs from the watershed and low concentrations of incoming particulate matter accumulating in the sediments.

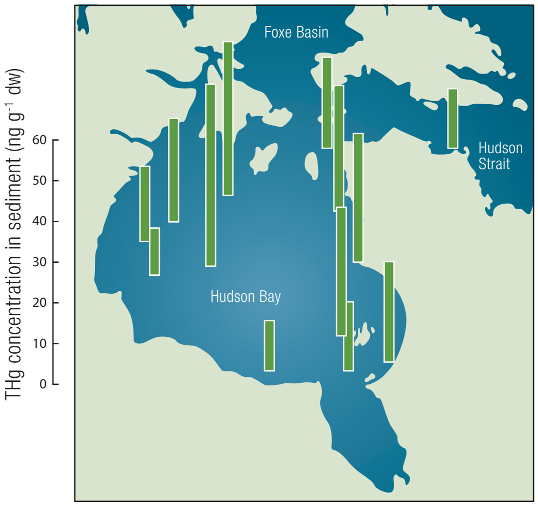

FIGURE 6.5

Core locations in Hudson Bay and Hudson Strait with concentrations of THg (ng g^{-1} dw) in surface sediment (Hare et al. 2010).

Spatial variation of THg in Hudson Bay sediments appeared to be largely controlled by the sorting of Hg-bearing particles from the water column. Surface THg concentrations were positively related to the fraction of surficial silt and clay (percent < 63 µm) and to water depth, but not distance offshore. Two deepwater cores (sites 3 and 15; Fig. 6.5) from depths > 400 m in the Hudson Strait were exceptions and had low silt and clay content and low sediment Hg concentrations for their depth. Sediments in the Hudson Strait may receive particles from a different source such as ice-rafted particulate matter from the shallow Foxe Basin (Kuzyk et al. 2008).

Vertical profiles of THg in Hudson Bay sediments were examined for historical changes related to atmospheric deposition of anthropogenic Hg (Hare et al. 2010). Sediment mixing processes were modelled to determine the simplest chronology of Hg inputs that could explain the observed vertical THg profiles in Hudson Bay sediments. The model assumed steady

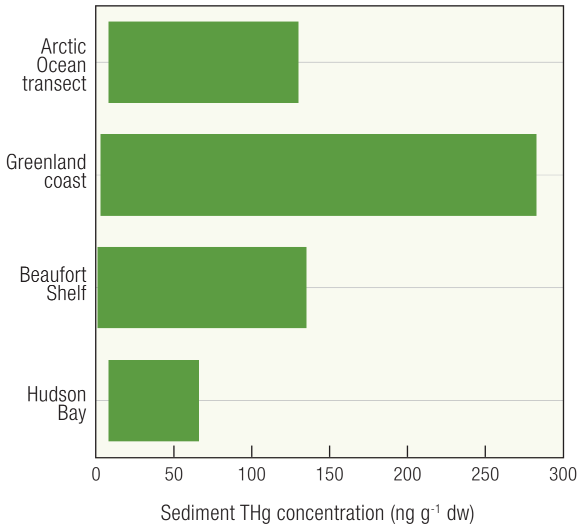

FIGURE 6.6

Ranges of sediment THg concentrations in Hudson Bay (Hare et al. 2010) and other marine regions in the circumpolar Arctic, specifically the Arctic Ocean (Gobeil et al. 1999), the Beaufort Shelf (Macdonald and Thomas 1991), and the Greenland coast (Asmund and Nielsen 2000).

sedimentation and mixing rates over time, and negligible diagenetic mixing of Hg relative to biomixing. These assumptions were supported by several recent geochemical studies of the same core set (Kuzyk et al. 2008, 2009). The modelled chronologies demonstrated that stable Hg concentrations existed in most cores at sediment depths accumulated before the industrial age (circa 150 years ago). In these cores, sediment THg fluxes increased during the early days of the Industrial Era—1870 to 1930—followed by stable or declining Hg inputs toward the end of the 20th Century. The flux patterns were consistent with general historical patterns of anthropogenic Hg emissions in North America. Several other cores in Hudson Bay displayed consistent increases in THg concentration in deep sediment accumulated before the Industrial Era. Concomitant increases in the proportion of marine carbon in some of the latter cores suggest that natural changes in the materials incorporated into sediments can increase THg concentrations as much as those potentially caused by anthropogenic impact. Based on differences between background and surface THg concentrations, Hare et al. (2010) estimated that Hudson Bay has accumulated 52–80 t of anthropogenic Hg in its sediments since the onset of the Industrial Era. This estimate of anthropogenic Hg addition in Hudson Bay was deemed small relative to recent annual rates of anthropogenic Hg emissions in North America (Hare et al. 2010).

6.6 Mass balance models

6.6.1 Mass balance inventory of Hg for the Arctic Ocean

The Arctic Ocean is a semi-enclosed mediterranean sea, which provides the opportunity to calculate a Hg budget and make direct comparisons of the relative importance of inputs to the system (Fig. 6.7; Outridge et al. 2008). An inventory of THg in the Arctic Ocean was developed with a reasonably well-balanced sediment and water budget (Stein and Macdonald 2004). Fluxes for the various pathways were estimated with THg concentrations and fluxes of media obtained through a comprehensive literature survey. A modified version of the Global/Regional Atmospheric Heavy Metal (GRAHM) model (Dastoor and Larocque 2004) was used to estimate the net atmospheric flux of THg into the Arctic Ocean. Detailed discussions of how the budget components were calculated and potential errors in the estimates are given in Outridge et al. (2008).

For the Arctic Ocean as a whole, the largest single source of Hg was net atmospheric deposition, which contributed 48% (98 t y^{-1}) of the total annual Hg input (Fig. 6.7; Outridge et al. 2008). The GRAHM model incorporated a re-emission term of 133 t y^{-1} (volatilization of Hg from snowpack, snowmelt, and runoff, equivalent to 55% of gross deposition) and evasion term of 12 t y^{-1}. Thus, the gross atmosphere-ocean flux was 243 t y^{-1}. Of the net flux, 46% (45 t) occurred

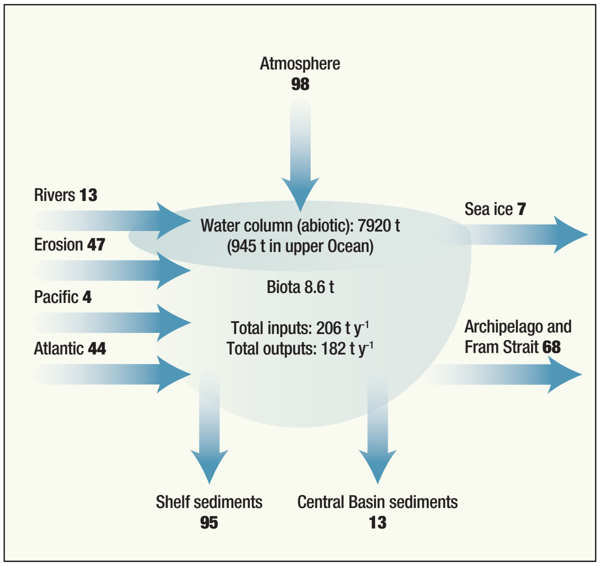

FIGURE 6.7

Mass balance model of THg (t y^{-1}) in the Arctic Ocean with flux components. Reprinted with permission from Outridge et al. (2008). © CSIRO Publishing.

during springtime and 54% (53 t) during the rest of the year. Inflows from the Atlantic and Pacific oceans (23%), and coastal erosion (23%) contributed most of the remaining THg to the system. Collectively, rivers were a minor source.

Sedimentation was the single largest loss pathway for Arctic marine THg and represented 59% of the total 182 t y^{-1} removed from the Arctic Ocean by all processes. In the Arctic Ocean, an estimated sediment Hg flux of 95 t y^{-1} occurs in the continental shelf seas and 13 t y^{-1} in the Central Basin for a total sequestration of 108 t y^{-1}. It is also noteworthy that the shelf sequestration rate almost balanced the net atmospheric Hg input of 98 t y^{-1}, which suggests the possibility of a scavenging of deposited atmospheric Hg from the shelf water column into sediments (Outridge et al. 2008). Seawater export through the Arctic Archipelago and the Fram Strait represented 37% of THg output from the Arctic Ocean. Total estimates of Hg inputs and outputs were almost in balance with a net annual increase of 0.3% in total mass.

Outridge et al. (2008) concluded that Hg deposition from AMDEs and atmospheric deposition of Hg in general, do not have the capacity to produce the short-term Hg variations observed in some marine mammals in the Arctic Ocean, especially in light of inputs from riverine fluxes, erosion, and ocean exchange. The top 200 m of the Arctic Ocean was estimated to contain 600–1,000 t of THg, which is far larger than estimated annual fluxes into or out of the surface Arctic Ocean. From this perspective, the proportion of AMDE Hg entering the upper ocean contributes a relatively small amount to what is already a large reservoir of inorganic Hg (Outridge et al. 2008). This reservoir is maintained through atmosphere and ocean transport of Hg, and includes a natural background augmented during the past two centuries by human activities (Sunderland and Mason 2007). However, significant limitations remain in our understanding of the fundamental physical and chemical properties of newly deposited Hg and, hence, its post-depositional fate is unclear. Further, our understanding of methylation processes that supply MeHg to food webs in the Arctic is also currently limited.

6.6.2 Mercury budget for Hudson Bay

Mass balances of THg were modelled for Hudson Bay to investigate pre-industrial and contemporary sources and pathways of THg and their sensitivity to projected climate change (Hare et al. 2008). Estimates of atmospheric deposition, mass exchange of marine water, river inputs, and sedimentation of THg were primarily based on measurements taken within the Hudson Bay system by Hare et al. (2008), Kirk et al. (2006), and Sanei et al. (2010), while estimates of Hg fluxes from coastal erosion and surface water evasion were modelled from measurements taken in similar marine environments (Hare et al. 2008).

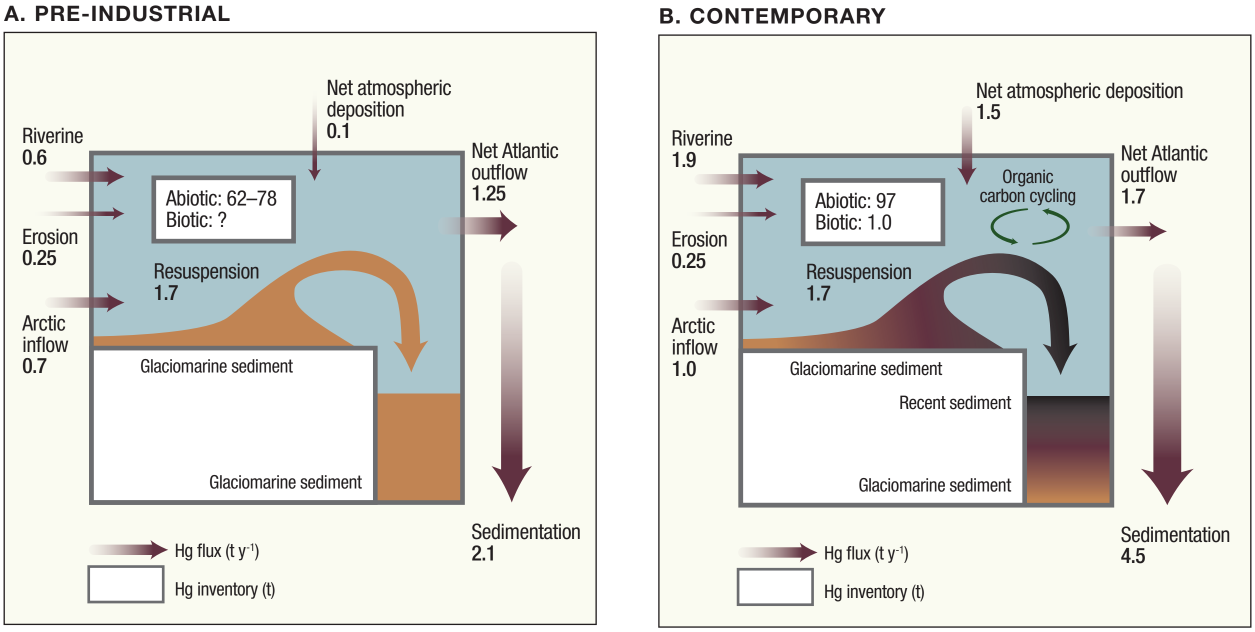

FIGURE 6.8

Pre-industrial and contemporary mass budgets of THg in Hudson Bay. Reprinted with permission from Hare et al. (2008). © Elsevier.

The net Hg influx to and flux out of Hudson Bay in pre-industrial times were both estimated at 3.4 t y^{-1} and appear to have been in a steady state over decades to centuries based on post-glacial sediment cores showing consistent THg concentrations in depths not reached by contemporary inputs (Fig. 6.8; Hare et al. 2008, 2010). Pre-industrial Hg fluxes appear dominated by the resuspension of shallow coastal sediments in the south (< 2 t y^{-1}) and relatively low inputs from rivers and marine water masses (< 1 t y^{-1} each). Primary Hg losses from the system occurred through sedimentation and mass export of marine water (~2 and 1 t y^{-1}, respectively). The net exchange of Hg between the ocean and atmosphere estimated by Hare et al. (2008) was the least-constrained flux because models and field data range widely, but most likely contributed a negligible net amount of Hg during the pre-Industrial Era (0.1 t y^{-1}).

The total contemporary Hg influx and outflux, estimated at 6.3 t y^{-1} each, represent a two-fold increase from pre-industrial fluxes (Fig. 6.8). The most notable change was increased Hg burial in recent sediments (4.5 t y^{-1}), implying that much of the modern Hg load entering this system is buried in the sediments (Hare et al. 2008). Higher river inputs (1.9 t y^{-1}) and a larger net gain from the atmosphere (1.5 t y^{-1}) may reflect increased contemporary atmospheric deposition to Hudson Bay and its watershed. The contemporary THg inventory in the Hudson Bay water column was estimated at 98 t, about 1% of which is present in marine organisms and the remainder in abiotic forms (Hare et al. 2008).

Fluxes of Hg in the Hudson Bay region are susceptible to change with climate warming. The ocean-atmosphere exchange of Hg in Hudson Bay is strongly influenced by sea ice because it prevents Hg evasion from seawater and also intercepts wet and dry precipitation, thereby further exposing the deposited Hg to photoreduction and re-emission to the atmosphere. The large Hg flux from resuspension of shallow coastal sediment is also impacted by sea ice that scours sediments during ice movement and dampens the effect of wave-induced sediment resuspension. Similarly, the transport of watershed Hg to Hudson Bay is strongly influenced by the hydrological regimes of rivers (Kirk and St. Louis 2009), which contribute a larger proportion of Hg to this system than typical of most seas. Given that both sea ice and watershed runoff in Hudson Bay appear particularly sensitive to climate change, magnitudes of Hg fluxes in this region are likely to increase if projected climate models are realized (Gagnon and Gough 2005a, Gough et al. 2004, Gough and Wolfe 2001, Macdonald et al. 2005, Saucier and Dionne 1998). In view of both positive and negative influences of a changing climate on Hg fluxes as well as uncertainties in climate and atmospheric modelling, the net impact on the retention and export of Hg in Hudson Bay is unclear. Elsewhere, absent ice cover or longer ice-free seasons in marine ecosystems are associated with higher net fluxes of Hg from the ocean to the atmosphere (Strode et al. 2007).

While the THg mass balances for Hudson Bay indicate recent human impact, information on MeHg fluxes may be more relevant to concerns of bioaccumulation in food webs. In other oceans, solid-phase MeHg tends to compose a small portion of THg in marine sediment (e.g., < 1%) but a much larger proportion of pore-water THg (e.g., up to 50%) (Fitzgerald et al. 2007). Pore-water fluxes of MeHg from coastal shelf sediments to the overlying water column can be significant enough to explain a large proportion of MeHg burdens in high trophic level organisms (Hammerschmidt et al. 2004, Hammerschmidt and Fitzgerald 2006). Information on MeHg fluxes for Hudson Bay would prove useful in determining the relative contribution of potential sources including coastal shelf sediments, river watersheds, and the Hudson Bay water column. Kirk et al. (2009) examined the importance of MeHg fluxes from two large rivers as sources to Hudson Bay and suggested that internal MeHg production in the bay may be significant. These measurements are also particularly relevant given the potential for increased MeHg loadings from the vast wetlands of the southern Hudson Bay watershed under warming climate conditions (Macdonald et al. 2005).

6.6.3 Comparison of Hg budgets for the Arctic Ocean and Hudson Bay

The estimated flux from atmospheric deposition to the Arctic Ocean suggests it is the largest single source of THg in that system (Table 6.2). In contrast, rivers were identified as the largest single source (41%) for Hudson Bay followed in importance by the atmosphere (32%) and ocean inflow (22%) Hg fluxes (Hare et al. 2008). The difference between the Arctic Ocean and Hudson Bay can be partly attributed to the *edge effect* of riverine inputs which are larger for smaller bodies of water. Modelling versus field measurement approaches to estimate net atmospheric deposition may also explain variation in the relative importance of Hg sources. In contrast with the mass balance for the Arctic Ocean, net atmospheric deposition (1.5 t y^{-1}) to Hudson Bay was estimated from field measurements of atmospheric deposition, snowpack re-emission, and seawater evasion, although the GRAHM model was used to set the maximum upper limit of 13.6 t y^{-1}.

The large difference between GRAHM model results and field measurements of atmospheric inputs in these budgets underscores what is widely regarded as one of the central uncertainties in marine Arctic Hg science—what is the actual net deposition from the atmosphere that accumulates in the water? Recent model estimates of net deposition that incorporate AMDE effects and re-emission of Hg from snowpack following AMDEs range from 110–143 t y^{-1} for the circumpolar Arctic (section 3.6.2). For the Arctic Ocean only, Outridge et al. (2008) calculated an independent estimate of net atmospheric input, based on field measurements of parameters associated with atmospheric flux. This approach, which employed data from direct measurements of wet deposition in the Arctic (Fitzgerald et al. 2005, Sanei et al. 2010), springtime snowmelt at sites across the Arctic, and shipboard-based estimates of evasion from the ocean, produced a net atmospheric Hg deposition to the Arctic Ocean of 8.4 t y^{-1}, over an order of magnitude lower than that of 98 t y^{-1} provided by the GRAHM model (Outridge et al. 2008). The field estimate of net deposition rate that included re-emission but not evasion (18.4 t y^{-1}) was similar to the 23 t y^{-1} calculated for the Arctic Ocean on the basis of snowpack sampling by Lu et al. (2001), and to the 27 ± 7 t y^{-1} calculated from a net atmospheric flux of 2.8 + 0.7 µg m^{-2} y^{-1} to lake sediments in northern Alaska (Fitzgerald et al. 2005). More work using a variety of methodological approaches is essential to resolve the uncertainty in the atmospheric Hg contribution to Arctic ecosystems, especially since this flux should be most immediately affected by emission controls.

6.7 Biogeochemical cycling of Hg

6.7.1 Methylation and demethylation

The main sites of MeHg production in the Canadian Arctic have not yet been identified, although it is known that inorganic Hg is methylated in the marine environment (Fitzgerald et al. 2007). Based on studies from other regions, methylation of inorganic Hg may potentially occur in sediments (particularly in shallow coastal shelves), in the water column or on snow and sea ice (Macdonald and Loseto 2010). The breakdown of Me_2Hg in the water column or in the air after its evasion may also be a source of MeHg for biological uptake (St. Louis et al. 2005, 2007, Kirk et al. 2008). It has been suggested that open polynyas are important sites for Me_2Hg evasion (St. Louis et al. 2005, 2007), bringing up the possibility of Hg methylation by phytoplankton (Pongratz and Heumann 1998). Production of Me_2Hg has been demonstrated for pure cultures of marine bacteria collected from Antarctic surface waters and macroalgae isolated from an Arctic fjord (Pongratz and Heumann 1998, 1999). However, recent work on the Arctic Ocean suggests that Me_2Hg can also be produced in deep waters (section 6.3.1; Kirk et al. 2008). A possible mechanism for phytoplankton-associated methylation was very recently proposed by Larose et al. (2010) implicating transmethylation reactions (Bentley and Chasteen 2002) that are involved in the degradation of the phytoplankton osmolyte dimethylsulfoniopropionate (DMSP). Other studies in lower-latitude oceans have also challenged the current paradigm that only anaerobic conditions support significant MeHg production or net rates of methylation (Mason and Fitzgerald 1990, Sunderland et al. 2009, Heimbürger et al. 2010, Cossa et al. 2011) and underscore the need for more fundamental research examining mechanistic aspects of Hg methylation in polar regions.

TABLE 6.2 Fluxes and relative loadings of THg from major input pathways to the Arctic Ocean (Outridge et al. 2008) and Hudson Bay (Hare et al. 2008). Fluxes are expressed as t y^{-1}; relative loadings (in square brackets) are expressed as percentages of the total inputs. Hudson Bay data are for the modern era (see Fig. 6.8). (source: AMAP 2011)

Pathway	Arctic Ocean	Hudson Bay
Ocean Currents	48 [23%]	1.0 [22%]
Net Atmospheric Deposition	98 [48%]	1.5 [32%]
Rivers	13 [6%]	1.9 [41%]
Coastal Erosion	47 [23%]	0.25 [5%]
Total Inputs (t y^{-1})	206	4.65

The first measurements of methylation rates in marine waters of the Canadian Arctic are consistent with the notion that the water column is an important site for MeHg production (Lehnherr et al. 2011). Seawater samples were amended with isotopically labelled Hg(II) and MeHg and incubated to quantify rates of methylation and demethylation. The results of these experiments demonstrated that both MeHg and Me_2Hg are produced in polar marine waters by methylation of Hg(II). Additionally, Me_2Hg can also be formed from methylation of MeHg, but in general, Me_2Hg production was slower than MeHg production. Methylmercury can be demethylated under both light and dark conditions as a result of photodemethylation and microbial demethylation, respectively.

A numerical model using the rate constants of MeHg production and degradation measured during these incubations estimated that, on average, Hg(II) methylation in the water column accounted for about half (47 ± 62%) of the MeHg present in marine waters of the Canadian Arctic Archipelago and is therefore the single largest source of MeHg to Arctic marine waters and food webs (Lehnherr et al. 2011). Furthermore, the model estimates indicated that MeHg demethylation in the water column limits how far MeHg can be transported by ocean currents. For example, 90% of the MeHg in a particular water mass is likely demethylated in the time it takes for that water to travel 20–200 km. Therefore, highlighting the importance of Hg(II) methylation in the water column, the majority of MeHg occurring in Arctic marine waters probably does not originate from distant sources. The model results also indicated that some sites appear to be net sources of MeHg (methylation hotspots) while others can be net sinks for MeHg, depending on the availability of Hg(II), which limits MeHg production. The implication of this finding is that if concentrations of Hg(II) in Arctic marine waters were to increase as a result of either increased anthropogenic inputs or environmental change, the production of MeHg in the water column from Hg(II) methylation is also likely to increase.

While little information currently exists on photodemethylation in polar seawater, this loss process likely occurs near the water-column surface where UV light can penetrate (e.g., Monperrus et al. 2007). However, recent experimental work suggests that MeHg-chloride complexes, which are the dominant form in seawater, are more resistant to photodemethylation, particularly in comparison to aqueous MeHg complexes in fresh water (Zhang and Hsu-Kim 2010). Sea ice and snow cover strongly reduce light penetration and, hence, photodemethylation in the Arctic Ocean for much of the year. Reductions in summer sea-ice extent associated with climate change will most probably increase photodemethylation in the marine Hg cycle and possibly decrease MeHg levels. Using emerging new techniques, Point et al. (2011) measured the stable isotope composition of Hg in seabird eggs along a latitudinal gradient in Alaska and found isotopic evidence that less photodemethylation occurs at the northern, more ice-covered sites. Photodemethylation of aqueous MeHg produces distinct Hg isotope fractionation patterns (Bergquist and Blum 2007, 2009), and the extent of this fractionation in Hg bioaccumulated by seabirds was significantly correlated to the percent of sea-ice cover. The authors predicted that further loss of sea ice will enhance the breakdown of MeHg in polar marine environments (Point et al. 2011).

6.7.2 Reduction and oxidation

Reduction-oxidation reactions (redox) of Hg can be driven by photochemical or microbial processes, which are described separately as follows. While photochemical reactions appear to dominate the redox cycle in fresh waters, recent evidence from the Arctic suggests microbes may be key players in altering Hg redox chemistry in salt water.

Microbial redox processes

Microbes are well known for their capacity to form the neurotoxin MeHg, but they also have the capacity to detoxify this metal via Hg resistance enzymes (Barkay et al. 2003). Some types of bacteria have a mercuric reductase enzyme (called merA) that reduces Hg(II) to volatile Hg(0), and some also have an enzyme (merB) that decomposes organomercurial species (Barkay et al. 2003). In a study of Arctic microbial communities associated with seawater, microbial mats and macroalgae, Poulain et al. (2007c) found evidence from messenger RNA detection that marine, cold-adapted microbes expressed diverse forms of the merA gene coding for the mercuric reductase enzyme. This discovery of expressed Hg resistance genes suggests that microbial populations in the Arctic marine environment are sensitive to Hg contamination. However, THg concentrations were low in water and sediments from which these microbial populations were sampled and were characteristic of pristine environments. Therefore, the induction of Hg resistance may reflect: 1) the existence of environmental conditions that enhance Hg bioavailability due to microbe physiology or to the physical and chemical properties of the Arctic environment; or, 2) the presence of micro-niches with high Hg concentrations, such as those encountered in snow and frost flowers (Douglas et al. 2008). Investigations in northeastern

Greenland have recently found that mercury-resistant bacteria occur in snow and to a lesser extent in brine associated with Arctic sea ice (Møller et al. 2011).

Poulain et al. (2007c) modelled the potential for microbes to reduce Hg in the High Arctic. Results of their kinetic redox model suggest that microbially mediated reduction of Hg(II) could account for most of the Hg(0) present in the surface waters of the Arctic Ocean. Studies conducted in the circumpolar Arctic show ranges of dissolved gaseous Hg (DGM) concentrations in seawater that broadly overlap with those of freshwater systems (60–643 fM; Table 6.3). Virtually nothing is known about Hg redox chemistry deeper in the ocean water column or in sediments. Elevated GEM concentrations were measured in deep ocean water beyond light penetration (section 6.3.1), suggesting that microbes may be important mediators of Hg redox transformations in that environment. The production of GEM in seawater collected from the aphotic zone and incubated in the absence of light supports this notion (Lehnherr et al. 2011). In addition, elevated levels of DGM were recorded under sea ice in the Canadian Arctic Ocean (St. Louis et al. 2007, Andersson et al. 2008), suggesting that DGM builds up under sea ice. This DGM is likely composed of a mixture of Me_2Hg and Hg(0), both of which are volatile and may be of microbial origin. The rate of microbial metabolism is dictated by temperature, and the Arctic is undergoing climate warming. Therefore, it is critical that further research be conducted at the cellular level on microbial contributions to Hg transformations in the polar marine environment. Investigations should focus on microbial species currently adapted to the Arctic and species likely to arise and become dominant with climate change.

Photochemical redox processes

Halogens such as chlorine and bromine and their reactive derivatives are involved in photooxidation of Hg(0) to Hg(II). This process has been documented in the atmosphere (Ariya et al. 2008), in snow (Dommergue et al. 2003, Lalonde et al. 2003, Poulain et al. 2004, Steffen et al. 2008) as well as in surface waters of the St. Lawrence River (Lalonde et al. 2001) and the coastal Atlantic Ocean (Whalin and Mason 2006). In the presence of light, elemental Hg is more likely to be oxidized back to divalent Hg in marine systems than in fresh waters. During bottle incubation experiments conducted with coastal waters near Cornwallis Island, Poulain et al. (2007b) showed that the light-induced production of elemental Hg greatly decreased with increasing salinity, reaching virtually no net production in undiluted coastal seawater. Altogether these data suggest that conditions encountered in coastal and marine systems tend to increase the preponderance of inorganic Hg(II) by favouring oxidation reactions.

TABLE 6.3 Water concentrations of DGM in surface waters of Arctic marine ecosystems.

Location	Sampling Date	*n*	DGM Concentration (fM)	Reference
North Atlantic and Arctic oceans (near the Fram Strait)	2004	> 400	54–174	Temme et al. (2005)
Arctic Ocean (Griffith Island and Resolute Passage, NU)	2004–2005	8	643 ± 179[a]	St. Louis et al. (2007)
Canadian Arctic Archipelago[b]	2005	11	127 ± 51[a]	Kirk et al. (2008)
Hudson Bay region[b]	2005	13	148 ± 25[a]	Kirk et al. (2008)
Arctic Ocean (including the Canadian Archipelago, Beaufort Sea, Chukchi Sea and Central Basin)	2005	Continuous measures[c]	220 ± 110	Andersson et al. (2008)
Arctic Ocean (near Kongsfjorden, Norway)	2002	5	60–349	Sommar et al. (2007)

[a] GEM measurements.

[b] only surface water data are presented here.

[c] i.e. every 10 minutes over a 3-month period.

n = number of sites sampled, fM = femtomoles L^{-1} (i.e. 10^{-15} M).

6.8 Food webs

6.8.1 Trophic transfer of Hg through marine food webs in the Canadian Arctic

The first investigations of Hg biomagnification in marine food webs were reported for the Canadian Arctic in the previous *CACAR* (Fisk et al. 2003). Two separate studies that focused on Lancaster Sound (Atwell et al. 1998) and the North Water Polynya (Campbell et al. 2005) indicated that Hg concentrations increase several-fold from ice algae (North Water Polynya) and particulate organic matter (Lancaster Sound) up through invertebrates and fish to seabirds and marine mammals. More recently, Hg biomagnification in marine food webs was investigated in Cumberland Sound, in Hudson Bay, and in the Beaufort Sea and Amundsen Gulf.

Cumberland Sound

In Cumberland Sound, located on the east coast of Baffin Island, THg concentrations in biota (log-transformed, µg g^{-1} wet weight [ww]) were positively correlated with trophic position, estimated by their $\delta^{15}N$ ratios (Fig. 6.9). Muscle tissue was analyzed for all species except mixed zooplankton, which were analyzed whole. Concentrations increased from invertebrates (clams and zooplankton) to mid-trophic position consumers like seal and sculpin, and the top predator, Greenland shark. However, ringed and harp seals were at a similar trophic position to capelin and sculpin, but had higher THg concentrations (Fig. 6.9), which could be attributed to differences in bioaccumulation processes between these biota or differences in sources of dietary Hg. Ringed seals from Cumberland Sound (0.249 ± 0.198 µg g^{-1} ww) also had higher THg than conspecifics from West Greenland waters (Rigét et al. 2007b). Greenland sharks sampled in Cumberland Sound in August 2007 had similar muscle THg concentrations to Cumberland sharks collected in 2008 and 2009 (McMeans and Fisk 2010).

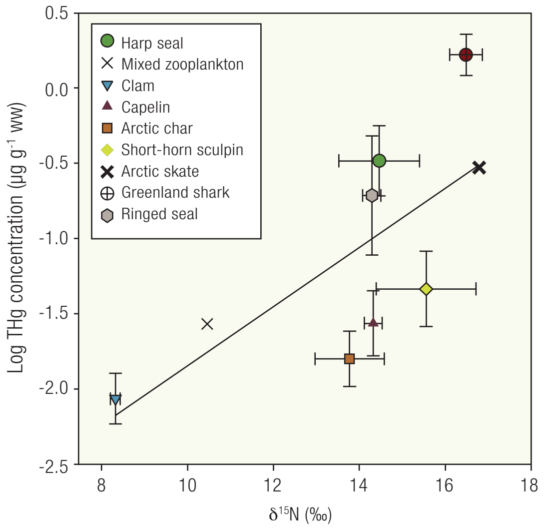

FIGURE 6.9

Relationship between $\delta^{15}N$ ratios (‰) and log-transformed THg concentrations (µg g^{-1} ww) in species sampled in August 2007 in Cumberland Sound, Baffin Island (McMeans and Fisk 2010). The linear regression model for this relationship was: $\log THg = -3.69 + 0.193 * \delta^{15}N$ ($r^2 = 0.48$, $p < 0.001$, $n = 9$).

Hudson Bay

Mercury biomagnification was investigated in the marine food web of Hudson Bay using muscle data for three species of seals sampled from western Hudson Bay (Young et al. 2010) and one seabird species—the thick-billed murre—sampled from Coats Island in northern Hudson Bay, as well as whole fish and invertebrates delivered by thick-billed murres to nestlings on the breeding ledges on Coats Island, supplemented by invertebrates sampled from plankton tows near the Nuvuk Islands. A positive relationship was found between log THg concentrations and $\delta^{15}N$ ratios in the biota sampled from Hudson Bay (Fig. 6.10). Concentrations of THg increased from invertebrates through fish to the top trophic consumers, seabirds and seals. The mean muscle $\delta^{15}N$ ratio and THg concentration for thick-billed murres were very similar to that found in muscle of adult ringed seals.

Beaufort Sea and Amundsen Gulf

Mercury biomagnification was evaluated for food webs in different habitats of the eastern Beaufort Sea and Amundsen Gulf, specifically a shallow estuarine shelf, deep pelagic water, and epibenthic zones. Biomagnification rates were estimated using the slopes of THg concentration versus trophic position of sampled invertebrates, fish, and beluga (Loseto et al. 2008a). The biomagnification slopes for THg ranged from 0.23 to 0.26 and did not differ significantly between the estuarine, pelagic, and epibenthic food webs. The biomagnification slopes were similar to the value calculated for Hudson Bay (slope of 0.23) and slightly higher yet comparable to values calculated in Cumberland Sound (slope of 0.193), the North Water Polynya near Baffin Bay (slope of 0.197; Campbell et al. 2005), and West Greenland waters (slope of 0.183; Rigét et al. 2007b). Thus, similar Hg biomagnification slopes have been observed among disparate Arctic marine habitats (Rigét et al. 2007b).

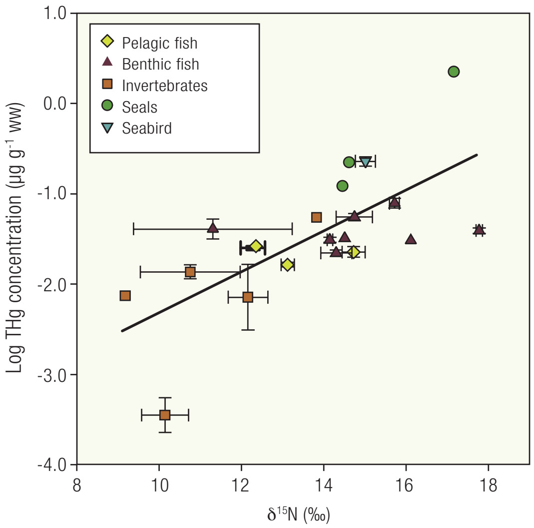

FIGURE 6.10

Relationship between δ^{15}N ratios (‰) and log-transformed THg concentrations (µg g^{-1} ww) in a marine food web of Hudson Bay (linear regression model: log THg = $-4.57 + 0.225 \times \delta^{15}\text{N}$, $r^2 = 0.47$, $p < 0.001$, $n = 20$). Invertebrates (sea angel, jellyfish, squid, amphipods, euphausiids), pelagic fish (Arctic cod, capelin, sandlance), and benthic fish (sculpins, gunnels, Arctic and daubed shannies, eelpout, Atlantic poacher, blennies, snailfish) were sampled from 2007 to 2009. Seabirds (thick-billed murre) were sampled in 2007. Ringed seals were sampled in 2006, bearded seals in 2005, and harbour seals from 1999 to 2006. Fish, invertebrate, and seabird data are from Braune (2010b). Seal data are from Young et al. (2010).

6.8.2 Mercury exposure, habitat and feeding behaviour of beluga whales in the eastern Beaufort Sea

Representing one of the largest summer aggregations of beluga whales, beluga from the eastern Beaufort Sea population arrive at the Mackenzie estuary by the thousands (Fraker et al. 1979). The summer harvest of beluga by communities of the Inuvialuit Settlement Region (Northwest Territories) is an important component of the Inuvialuit subsistence lifestyle (Usher 2002). The Beaufort beluga whale population is one of Canada's largest and is estimated at a minimum of 40,000 individuals (COSEWIC 2004). At present, there is no indication of population decline.

Travelling to the Bering Sea in the winter and to the eastern Beaufort Sea and Amundsen Gulf in the summer, the Beaufort beluga population undergoes one of the longest migrations (Richard et al. 2001). As a result, there is high seasonal and regional complexity in diet and dietary sources of Hg.

Concern over high Hg levels found in Beaufort Sea beluga whales (Lockhart et al. 2005b) led to several studies to better understand sources of Hg and their exposure as top predators in the marine environment. A conceptual model of Hg accumulation in beluga was developed by categorizing *top-down* and *bottom-up* processes of Hg exposure (Fig. 6.11). Top-down investigations of beluga habitat use were conducted to gain knowledge of feeding areas followed by studies to understand diet preferences, while bottom-up studies on local food webs (i.e. trophic structure and sources of Hg) were carried out to better understand exposure (Loseto et al. 2006, 2007, 2008a, 2008b, 2009).

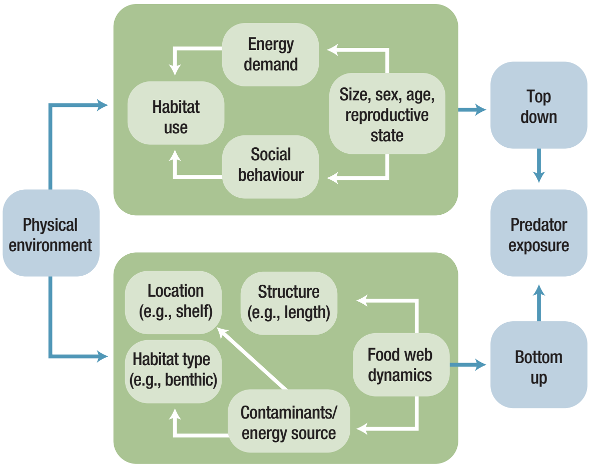

FIGURE 6.11

Conceptual model of processes affecting the Hg concentration in a predator species such as beluga whales. Food web interactions determine the sources, bioaccumulation, and biomagnification of Hg in the form of MeHg. Dietary preferences are shaped by many factors including the sex, size, age, and reproductive status of an animal. These, in turn, dictate the energy requirements and social behaviour that together describe habitat use (source: AMAP 2011).

BOX 6.1 Community-Based Monitoring in the Inuvialuit Settlement Region: Hendrickson Island Beluga Study (Northwest Territories)

The communities of the Inuvialuit Settlement Region have participated in research and monitoring of their environment and resources since the 1970s, in part due to their land claim settlement—the *Inuvialuit Final Agreement*—and oil and gas exploration activities in the region. The traditional subsistence hunt for beluga whales has included sample collection for contaminants since the 1980s, which has resulted in one of the longest datasets for an Arctic marine mammal.

Among the many monitoring programs within the region, the beluga program at Hendrickson Island, just 20 km from Tuktoyaktuk, has been a great success and is now a model for other community-based monitoring programs. The success of the program has been attributed to the strong relationships formed between Frank Pokiak and his family and the researchers involved in the beluga program.

Just over ten years ago, Frank Pokiak was hired and trained to take beluga tissue samples from harvested whales. Despite having to move their family to the island each year, Frank and his wife Nellie were passionate about the research program and remained dedicated to maintaining the program annually. By bringing their family, they are able to pass on traditional knowledge and teach their children and grandchildren how to sample whales. As well, more than ten community youths have been engaged in the program over the past six years. The stable, long-term involvement of the Pokiaks in the Hendrickson Island beluga research program has provided a solid foundation to grow from and has attracted new programs.

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Having Frank and Nellie Pokiak lead the monitoring and sampling components makes great sense. Local hunters know them and are therefore comfortable sharing their whales to be sampled. Hunters bring their whales to the island to butcher, and Frank assists them in bringing in the whales. Because the Pokiaks and other local hunters have a long history of observing whales, their movements, behaviour and anatomy, they add to the strength of the program and help shape the research direction.

Key elements that have led to the great success of the Hendrickson Island program include: 1) respect for each other's knowledge and acknowledgement of each other's roles in maintaining a successful program; 2) sharing knowledge at meetings that has fostered strong communication and relationship building; and, 3) a common interest in studying and learning more about the environment to enable better decision making and ensure a viable future for beluga whales.

The habitat selected by beluga was characterized using resource selection function analysis (Arthur et al. 1996) to evaluate top-down influences on their Hg exposure. Satellite telemetry data from tagged beluga provided the information to evaluate daily habitat selection defined by sea ice, bathymetry, and distance to the shoreline. Beaufort Sea beluga were found to sexually segregate during summer movements (Loseto et al. 2006). Based on sea-ice concentration and shelf habitat, three different habitat use groups were defined in relation to beluga length, sex, and reproductive status as follows: 1) females with and without calves and small males (< 4 m) selected shallow open water near the mainland; 2) medium length males (3.8–4.3 m) and a few females (> 3.4 m) without neonates selected the sea ice edge; and, 3) the largest males (4–4.6 m) selected heavy sea-ice concentrations in deep, offshore waters. It was hypothesized that such intra-species segregation of habitat use has consequences for their feeding ecology and Hg exposure. A habitat segregation model was therefore used to evaluate potential Hg sources and food web biomagnification to different sex and size classes of beluga whales (Loseto et al. 2008a).

Hypothetical feeding groups were developed to merge the influences of predator behaviour and food web dynamics on Hg levels in beluga. The following food webs were grouped to beluga habitat use: 1) beluga using shallow, open-water habitats likely use the Mackenzie Delta extensively and were therefore paired with the estuarine-shelf food web; 2) beluga selecting

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the ice edge likely feed on Arctic cod associated with sea ice (Gradinger and Bluhm 2004) and/or in pelagic food webs, and were paired with the Amundsen Gulf food web that included organisms collected offshore in the pelagic environment; and, 3) beluga selecting high levels of sea ice concentration do so in deep waters where they dive up to 800 m (Richard et al. 1997), likely feeding in benthic habitats, and were paired with the epibenthic food web.

Despite the high influx of inorganic Hg and MeHg to the Mackenzie River Delta (Leitch et al. 2007), prey Hg levels associated with the estuarine-shelf food web were among the lowest (Fig. 6.12). Concentrations of THg in fish muscle ranged from 0.12 µg g^{-1} dw in Pacific herring to 0.31 µg g^{-1} dw in saffron cod. The beluga whale group hypothesized to feed there also had the lowest THg levels (muscle: 2.6 µg g^{-1} dw) (Loseto et al. 2008a). Beluga hypothesized to feed in the epibenthic and Amundsen Gulf food webs had the highest THg concentrations and did not differ significantly (muscle: 6.0 and 4.4 µg g^{-1} dw, respectively). Potential prey from the offshore and benthic habitats had THg concentrations that were generally higher than prey from the estuarine shelf, including Arctic cod found in both areas (Fig. 6.12), which was consistent with the differences in THg between nearshore and offshore beluga. Among the benthic fish species, a range in THg concentrations was observed, and fourhorn sculpin had the highest levels (0.53 µg g^{-1} dw). Although the assigned beluga feeding groups were an oversimplification of temporal and spatial complexities of beluga movement and seasonal feeding preferences, the exercise was valuable in understanding trophic transfer in the various food webs.

Fatty acids were measured on prey items collected in the near shore, offshore pelagic, and epibenthic food webs to trace diet sources of beluga. Overall, it appeared that beluga diet was dominated by Arctic cod (Loseto et al. 2009). However, size-related differences in diet were evident, and larger-sized beluga appeared to consume more offshore Arctic cod, whereas smaller-sized beluga appeared to feed on prey in near-shore habitats that included nearshore Arctic cod. The fatty acid results also revealed the lack of benthic sources in the diet of beluga whales.

Both bottom-up and top-down processes explained the observed Hg concentrations in beluga, which varied within the population due to size and sex segregation. These findings highlight the range and variability of prey resources consumed by Arctic marine mammals (Cardona-Marek et al. 2009). It remains unclear why

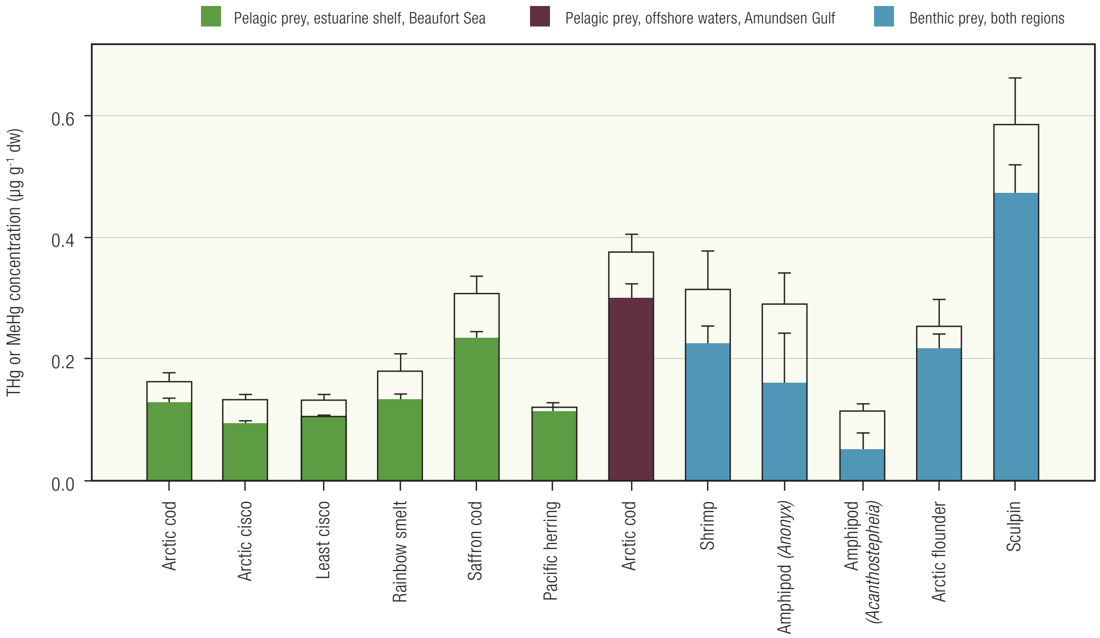

FIGURE 6.12

Concentrations of THg and MeHg (µg g^{-1} dw) (white and coloured bars, respectively) in potential prey species collected from different habitats of the Beaufort Sea and Amundsen Gulf. Pelagic and benthic prey were collected from the shallow estuarine shelf of the Beaufort Sea and offshore waters in the Amundsen Gulf. Muscle was analyzed for fish while invertebrates were analyzed whole. Data are from Loseto et al. (2008a).

there was unexpectedly greater Hg bioaccumulation in beluga and their associated food web in offshore waters compared to those feeding in the estuarine shelf. This observation suggests the large Hg loadings from the Mackenzie River may not be bioavailable upon entering the Delta. Instead, this Hg may be deposited and redistributed, perhaps becoming more readily available to the offshore food web.

6.9 Marine fish

6.9.1 Sea-run Arctic char

Spatial patterns of Hg concentrations

Arctic char is a member of the salmonid family, which also includes lake trout and Dolly Varden, and has the most northern distribution of the three species. The taxonomic differentiation of Dolly Varden and Arctic char has long been debated, and for the purposes of this assessment, Dolly Varden was considered to have a limited distribution within the Canadian Arctic and occur primarily west of the Mackenzie River Delta (Reist and Sawatzky 2010). Arctic char and Dolly Varden are freshwater species that either reside entirely in lakes and rivers or are anadromous and migrate to the ocean for several weeks during the Arctic summer to feed on marine invertebrates and small fish. This migratory behaviour facilitates greater accessibility to food and may result in a more rapid growth rate than would be observed in inland waters where invertebrates are smaller and forage fish are often scarce. Sea-run Arctic char is an important traditional food harvested by coastal Arctic communities for domestic consumption and, in some areas (e.g., Cambridge Bay), in support of commercial fisheries. Sea-run char is the dominant species captured in marine fisheries in the Arctic Archipelago (Zeller et al. 2011).

The earliest information on Hg levels in sea-run char is from the periodic assessments of commercial and domestic fisheries, determined primarily over the past three decades and summarized in Lockhart et al. (2005a). Sample sizes from these assessments were small, and ancillary measurements were often limited to length and weight. Levels of Hg were measured in sea-run Arctic char collected from northern Quebec and Labrador in the late 1990s (Evans et al. 2005) with additional measurements such as fish age also determined.

During NCP III, a systematic scientific survey was made of THg concentrations in sea-run char at 20 locations across northern Canada including the western Arctic (e.g., Holman Island, Paulatuk), central Arctic (e.g., Cambridge Bay, Gjoa Haven), northern Baffin Island area (e.g., Resolute Bay, Pond Inlet), and Labrador (e.g., Nain, Hopedale) (Evans and Muir 2005, 2006, 2007, 2008, 2009, 2010). Two of those sites were west of the Mackenzie River on the Rat and Vittrekwa rivers where Hg levels were measured in Dolly Varden char. Sampling sites were selected to provide broad spatial coverage of regions under different watershed influences, climatic regimes, and latitudinal gradients. Some sites were sampled more than once to investigate temporal variability and, for a subset of sites, to track longer-term temporal trends as part of NCP's monitoring program.

Average fork length in fish ranged from 408 ± 10 mm at Nain to 732 ± 17 mm at Gjoa Haven (Fig. 6.13). Char muscle had extremely low concentrations of THg across the Canadian Arctic, with site averages ranging from 0.014 ± 0.002 μg g^{-1} ww at Cape Dorset to 0.097 ± 0.015 μg g^{-1} for the Vittrekwa River. Site-mean concentrations of THg in char were not significantly correlated with their mean length, age, $\delta^{13}C$ ratios or $\delta^{15}N$ ratios. Site-specific differences in Hg in the sea-run char most likely reflect variation in the Hg content of their diets, which may be related to localized inputs of Hg from large watersheds (e.g., the Mackenzie River), Hg methylation rates in the marine environment, and the types of prey available.

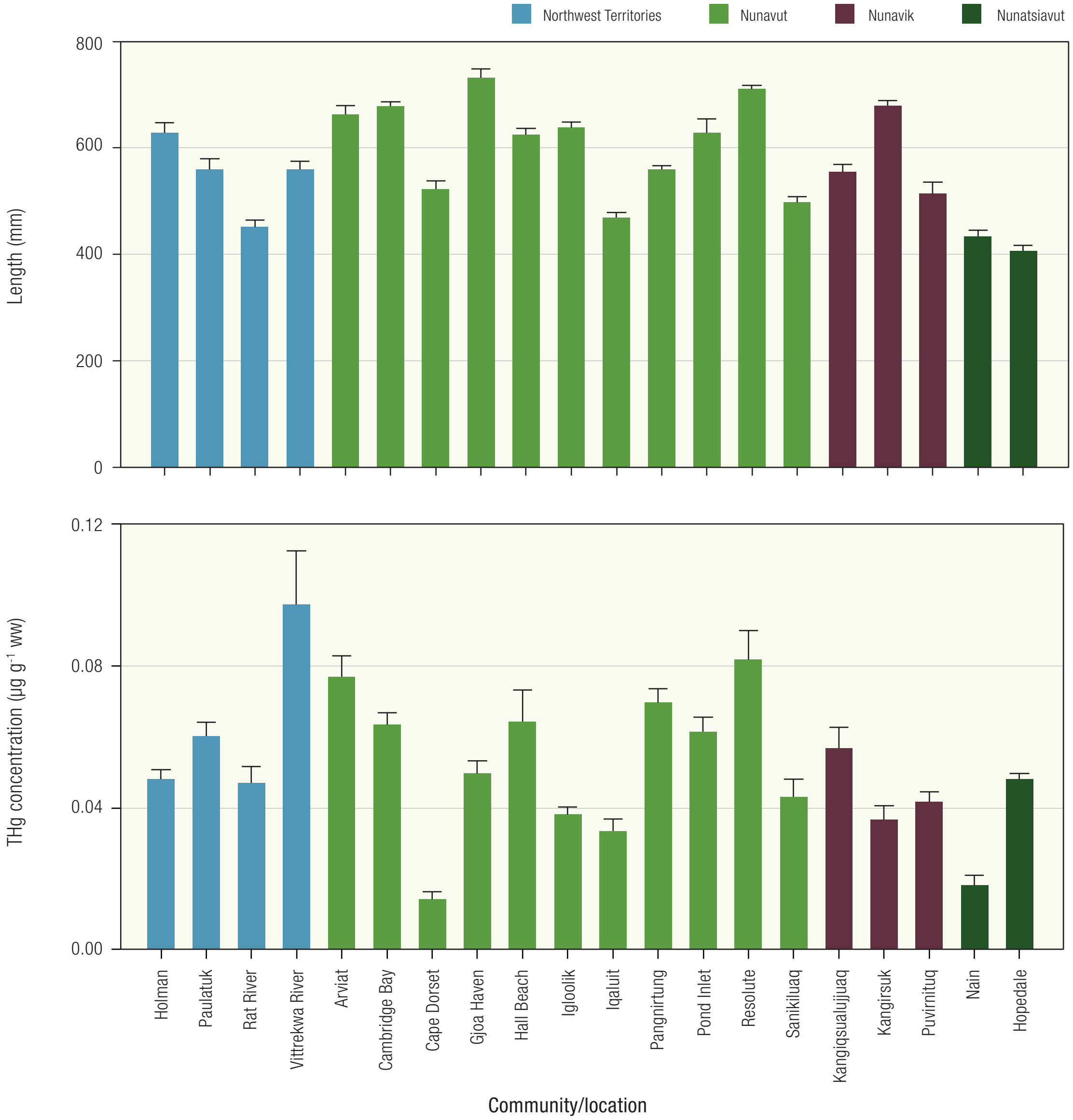

FIGURE 6.13

Average fork length and THg concentrations in sea-run Arctic char or Dolly Varden char (Rat and Vittrekwa rivers only) collected from 2004 to 2009 at 20 locations (Evans and Muir 2005, 2006, 2007, 2008, 2009, 2010).

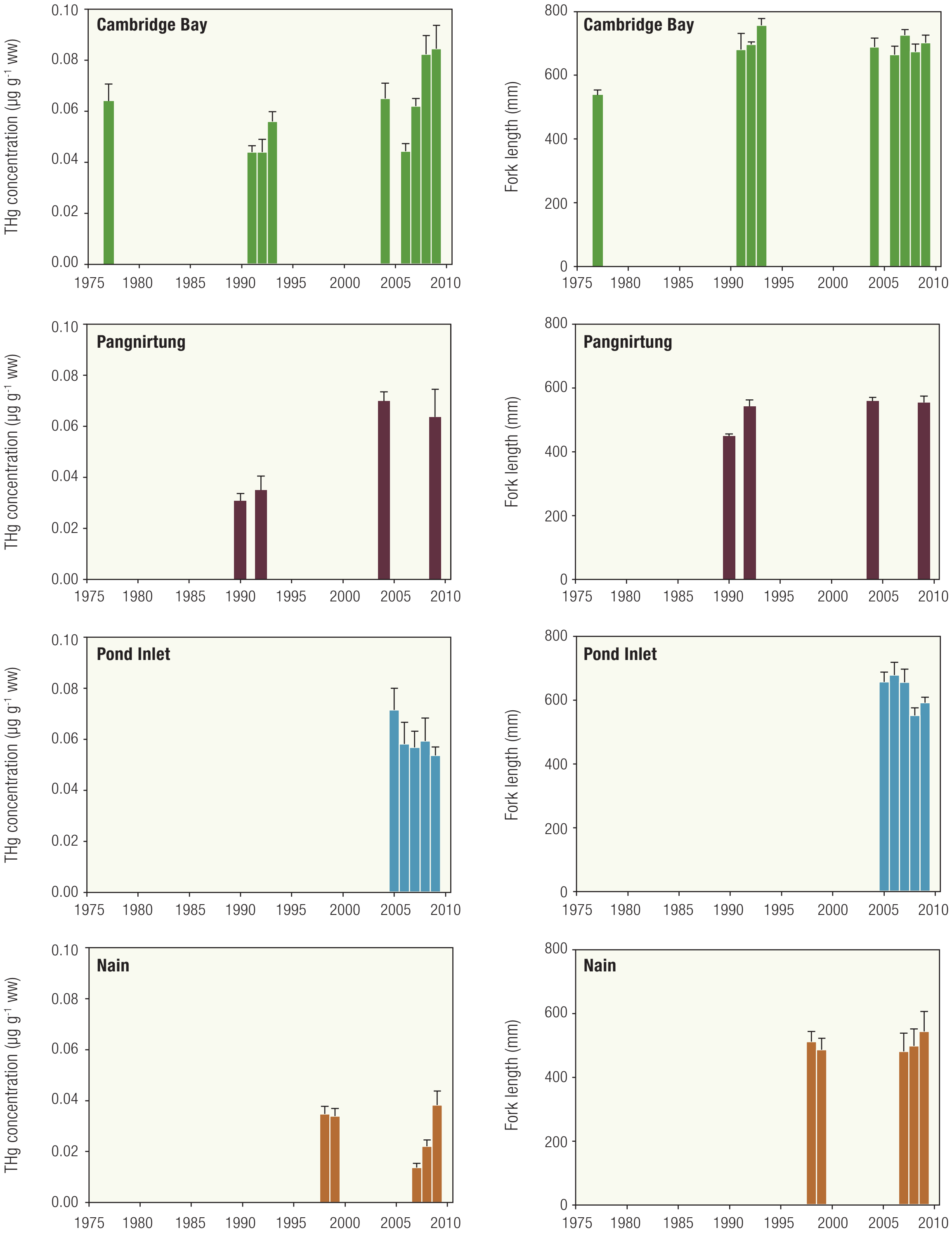

FIGURE 6.14

Temporal variation in the mean (± standard deviation) fork length and THg concentration in sea-run char from Cambridge Bay, Pangnirtung, Pond Inlet, and Nain (data from Lockhart et al. 2005a, Evans and Muir 2010). Statistical results are described in the text.

Temporal trends of Hg in sea-run char

Sea-run char were measured in the Canadian Arctic for their Hg content as early as the 1970s (Lockhart et al. 2005a) but with relatively few measurements and infrequent sampling, the sensitivity of these datasets to detect temporal trends was limited. The sea-run char program, initiated during NCP III, is better positioned to track trends with larger and consistent sample sizes and with more detailed ancillary measurements such as age, and carbon and nitrogen stable isotopes. From 2004 to 2009, sea-run char were collected at coastal communities and some communities were monitored more than once (Evans and Muir 2006, 2010). These collections, combined with earlier studies, allow for an investigation of temporal trends for populations at Cambridge Bay, Pangnirtung, Pond Inlet, and Nain.

Cambridge Bay has the longest record of THg concentrations in muscle of sea-run char, where fish were collected between 1977 and 2009 (Fig. 6.14). Fish collected in 1977 were much smaller than in other years and so were excluded from the trend analysis. The factors affecting log-transformed THg concentrations were explored with length, year, and a length-by-year interaction as independent variables in a general linear model. Cambridge Bay char showed a significant increase ($p < 0.001$) in THg concentrations from 1991 to 2009 and 2004 to 2009 ($p = 0.006$) as did Pangnirtung from 1990 to 2009 ($p < 0.001$). Pond Inlet, north of Pangnirtung, was investigated annually from 2004 to 2009, and variations in THg concentrations were only related to fish length. Finally, THg was measured in char from Nain in 1998 and 1999 and again from 2007 to 2009. Log THg concentrations showed a significant decrease ($p = 0.003$) for the entire dataset of 1998 to 2009 but a significant increase ($p < 0.001$) in the most recent years from 2007 to 2009. Since a limited number of years were sampled, the results should be interpreted with caution. However, the evidence suggests a trend of Hg increase in recent years with the exception of the most northerly sampling location, Pond Inlet.

Less detailed data exist for eight other communities, where char were sampled on only two or three occasions (Table 6.4). Muscle concentrations of THg (log-transformed) were not significantly different between years at Holman, Arviat, Hall Beach, and

TABLE 6.4 Mean (± standard deviation) fork length and THg concentration in sea-run char collected in two or three years (data from Lockhart et al. 2005a, Evans and Muir 2010).

Region	Community	Year	Fork Length (mm)	THg Concentration ($\mu g\ g^{-1}$ ww)
Northwest Territories	Holman Island	2004	631.5 ± 88.3	0.048 ± 0.008
		2008	520.2 ± 38.2	0.046 ± 0.019
		2009	656.3 ± 142.0	0.074 ± 0.045
	Paulatuk	1984	546.7 ± 58.8	0.042 ± 0.12
		2004	569.1 ± 42.8	0.064 ± 0.014
		2007*	501.0 ± 34.7	0.052 ± 0.016
Nunavut	Arviat	2005	680.4 ± 115.8	0.071± 0.026
		2008	657.5 ± 96.5	0.083 ± 0.027
	Hall Beach	1992	698.0 ± 56.3	0.054 ± 0.018
		2007	626.4 ± 111.9	0.064 ± 0.031
	Iqaluit	1991	302.0 ± 36.5	0.082 ± 0.019
		2004	470.9 ± 45.0	0.034 ± 0.011
		2009	522.8 ± 62.2	0.033 ± 0.015
Nunavik	Kangiqsualujjuaq	2006	567.0 ± 56.0	0.068 ± 0.044
		2007	544.0 ± 49.9	0.050 ± 0.025
		2008	549.0 ± 63.0	0.042 ± 0.022
	Kangirsuk	1998	460.0 ± 63.8	0.032 ± 0.026
		2007	682.5 ± 46.0	0.037 ± 0.012
Nunatsiavut	Hopedale	1999	415.8 ± 106.4	0.030 ± 0.010
		2007	422.1 ± 32.4	0.048 ± 0.009

* For Paulatuk, two fish with high THg levels were excluded from the analysis because they were potentially non-migratory

Kangiqsualujjuaq, while significant differences were observed for char from Kangirsuk, Hopedale, Iqaluit, and Paulatuk. Increasing temporal trends were found at all the latter sites except for Iqaluit. Trend analyses for these eight communities were based on infrequent sampling and should be interpreted with caution until more complete analyses are conducted.

6.9.2 Other marine fish

With the exception of sea-run Arctic char, there is currently very limited information on Hg concentrations in marine fish in the Canadian Arctic. Char spend most of their life in fresh water and only seasonally migrate to the ocean to feed. Little information exists for wholly marine fish species that may be important sources of dietary Hg for seabirds and large marine mammals such as seals and beluga. Arctic cod, a keystone species, has been sampled in several food web studies for the Beaufort Sea and Amundsen Gulf (Loseto et al. 2008a), the central basin of the Arctic Ocean (Stern and Macdonald 2005), and Lancaster Sound (Atwell et al. 1998). Arctic cod captured in offshore waters had higher THg concentrations than those in an estuarine shelf (Loseto et al. 2008a), and spatial variation in cod THg was also observed between deep basins in the Arctic Ocean (Stern and Macdonald 2005). In addition, several species of deep-water demersal fish were found to have higher THg concentrations relative to fish in an estuarine shelf (Loseto et al. 2008a). The factors that determine geographic and habitat variation of Hg bioaccumulation in marine fish remain unresolved and warrant future investigation to better understand Hg transfer to top predator animals in the marine environment.

6.10 Marine birds

6.10.1 Levels and spatial patterns

Eggs of thick-billed murres, northern fulmars, black-legged kittiwakes, black guillemots, and glaucous gulls were sampled from Prince Leopold Island in Lancaster Sound from 2003 to 2004 and again in 2008. Concentrations of THg in eggs varied significantly among the five seabird species sampled, with glaucous gull eggs having the highest concentrations and those of black-legged kittiwakes, the lowest (Fig. 6.15). Differences in egg THg concentrations among the species at Prince Leopold Island in 2008 were similar to those reported by Campbell et al. (2005) for birds collected in 1998 from the North Water Polynya. For those five species, Campbell et al. (2005) also found that the kittiwakes had the lowest THg concentrations but both northern fulmars and glaucous gulls had relatively high levels.

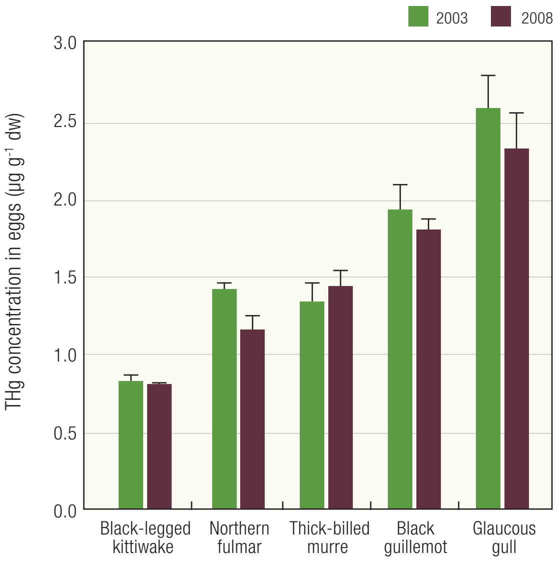

FIGURE 6.15

Mean concentrations (± standard error) of THg (µg g^{-1} dw) in eggs of five seabird species collected from Prince Leopold Island from 2003 to 2004 and again in 2008 (Braune 2009c).

Those authors attributed the differences to trophic position with the glaucous gulls feeding almost a full trophic level higher than the kittiwakes. At Prince Leopold Island, glaucous gulls feed their chicks mainly marine prey including fish and the eggs and nestlings of other seabirds (Nettleship et al. 1990), whereas breeding kittiwakes feed primarily on small surface-schooling fish as well as crustaceans and plankton at the surface (Baird 1994). However, many Arctic seabirds overwinter in areas far from their breeding colonies. Therefore, differences in diet as well as overwintering areas may affect their exposure to contaminants.

Eggs of Arctic terns sampled in 2008 from a colony just north of Cornwallis Island in the High Arctic (Akearok et al. 2010) had a mean THg concentration (2.1 ± 0.13 µg g^{-1} dw) very similar to those found in some glaucous gull and black guillemot eggs (Annex Table A2.5). However, THg concentrations (mean of 6.4 ± 5.17 µg g^{-1} dw) in eggs of ivory gulls from Seymour Island in the Canadian High Arctic in 2004 were among the highest ever recorded in Arctic seabird eggs (Braune et al. 2006). In contrast, Campbell et al. (2005) found that ivory gulls had lower THg concentrations than glaucous gulls collected from the North Water Polynya in 1998.

The last spatial survey of Hg levels in Canadian Arctic seabird eggs was carried out in 1993 (Braune et al. 2002). That survey included eggs of thick-billed murres from four Arctic locations: Prince Leopold Island and Coburg Island in the High Arctic, and Digges Island and Coats Island in northern Hudson Bay. The survey showed that concentrations of THg were significantly higher in murre eggs from the High Arctic sites—Prince Leopold Island, Coburg Island—compared with the lower Arctic murre colonies sampled—Coats Island, Digges Island—and there were no significant differences found for THg in eggs between the two Low Arctic colonies.

In 2007 and 2008, livers of adult thick-billed murres were sampled from five locations in the eastern Canadian Arctic (Fig. 6.16). A comparison of female hepatic THg concentrations from those five colonies found no significant differences among the three Low Arctic colonies—Coats Island, Digges Island, and Akpatok Island—in the northern Hudson Bay and Hudson Strait region, nor were there significant differences between the two higher latitude colonies at Prince Leopold Island and the Minarets (Fig. 6.17). However, hepatic THg concentrations in murres from Prince Leopold Island and the Minarets were significantly higher than from the three Hudson Strait/Hudson Bay colonies.

FIGURE 6.16

Five sampling locations for adult thick-billed murres collected in 2007 and 2008. Only females were collected at Coats Island.

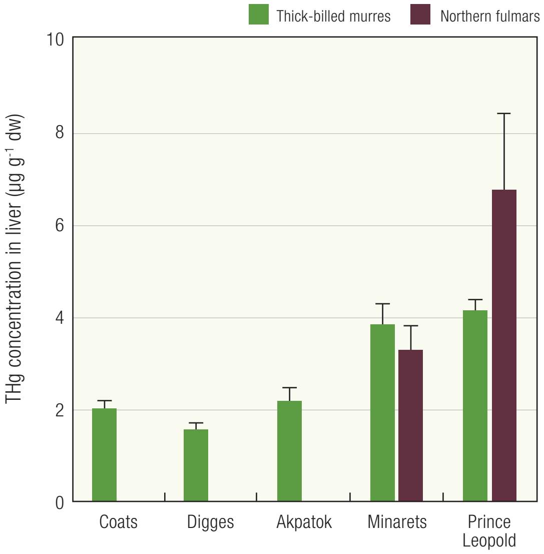

FIGURE 6.17

Mean concentrations (± standard error) of THg (µg g^{-1} dw) in livers of adult female thick-billed murres and northern fulmars collected from five colonies in 2008 (*n* = 5 for each species at each of the five colonies) (Braune 2009b).

Hepatic THg levels in female northern fulmars from the Minarets and Prince Leopold Island—the only two locations sampled for fulmars in 2008—were not significantly different although the birds from Prince Leopold Island had relatively higher Hg levels (Fig. 6.17). However, in 2003, both male and female northern fulmars sampled from Cape Vera on Devon Island had relatively higher hepatic THg concentrations (males: 11.6 ± 0.77 µg g^{-1} dw, females: 9.54 ± 1.08 µg g^{-1} dw) than those from Prince Leopold Island (males: 9.80 ± 0.98 µg g^{-1} dw, females: 7.31 ± 1.23 µg g^{-1} dw) (Braune et al. 2010). Hepatic THg concentrations in two fulmars sampled in 2001 from the Qikiqtarjuaq area (Mallory et al. 2004b) were much lower (0.8 and 0.9 µg g^{-1} dw) than levels found in fulmars from the nearby Minarets in 2008, and from Cape Vera and Prince Leopold Island in 2003 (see Braune et al. 2010).

The results from this recent spatial survey suggest there are latitudinal differences in the factors determining Hg exposure, with birds breeding at the higher latitude colonies accumulating more Hg. Similar latitudinal differences have been observed by Dietz et al. (1996) for Greenland species and Braune et al. (2002) in the Canadian Arctic. Akearok et al. (2010) also found that THg concentrations in common eider eggs were higher from a High Arctic colony just north of Cornwallis Island compared with eider eggs sampled from the sub-Arctic Southampton Island in 2008. However, Mallory et al. (2004a) found the opposite pattern for common eiders, with higher hepatic THg concentrations in eiders from Hudson Bay locations relative to birds sampled elsewhere throughout the Canadian Arctic. No spatial pattern was evident for hepatic concentrations of THg in long-tailed ducks sampled from nine locations throughout the Canadian Arctic (Braune et al. 2005).

6.10.2 Temporal trends

Seabird eggs from Prince Leopold Island on Lancaster Sound have been monitored for contaminants, including THg, since 1975 (Braune 2007). Interpretation of temporal trends of contaminants may be confounded if populations vary their diet over trophic levels through time (Hebert et al. 2000). Therefore, regression analyses, which were used to analyze temporal trends of THg in the seabird eggs, included $\delta^{15}N$ as a covariable to control for potential inter-year trophic variations in diet. Significant increases in THg concentrations occurred between 1975 and 2008 to 2009 in eggs of thick-billed murres ($n = 55$, $p < 0.001$), northern fulmars ($n = 56$, $p < 0.001$) and black-legged kittiwakes ($n = 26$, $p < 0.03$) (Fig. 6.18). However, since 2003, THg concentrations in the fulmars, and possibly the kittiwakes, no longer appear to be increasing. Concentrations of THg in eggs of the ivory gull collected from Seymour Island also increased between 1976 and 2004, although the increase was not statistically significant (Braune et al. 2006). Recent increases in Hg bioaccumulation in Canadian Arctic seabirds are consistent with a broader geographic analysis of temporal Hg trends in Arctic biota that showed a west-to-east circumpolar gradient in the occurrence of recently increasing Hg trends (Rigét et al. 2011). This gradient was due to a higher proportion of marine time series showing significant Hg increases in the Canadian and Greenland regions of the Arctic compared to the North Atlantic Arctic. The reasons for recent Hg increases in seabirds are unclear but may involve changes in atmospheric Hg deposition associated with anthropogenic and natural emissions, coupled with environmental and biological (e.g., food web) processes which may also be affected by climate change.

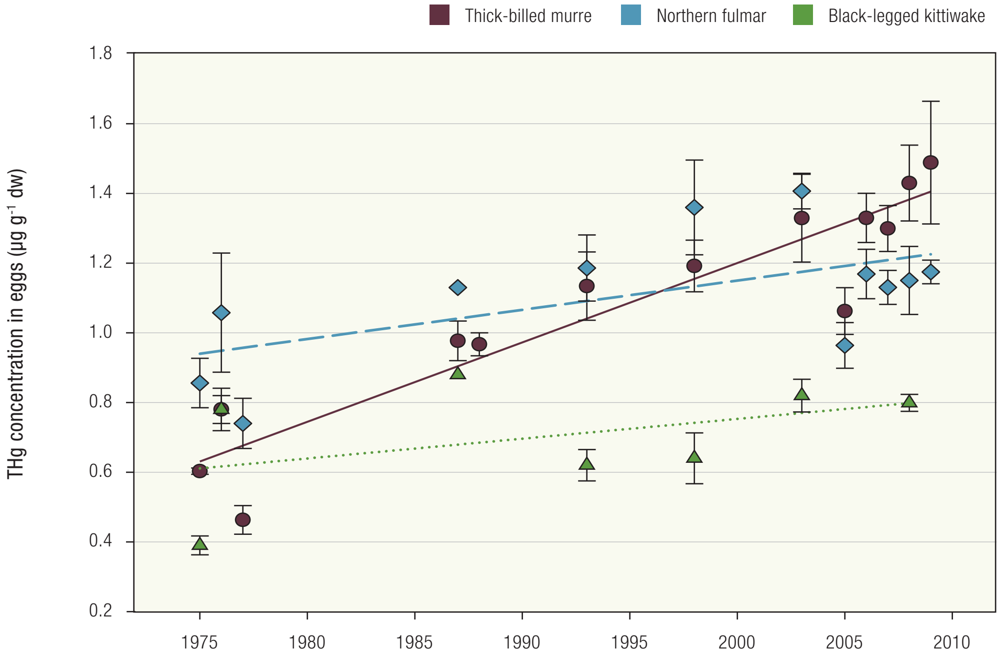

FIGURE 6.18

Mean concentrations (± standard error) of THg in eggs of thick-billed murres, northern fulmars and black-legged kittiwakes from Prince Leopold Island, 1975 to 2009 (Braune 2009c).

6.10.3 Statistical power of monitoring datasets to detect temporal trends

Long time series of monitored contaminants often show random inter-year variations that are not part of a trend (Bignert et al. 1993, 1994, Olsson 1995, 1998, Hebert and Weseloh 2003). The probability that a monitoring program will detect a temporal trend in contaminant concentrations, in spite of the inter-year fluctuations or *noise* in the data, represents its statistical power. The power to detect changes in contaminant levels with time depends on the pattern and magnitude of those changes (Nicholson and Fryer 1992) as well as the number of samples collected and the frequency of sample collection (Bignert et al. 1993, Hebert and Weseloh 2003). Implications of ignoring power include the collection of insufficient data to make reliable inferences about temporal trends and/or collection of extraneous data (Braune et al. 2003).

In order to examine inter-annual variation in contaminants data, and to improve the statistical power of the temporal trend data series for Canadian Arctic seabirds, eggs from northern fulmars and thick-billed murres have been collected annually from Prince Leopold Island since 2005. A statistical package (PIA) developed by Anders Bignert (Bignert 2007) to analyze trends in time series was used to compare the datasets for seabird eggs including the recent annual sampling (i.e. 2005 onwards) against results if sampling had continued on a five-year sampling cycle. The results show that recent annual sampling has greatly increased the power of the datasets for thick-billed murres and northern fulmars, and has lowered the lowest detectable change (Table 6.5).

6.10.4 Effect of climate change on temporal trends of Hg

Thick-billed murres breeding on Coats Island in northern Hudson Bay have been monitored for contaminants since 1993 (Braune et al. 2002). While THg levels in eggs of thick-billed murres have increased at Prince Leopold Island in the High Arctic, there is no apparent Hg trend at the Coats Island murre colony (Fig. 6.19). A decrease in ratios of $\delta^{15}N$ in thick-billed murre eggs from Coats Island indicate that adult murres have shifted their diet to a lower trophic position prior to laying, whereas murre eggs from Prince Leopold Island have not shown any consistent change over time (Braune 2009a).

Arctic cod is the main prey of thick-billed murres at Prince Leopold Island in the High Arctic (Gaston and Bradstreet 1993, Davidson et al. 2008) and, until the mid-1990s, was the most common prey item found in the diet of nestling murres throughout the Canadian Arctic (Gaston and Jones 1998). However, dietary studies have shown that between 1980 and 2002 there has been a shift from Arctic cod and benthic fish species to capelin and sandlance in the diet of thick-billed murres at Coats Island (Gaston et al. 2003). Warming ocean conditions and longer ice-free periods have been documented for Hudson Bay (Davidson et al. 2008, Gaston et al. 2009) leading Gaston et al. (2003) to suggest that the decline in Arctic cod and increase in capelin and sandlance were associated with a general warming of Hudson Bay waters. In contrast, there has been no overall change in summer ice cover reported at Prince Leopold Island (Davidson et al. 2008). After adjusting egg Hg concentrations from both murre

TABLE 6.5 A power analysis of THg concentrations measured from 1975 to 2009 in thick-billed murre and northern fulmar eggs from Prince Leopold Island. The results with the inclusion of annual sampling from 2005 to 2009 (left-hand column) are compared with results based on a five-year sampling regime (right-hand column) for each species.

	Thick-Billed Murre		Northern Fulmar	
	Annually 2005–2009 *n* = 55 # years = 13	Every 5 Years *n* = 35 # years = 9	Annually 2005–2009 *n* = 56 # years = 12	Every 5 Years *n* = 36 # years = 8
# Years Required*	12	12	10	10
Lowest Detectable Change**	4.1%	8.2%	3.7%	6.8%
Power of Time Series***	94%	41%	97%	55%

* Number of years required to detect an annual change of 5% with a power of 80%.

** Lowest detectable change in current time series.

*** Power of current time series to detect a log-linear trend of 5%.

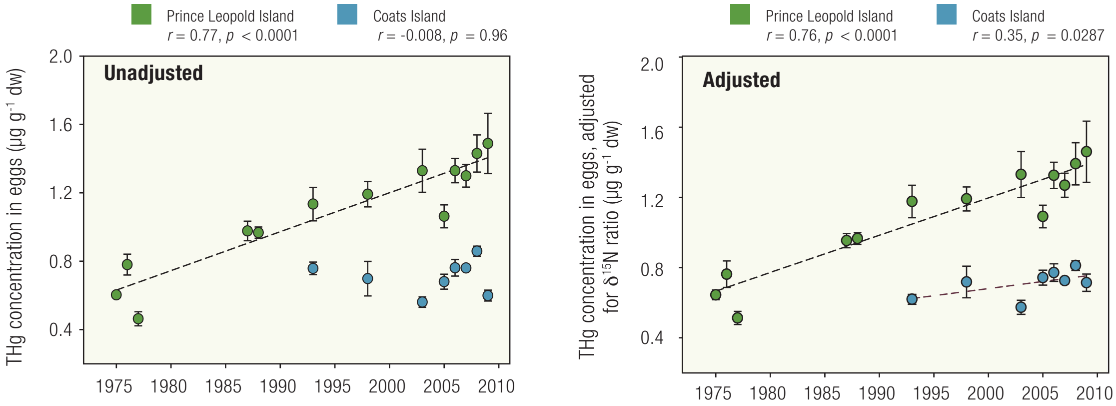

FIGURE 6.19

Unadjusted mean concentrations (± standard error) of THg in eggs of thick-billed murres from Prince Leopold Island and Coats Island compared to Hg concentrations adjusted for trophic position using δ^{15}N (Braune 2010b).

colonies for trophic position, little change was found in the Hg trend at Prince Leopold Island. However, the Coats Island colony showed a significant increase in Hg after trophic adjustment (Fig. 6.19), which indicates that the shift in diet which occurred for murres at Coats Island has affected the temporal Hg trend for that colony.

6.11 Marine mammals

6.11.1 Historical trends of Hg in marine mammals

Although Hg is a naturally occurring element in the environment, global human activity has led to a several-fold increase in Hg emissions to the atmosphere and in fluxes to marine and fresh waters (Mason et al. 1994). Calcified hard tissues such as teeth can be used to estimate the extent that global anthropogenic Hg emissions have increased Hg concentrations in Arctic biota since the pre-industrial period. Historical samples of hard tissues are often well-preserved in dry, cold climates like polar regions and are well represented in Arctic archaeological sites and museum collections. Teeth of marine mammals have THg concentrations that are correlated with THg levels in their soft tissues including muscle, kidney, liver and muktuk (Outridge et al. 2000). Therefore, analysis of historical and modern samples of teeth can be informative about the long-term changes of Hg intake and body burdens in wildlife.

Tooth THg content of Canadian Arctic beluga, walruses, and ringed seals has been examined from pre-industrial, historical (19th and early- to mid-20th Century) and present-day populations by Outridge et al. (2002, 2009, 2005). Long-term changes in THg content were assessed in the context of animal age structure and dietary information inferred from stable carbon and nitrogen isotopes in the teeth. The issue of preservation of the original Hg concentration in pre-industrial samples is addressed in detail by Outridge et al. (2005) and Dietz et al. (2009).

A significant increase in THg content was observed between historical and present-day teeth from ringed seals in Amundsen Gulf and from beluga in the Beaufort Sea and near Somerset Island (Fig. 6.20; Outridge et al. 2002, 2005, 2009). In the Beaufort Sea and Amundsen Gulf region, THg levels in marine biota increased after the late 19th Century, with the most substantial increases occurring in the mid-20th Century. Beluga teeth collected around Somerset Island from the late 19th Century to 1998 showed THg increases of 1.2- to 5.5-fold, but with no change until the 1920s to 1940s, which indicates that most, or all, of the increase took place after the early 20th Century (Outridge et al. 2005, Dietz et al. 2009). This historical trend in the Canadian Arctic is consistent with other Arctic observations from Greenland on the Hg content in animal hard-tissues (Dietz et al. 2009).

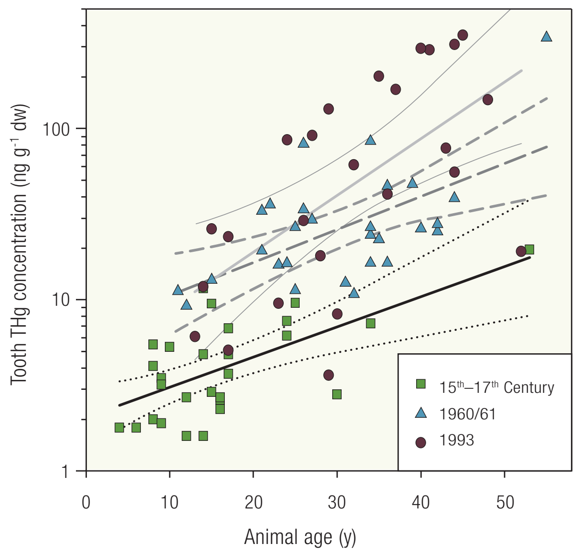

FIGURE 6.20

Tooth THg concentrations in relation to the age of Beaufort Sea beluga harvested in the 15th to 17th Centuries, in 1960 and 1961, and in 1993 in the Mackenzie Delta, with the respective linear regression lines and 95% confidence limits (source: Outridge et al. 2009).

Reflecting greater bioaccumulation in older individuals, the THg content of both pre-industrial and present-day teeth increased with animal age (Fig. 6.20). Likewise, the greatest differences between historical and modern samples of seals and beluga were observed for older individuals. For example, the teeth of modern seals 5 and 25 years old contained 8.8 and 17.2 times more Hg, respectively, than their 14th Century counterparts (Outridge et al. 2009). As suggested by some observed differences in carbon and nitrogen stable isotope ratios of teeth, historical Hg trends may also be influenced by changes over time in the feeding behaviour of seals and beluga (Dietz et al. 2009, Outridge et al. 2009). However, even after correcting for differences in trophic level of Somerset Island beluga using nitrogen stable isotopes, the THg content was still higher in modern teeth (Dietz et al. 2009).

Walrus was the only Arctic species studied so far that did not show greater THg in present-day teeth relative to pre-industrial samples obtained around Igloolik (Outridge et al. 2002). Walruses usually feed at a low trophic level, such as on clams and other bivalves, and may be less affected by increases in environmental Hg relative to other marine mammals at higher trophic levels, who are more impacted by biomagnification (Bernhard and Andreae 1984).

6.11.2 Ringed seal

The ringed seal is the most abundant Arctic pinniped with a circumpolar distribution, making it an ideal candidate for examining spatial trends of bioaccumulative contaminants. It has been a key species for biomonitoring in the Arctic Monitoring and Assessment Programme (AMAP) and in all phases of the NCP. Because of their high abundance, ubiquitous distribution, and central position in the food web, ringed seals play an important role in the ecology of Arctic marine ecosystems (Smith et al. 1991). The ringed seal diet consists of fish, mainly Arctic cod, polar cod, and crustaceans such as amphipods, mysids and euphausids (Reeves 1998). Recent measurements of fatty acids in ringed seal blubber suggest that Arctic cod are important for seals particularly in the eastern Arctic while capelin and sandlance may be more important in Hudson Bay (Thiemann et al. 2007). Fatty acid measurements also suggest that ringed seals forage within limited areas (Thiemann et al. 2007), consistent with other observations that they are relatively sedentary. Adult male seals may potentially occupy the same under-ice habitat for up to nine months (Smith 1987, Reeves 1998). Some large-scale movements, however, have been documented such as from the western Canadian Arctic islands to the East Cape of Siberia (Smith 1987) and between Greenland and Canadian waters (Teilmann et al. 1999).

Observations since the 1970s have found elevated THg concentrations in ringed seal liver and kidney (Smith and Armstrong 1978, Wagemann and Muir 1984, Muir et al. 1992). Early assessments also noted that concentrations were two- to three-fold higher in ringed seals in the southern Beaufort Sea—Sachs Harbour and Ulukhaktok—than in the eastern Canadian Arctic Archipelago (Muir et al. 1992, Wagemann et al. 1996), and higher in the Hudson Strait/Labrador region than in southern Hudson Bay (Dietz et al. 1998, Muir et al. 1999).

In 2005, the NCP extensively revised the monitoring program for ringed seal and initiated annual sampling at three locations—Sachs Harbour, Arviat, and Resolute Bay—with additional, one-time sampling at about ten other communities over a five-year cycle. Combined with previous sampling of ringed seals under Phase II of the NCP, there is currently a large biological dataset available for about 550 individual seals collected in the past ten years that includes measurements of age as well as carbon and nitrogen stable isotopes. Additional collections related to Hg measurements have been made by Fisheries and Oceans Canada (Gaden et al. 2009, Young et al. 2010, Brown and Reimer 2011).

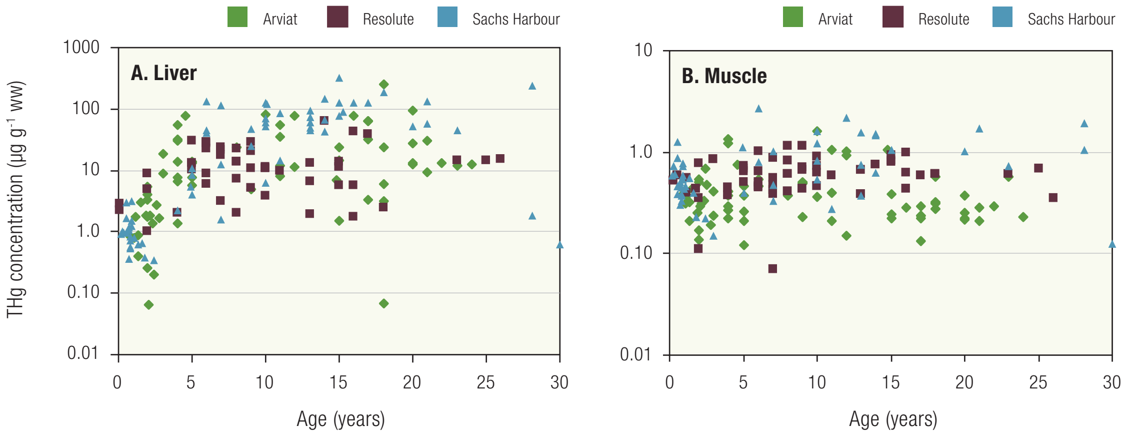

FIGURE 6.21

Influence of age on log-transformed THg concentrations (µg g^{-1} ww) in liver (A) and muscle (B) of ringed seals collected from Arviat, Resolute, and Sachs Harbour in 2005 to 2007 (Muir 2010).

Levels and spatial trends

Comparisons of average concentrations in ringed seal liver are challenging because Hg varies with age. The relationship of THg in liver and muscle with age was examined for seals collected from Arviat, Resolute, and Sachs Harbour in 2005 to 2007 (Fig. 6.21). Liver THg concentrations increased exponentially with age for seals less than five years and appeared to be relatively stable beyond that age. Adjusting for age is not always straightforward because the relationship between liver THg and age can vary among locations. Thus, for this assessment of spatial trends, only data for seals aged five years or older were used. Seal muscle concentrations of THg showed no overall trend with age (Fig. 6.21). Atwell et al. (1998) also found no correlation between muscle THg concentrations and age in ringed seals from Resolute. The lack of a relationship between muscle THg and age for seals and beluga has been interpreted as a result of more rapid elimination of the predominately MeHg in muscle, while for liver, the increase with age implies longer term storage of inorganic Hg (Dehn et al. 2005, Loseto et al. 2008b). Measurements of Hg elimination rates in seal muscle and liver would help to evaluate the appropriateness of these tissues for the long-term monitoring of Hg trends.

On average, THg concentrations in seal muscle were 32 times lower than in liver for tissues collected from the same location, and similar ratios have been reported elsewhere (Wagemann et al. 1996, Dehn et al. 2005). Most of the THg in liver is in the form of non-toxic, inorganic Hg-selenides (Wagemann et al. 2000). Dehn et al. (2005) showed that the proportion of THg as MeHg in ringed seal liver from Ulukhaktok ranged from 1–12% with lower proportions in animals greater than five years old, while Wagemann et al. (2000) reported an average MeHg fraction of 2% for ringed seals from the Canadian Arctic. In contrast, Dehn et al. (2005) showed that 81% of THg in muscle was in the form of MeHg.

Spatial trends of THg in ringed seal liver were examined using geometric means (± 95% confidence intervals) from each of 12 communities across the Canadian Arctic for which data were available post 2000 (Fig. 6.22). Levels for males and females were combined because, in general, THg concentrations in ringed seal liver are not influenced by the sex of the animals (Dehn et al. 2005). Sample sizes ranged from n = 6 (Inukjuaq) to n = 72 (Resolute). No statistical analysis was conducted for this assessment because different THg-age relationships among populations complicate age-adjustment of liver THg concentrations. Unadjusted THg concentrations in seal liver were higher in Sachs Harbour and Ulukhaktok compared to most sampling locations in the eastern Arctic including Arctic Bay, Arviat, Inukjuaq, Nain, Pangnirtung, Pond Inlet, Qikiqtarjuaq, and Resolute. Wagemann et al. (1996) and Rigét et al. (2005) reported a very similar geographic pattern of THg in liver with highest concentrations in ringed seals from the western Arctic Archipelago locations of Ulukhaktok and Sachs Harbour.

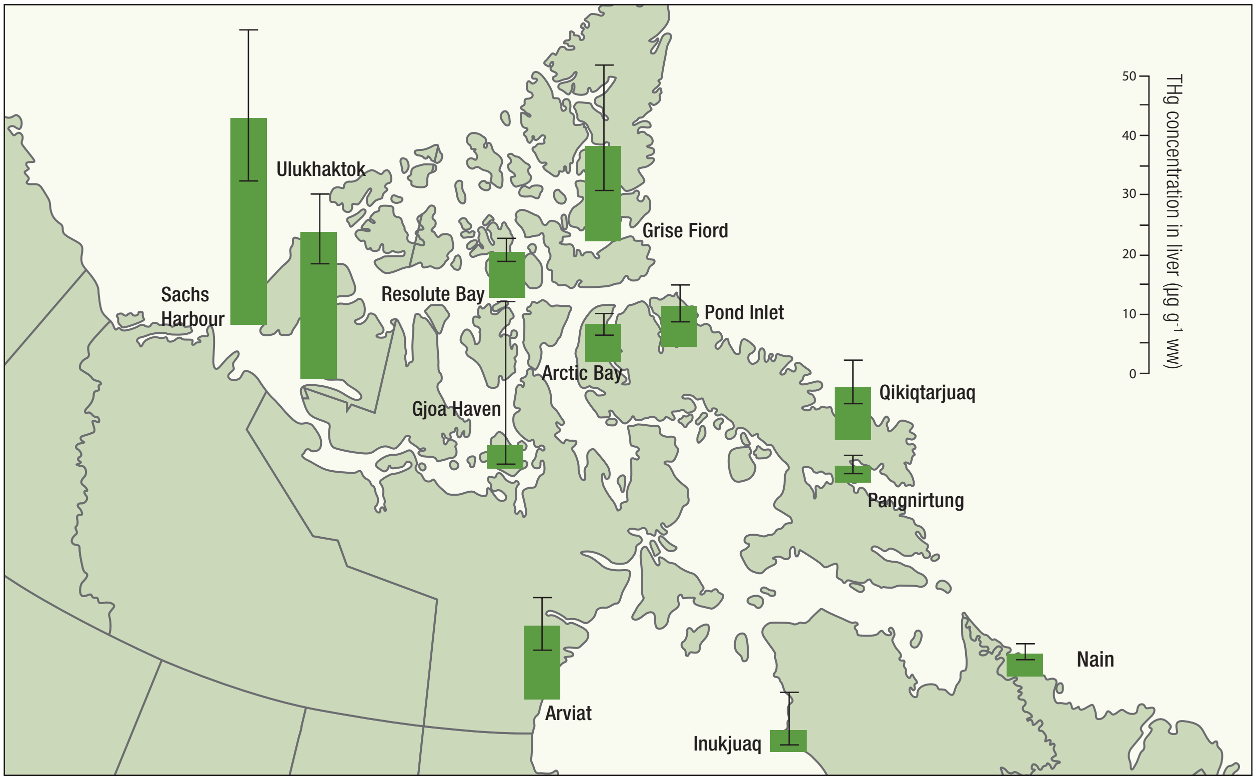

FIGURE 6.22

Concentrations of THg (geometric means ± 95% confidence intervals) in ringed seal liver from 12 communities sampled between 2000 and 2008. Only individuals aged five or older were included (Muir 2010).

Spatial trends of THg in muscle of adult ringed seals were examined using length-adjusted means from each of 12 communities across the Canadian Arctic for which data were available post 1999 (Fig. 6.23). Muscle THg concentrations varied significantly among locations in the eastern and western Arctic, and were significantly higher in Sachs Harbour, Ulukhaktok (Gaden et al. 2009), Resolute, and Qikiqtarjuaq compared with other communities sampled. Similar to results for liver, higher concentrations in ringed seal muscle were found in the western Arctic—Sachs Harbour, Ulukhaktok—but in addition, two other High Arctic sites—Resolute, Qikiqtarjuaq—showed elevated THg levels. It is unclear what factors are driving these geographic trends.

In a separate investigation in Nunatsiavut, ringed seals were collected in 2008 and 2009 from the Nachvak, Saglek, Okak, and Anaktalak fjords (Brown and Reimer 2011). Average THg concentrations in ringed seal muscle were low and ranged from 0.128–0.230 µg g^{-1} ww among the four fjords (Fig. 6.24). Average THg concentrations were slightly higher in seals from Okak Fjord than from Nachvak, Saglek, and Anaktalak fjords. Overall, these results show relatively little spatial variation among Labrador fjords and are consistent with low muscle concentrations of THg measured in ringed seals at two other sites in the region (Fig. 6.23).

Comparison of Hg concentrations in ringed seal with other seal species

Ringed seals, harbour seals, and bearded seals were sampled near communities in western Hudson Bay to examine differences in diet and THg concentrations between size classes (Young et al. 2010). Suggesting age-specific differences in foraging, muscle THg concentrations and $\delta^{15}N$ ratios varied between pups, juveniles, and adults of ringed and bearded seals. Ringed and bearded seals had similar THg concentrations in muscle, but harbour seals had levels an order of magnitude higher. For liver, ringed seals had lower THg concentrations than both bearded and harbour seals. Based on carbon stable isotope ratios, bearded seals appeared to feed more in the benthic food web. Elevated liver and muscle THg concentrations in harbour seals compared to the other two species were likely due to harbour seals feeding at a higher trophic level.

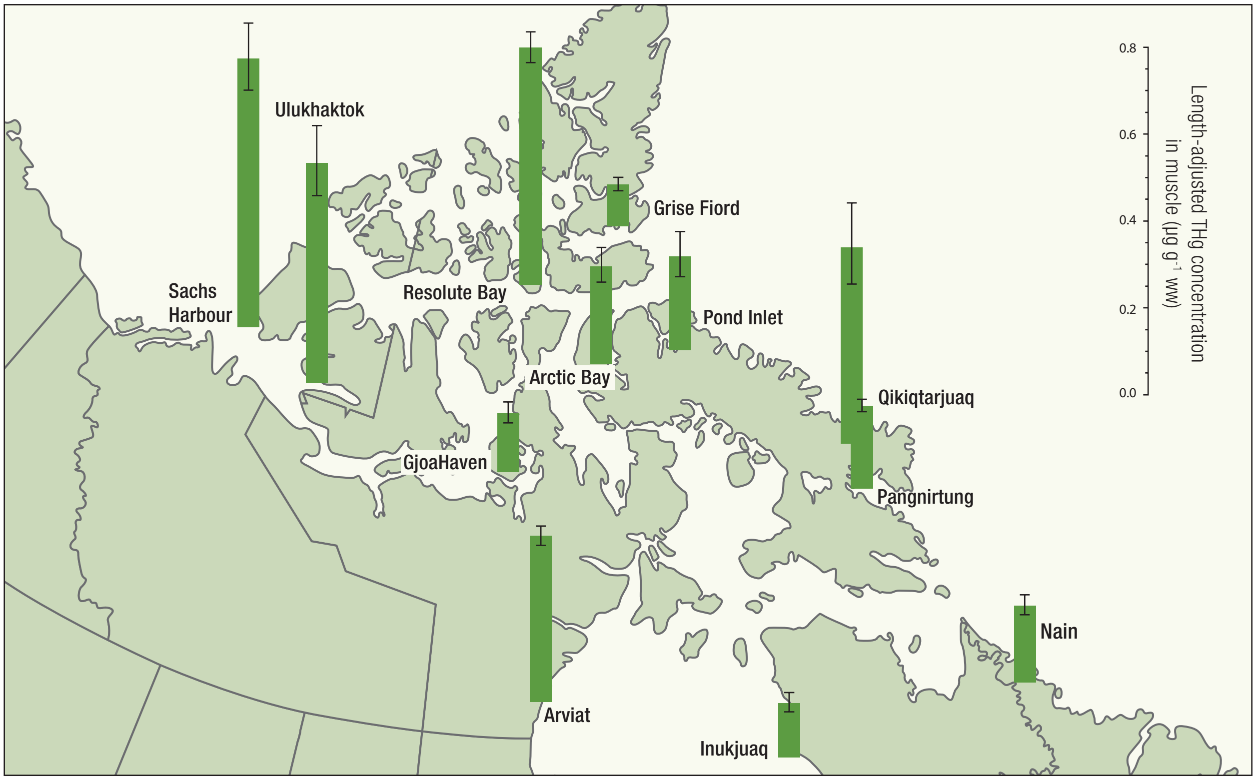

FIGURE 6.23

Concentrations of THg (length-adjusted geometric means ± 95% confidence intervals) in ringed seal muscle from 12 communities sampled between 1999 and 2009 (Muir 2010).

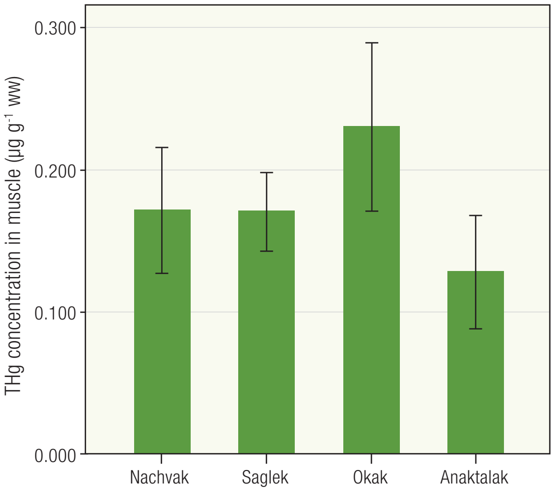

FIGURE 6.24

Average THg concentrations (± 95% confidence intervals) in ringed seal muscle collected from Nunatsiavut fjords in 2008 and 2009 (Brown and Reimer 2011).

Temporal Hg trends in ringed seals

No significant trends in THg concentration in ringed seal muscle were found in the communities of Arviat, Resolute, Sachs Harbour, or Pangnirtung from 1999 to 2009 (Fig. 6.25). While declines were observed in seals from Arviat and Resolute, the trends were not statistically significant, perhaps due to low sample sizes and relatively high within-year variance. This overall finding is in general agreement with results for THg in ringed seal muscle from Ulukhaktok where no significant trends were observed during 2002 to 2007 (Gaden et al. 2009).

Nitrogen stable isotope ratios were available for ringed seal muscle from Arviat, Resolute, and Sachs Harbour to determine if ringed seal diets are changing over time (Fig. 6.25). Average $\delta^{15}N$ ratios changed only modestly (± 1‰) over the study period which suggests seals were feeding at the same trophic level. Earlier measurements of $\delta^{15}N$ in ringed seal muscle collected in Resolute from 1988 to 1990 averaged 16.4 ± 0.2‰ (Atwell et al. 1998), which is within the range observed for samples collected between 2004 and 2009. Gaden et al. (2009) also found only minor shifts in average $\delta^{15}N$ of < 1‰ in seals from Ulukhaktok over a 14-year period.

BOX 6.2 Nunatsiavut Community Involvement in Ringed Seal Research


Sam Bentley

Nunatsiavut communities and their members have played a vital role in the ArcticNet Nunatsiavut Nuluak Program since its inception in 2006 to address Inuit concerns about the impacts of climate change, modernization, and contaminants on the health of marine ecosystems and communities. Inuit have been involved in all aspects of the program, including the direction of research, design and planning of projects, field work, and reporting of results. The close partnership between Nunatsiavut community members and researchers has fostered exchanges and connections of Inuit culture and history—through the life stories, practices, and experiences of people from the local area—with science and research—through presentations, activities, lab work, group projects and hands-on field work. The knowledge, experience, and insight of community members have been invaluable to the success of the program, and their participation in the research has enhanced the studies at a local level and made it more relevant to Inuit.

Recognizing the importance of collaboration among research programs, the NCP is supporting current initiatives under ArcticNet's Nunatsiavut Nuluak Program to better understand changes in ringed seal contaminant trends and health. Scientific study on contaminant levels in ringed seals, genomic-based health indices, ringed seal telemetry, and the marine food web will be complemented by Nunatsiavimmiut Knowledge—traditional Inuit knowledge of Nunatsiavut—on ringed seal behaviour, ecology, distribution, and health. The Inuit Knowledge information is being collected from local hunters and documented to improve our understanding of ecosystem changes that may be related to or influencing contaminant trends in Nunatsiavut ringed seals. The linked investigation using Nunatsiavimmiut Knowledge and science may prove to be the most valuable aspect of this collaborative project.

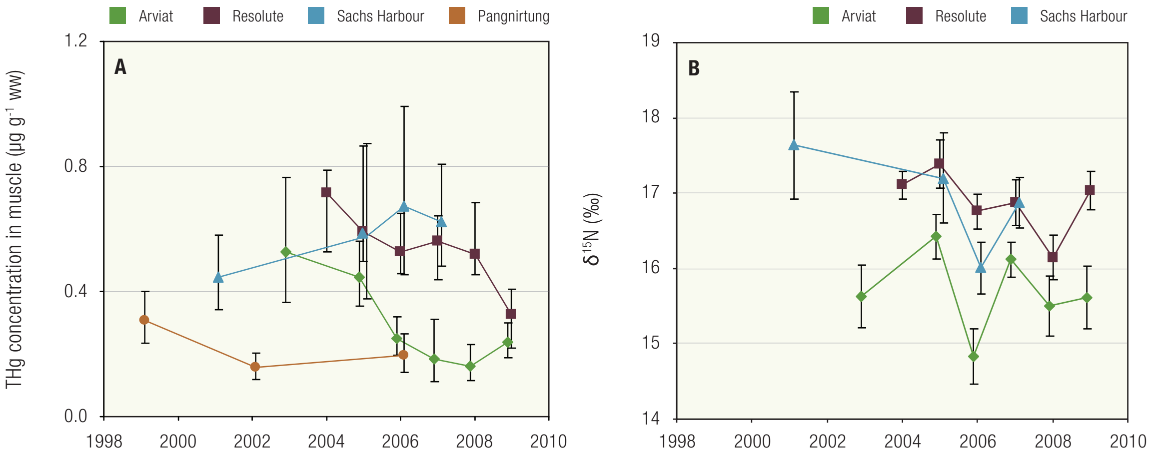

FIGURE 6.25

Temporal trends of THg concentrations (A) and $\delta^{15}N$ ratios (B) in ringed seal muscle from Arviat, Resolute Bay, Sachs Harbour, and Pangnirtung. Values are geometric means ± 95% confidence limits. $\delta^{15}N$ ratios were not available yet for Pangnirtung (Muir 2010).

Temporal trends for THg in ringed seal liver are also available as a result of the past focus on the analysis of livers at many Canadian Arctic sampling locations (Wagemann et al. 1996, Fisk et al. 2003) and more recent measurements (Muir et al. 2008). However, year-to-year variation in liver THg concentrations is often very high (Muir et al. 2008), making assessment of temporal trends difficult.

Temporal Hg trends in ringed seals in relation to ice cover

Gaden et al. (2009) reviewed THg concentrations in the muscle tissue of adult ringed seals (aged ≥ 7 years) sampled at Ulukhaktok (Holman) from 1973 to 2007. Stable isotope ratios of nitrogen and carbon were also analyzed from the muscle tissue. Seal THg, $\delta^{15}N$ and $\delta^{13}C$, together with age and sex were examined for temporal trends and associations with the length of the ice-free season in the eastern Amundsen Gulf.

No linear trends were observed for unadjusted THg concentrations in ringed seal muscle (Fig. 6.26, top panel). Age influenced the THg concentrations but there was no significant difference between levels in males and females. Age-adjusted THg concentrations displayed a second-order polynomial relationship with the length of the ice-free season in the year prior to seal collection. Seal THg concentrations were higher for years with short (~ 60 days) and long (greater than ~ 140 days) ice-free seasons (Fig. 6.26, bottom panel).

Gaden et al. (2009) hypothesized that the THg trend reflected the influence of the previous year's environmental conditions on prey consumption by ringed seals, although seal $\delta^{15}N$ and $\delta^{13}C$ ratios were not strongly associated with the length of the ice-free season possibly due to a mismatch in turnover times between THg and stable isotope ratios in muscle. Variation in the length of the ice-free season may have influenced the overall dietary exposure of Hg to seals by affecting the population of Arctic cod—the preferred prey of ringed seals in winter (Smith 1987). Additional field study is necessary to confirm the cause of this apparent trend, which may be quite important in the context of climate change.

6.11.3 Beluga, walrus, and narwhal

As part of ongoing NCP monitoring of whales and walruses—supported in the eastern Arctic by the Nunavut Wildlife Management Board and the Nunavut Implementation Fund, and in the western Arctic by the Fisheries Joint Management Committee and Fisheries and Oceans Canada—samples were collected by hunters during their subsistence hunts using standardized marine mammal sampling kits. Blubber, muktuk, kidney, liver, muscle, and the lower jaw, as well as morphometric data were collected for each animal. Animals were harvested by native hunters using their preferred methods.

Measurements of THg levels in tissues and organs of beluga, narwhals, and walruses have been on-going from, in some cases, the early 1980s (Lockhart et al. 2005b). Concentrations of THg vary from organ to organ within an animal and from animal to animal for a given site and time of collection. Of the organs analyzed, liver generally has the highest concentrations of THg, followed by kidney, muscle, and muktuk.

Levels in beluga from Hudson Bay

Gaden and Stern (2010) analyzed wet weight concentrations of THg in liver and muscle tissues from Hudson Bay beluga on the west coast (Arviat, 1984 to 2008) and the east coast (Sanikiluaq, 1994 to 2008). Mean annual concentrations of THg in liver were between 5–21 $\mu g\ g^{-1}$ and 5–43 $\mu g\ g^{-1}$ in beluga sampled at Arviat and Sanikiluaq, respectively. Despite some higher liver concentrations observed in Sanikiluaq, location was not a significant factor influencing THg levels in the beluga. Average THg levels in muscle were between 0.68–1.3 $\mu g\ g^{-1}$ and 0.49–1.8 $\mu g\ g^{-1}$ in western and eastern Hudson Bay beluga, respectively (Gaden and Stern 2010). From 1994 to 2008, beluga muscle appeared to have higher THg in females at Arviat compared to females at Sanikiluaq. However, the difference was not significant after accounting for inter-year variation.

Eric Loring

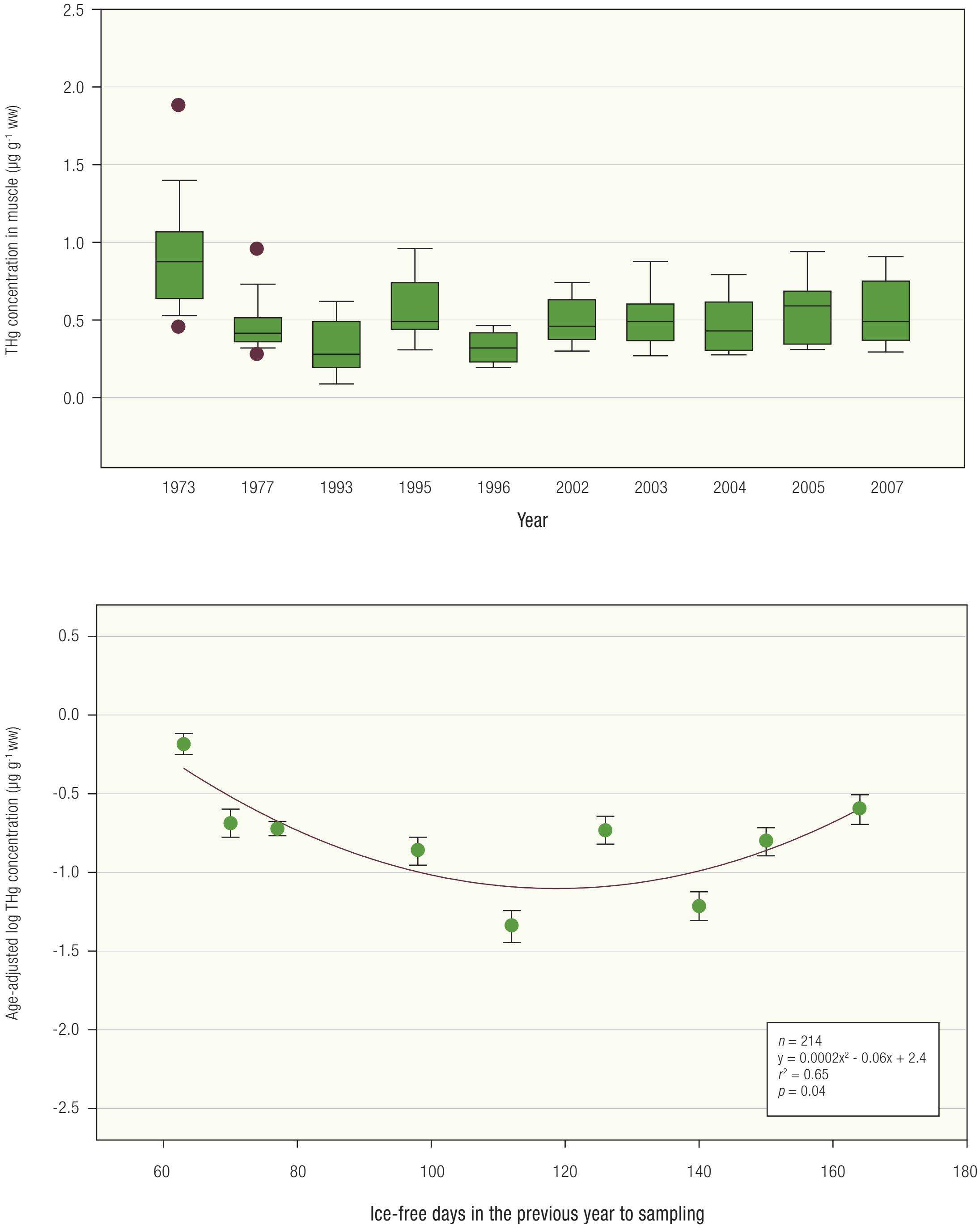

FIGURE 6.26

Box and whisker plots of THg concentrations in muscle of adult ringed seals (≥ 7 years, sexes pooled) at Ulukhaktok (Holman) from 1973 to 2007 (upper panel). Lines in boxes represent mean concentrations; whiskers represent 5th and 95th percentiles. Age-adjusted, log-transformed means (± standard error) of the THg data were plotted against the length of the ice-free season in the year previous to seal collection. Reprinted with permission from Gaden et al. (2009). © American Chemical Society.

Temporal trends in beluga from Hudson Bay

Temporal THg trends in muscle and liver of Hudson Bay beluga were examined for associations with δ^{15}N and δ^{13}C ratios. A general linear model was used to examine trends while accounting for age, sex, and location. Due to differences in THg between males and females in some years, sexes were analyzed separately.

No temporal trends were observed with unadjusted annual means of THg concentrations in beluga liver (Fig. 6.27) but a significant decrease in muscle THg (r = -0.28, p = 0.03) was observed for Sanikiluaq male beluga (Fig. 6.28). Age was correlated with THg concentrations and after accounting for age in the analysis, only THg in female beluga from Arviat varied significantly with time. Concentrations of THg increased in liver (r = 0.87, p = 0.02) but decreased in muscle (r = -0.80, p = 0.02) of female beluga from Arviat. The absence of corresponding trends in males from Arviat suggests there may be differences in habitat and prey selection between sexes (Loseto et al. 2006, 2008a). The rise of THg in beluga liver over the study period may have partly resulted from the accumulation of inorganic Hg which has a slow elimination rate in mammalian livers (Friberg et al. 1979).

Dietary tracers measured in beluga liver (δ^{15}N and δ^{13}C ratios) generally did not explain THg concentrations in liver or muscle of Arviat and Sanikiluaq beluga (Gaden and Stern 2010). Sexes were pooled in this analysis because of no significant gender differences in stable isotope ratios or temporal trends. Only δ^{13}C ratios in Arviat beluga showed a significant temporal trend, becoming more depleted between 1984 and 2008 (p = 0.02). The observed decrease in muscle THg and depletion in δ^{13}C ratios in beluga from Arviat suggest that these marine mammals may have shifted towards a less contaminated, more pelagic diet (Cherel and Hobson 2007, Ohizumi and Miyazaki 2010). The lack of trends in δ^{15}N ratios suggest beluga in Hudson Bay have not switched to prey of a different trophic level (Hobson and Welch 1992). Polar bears in western Hudson Bay have similarly shown a negative shift in δ^{13}C ratios from 1991 to 2007 (McKinney et al. 2009).

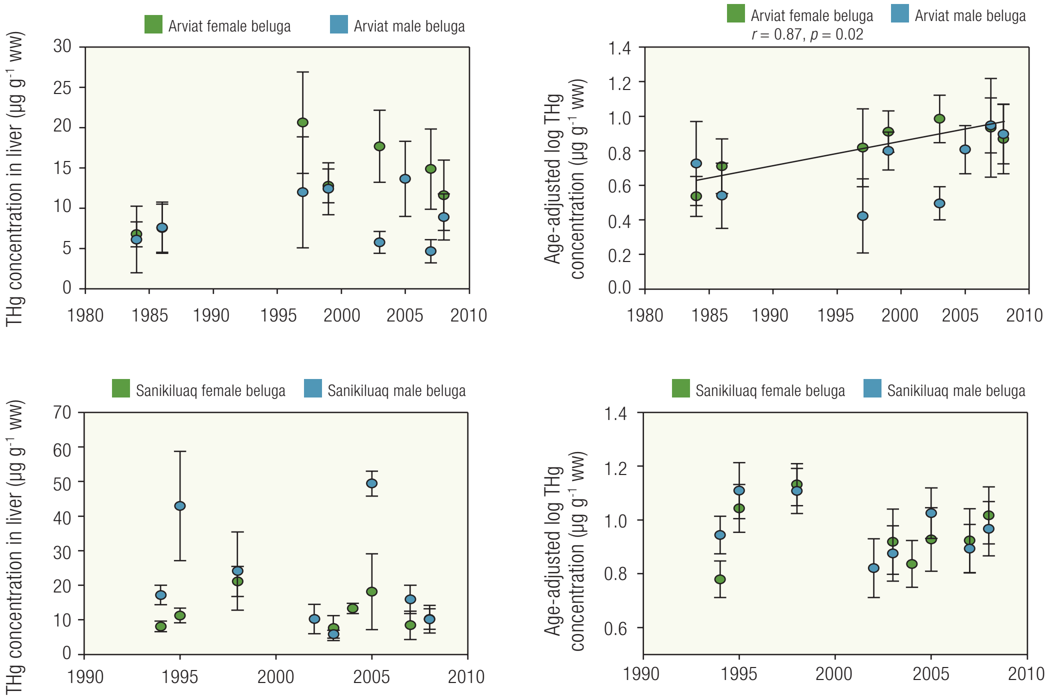

FIGURE 6.27

Annual means of THg concentrations (± standard error) in beluga liver sampled at Arviat (top panels) and Sanikiluaq (bottom panels) from 1984 to 2008 (data from Gaden and Stern 2010). Unadjusted and log-transformed, age-adjusted concentrations are presented in the left and right panels, respectively.

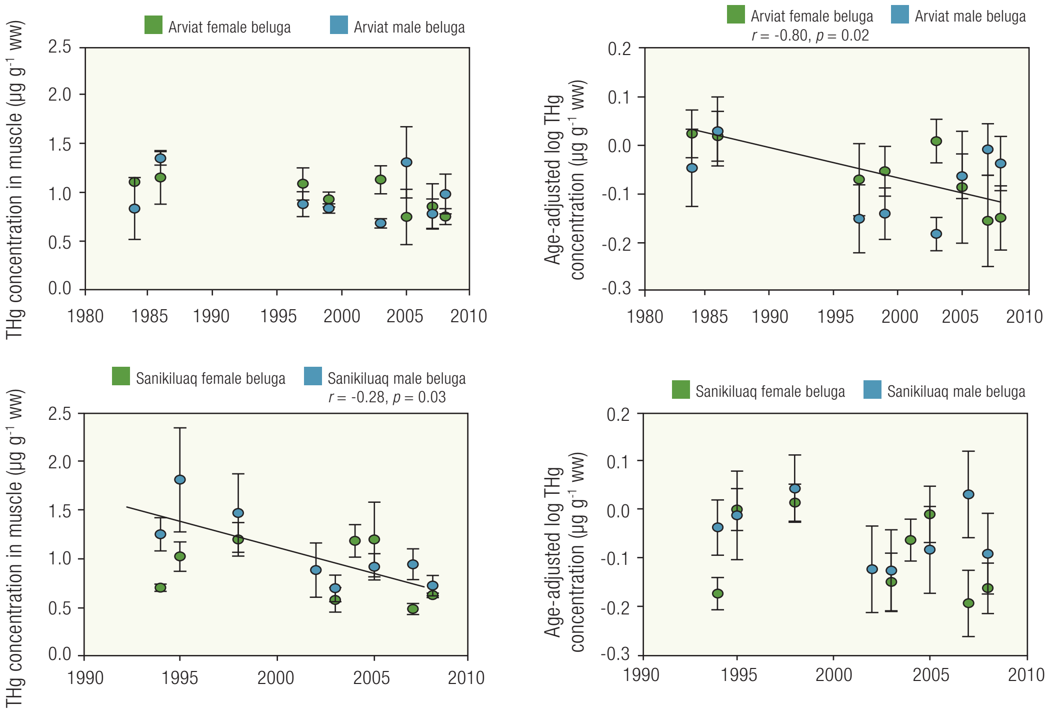

FIGURE 6.28

Annual means of THg concentrations (± standard error) in beluga muscle sampled at Arviat (top panel) and Sanikiluaq (bottom panel) from 1984 to 2008 (data from Gaden and Stern 2010). Unadjusted and log-transformed, age-adjusted concentrations are presented in the left and right panels, respectively.

The declining dietary exposure of THg in some Hudson Bay beluga may be affected by a longer ice-free season in Hudson Bay (Gagnon and Gough 2005, Ford et al. 2009, Rodrigues 2009, Hochheim et al. 2010).

Temporal trends in beluga from the western Arctic

A previous analysis of THg temporal trends in beluga whales from the western Arctic revealed that liver concentrations increased from the 1980s to the 1990s followed by stable, high levels in the early 2000s (Lockhart et al. 2005b). For this analysis, liver THg concentrations were age-adjusted to correct for long-term bioaccumulation, assuming that the rate of Hg accumulation over time is consistent and thus comparable across years. More recent analysis of the beluga data (by ANCOVA test for homogeneity of slopes) indicated that this assumption no longer holds for the dataset. Therefore, a different approach was adopted whereby THg concentrations of liver and muscle were averaged for specific age or size classes, respectively, to evaluate whether levels in beluga whales have increased, decreased, or remained stable since the previous assessment of temporal trends.

Both liver and muscle were examined because THg species and the physiological processes that govern its speciation differ between tissue types. Mercury in liver is largely inorganic and bound to selenium, thus rendering it biologically unavailable following demethylation. On the other hand, Hg in muscle is largely in the form of MeHg and better reflects recent diet exposure related to beluga size and habitat use (Loseto et al. 2008b).

Two age groups were defined to examine temporal trends in liver THg concentrations based on representation within the dataset of the age groups across years and the inclusion of both sexes. The first age group included whales from 16 to 35 years of age and the second included whales from 36 to 55 years. Mercury concentrations were significantly different between the age groups ($p < 0.0001$) when averaged over all years (16–35 years: 18.84 ± 1.1 $\mu g\ g^{-1}$ ww; 36–55 years: 53.38 ± 3.9 $\mu g\ g^{-1}$ ww). Age was estimated based on one growth layer group of dentine (Stewart et al. 2006).

In addition, to examine temporal trends in muscle THg concentrations, only male beluga were grouped by size. The size groups were defined according to

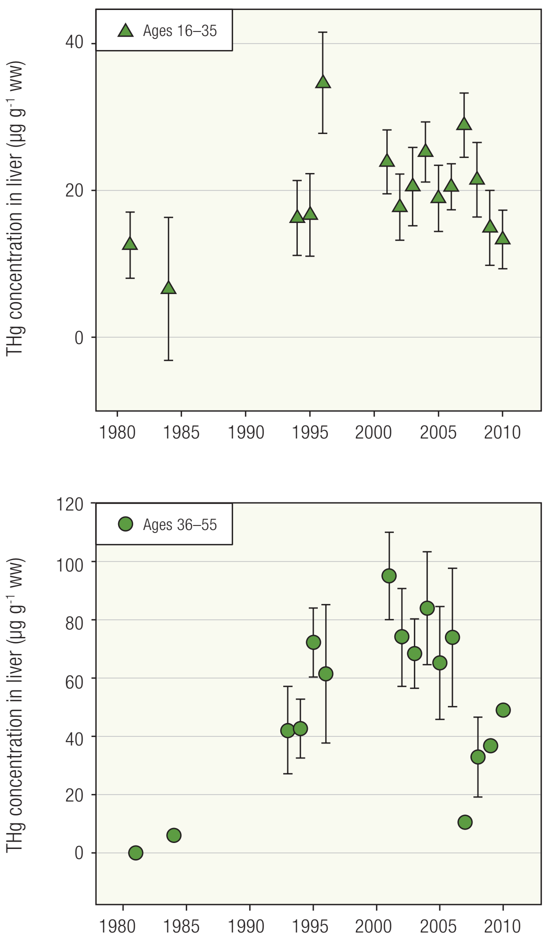

FIGURE 6.29

Temporal trends in liver THg concentrations in beluga whales of two age classes (16–35 and 36–55 years old) (Loseto and Stern 2011).

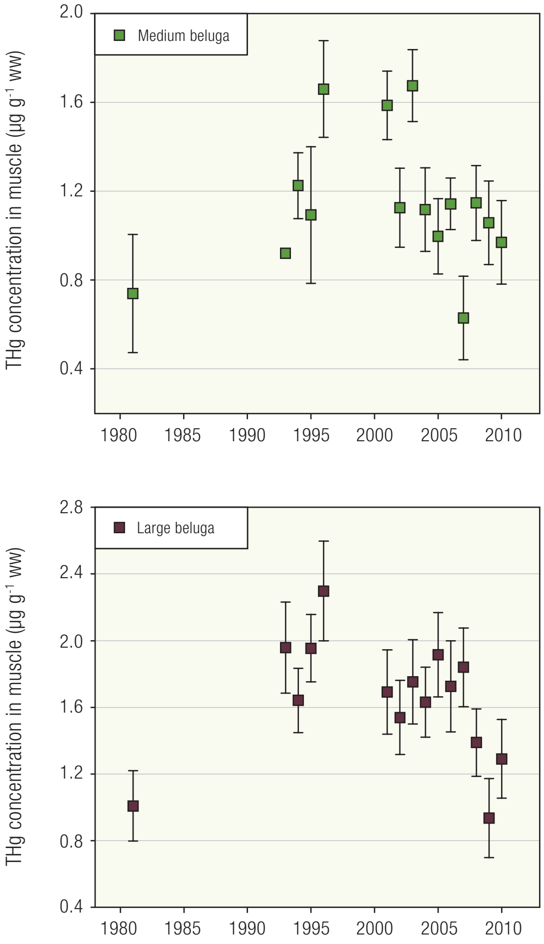

FIGURE 6.30

Temporal trends in muscle THg concentrations in beluga whales of two size classes (medium: 380–420 cm, large: > 420 cm) (Loseto and Stern 2011).

habitat use and relationships found with diet (Loseto et al. 2006, 2008a, 2009), with medium-sized males (380–420 cm) and large-sized males (> 420 cm). When averaged over all years, THg concentrations were significantly higher in the large males (1.60 ± 0.06 µg g^{-1} ww) than in the medium-sized males (1.18 ± 0.05 µg g^{-1} ww) ($p < 0.0001$).

A linear regression analysis of both muscle and liver THg concentrations for the two size or age groups was performed for data from 1981 to 2002. Consistent with the findings of Lockhart et al. (2005b), results in all cases supported increasing log-transformed THg concentrations in liver and muscle from 1981 to 2002. Linear regression analysis was then applied to the data spanning from 2002 to 2010 to determine if THg concentrations in beluga whales have increased, decreased, or remained stable since the last documented increase during the 1990s. Liver THg concentrations in young whales showed no significant change ($r^2 = 0.1$, $p = 0.3$) while older whales showed a slight decreasing trend that was not significant ($r^2 = 0.3$; $p = 0.1$) (Fig. 6.29). In contrast, muscle THg concentrations decreased significantly in both size classes (medium beluga: $r^2 = 0.25$, $p = 0.002$; large whales: $r^2 = 0.22$, $p = 0.032$) (Fig. 6.30). Further investigation is needed to determine what factors may be reducing MeHg exposure in western Arctic beluga.

Temporal trends in walruses and narwhals

Walruses and narwhals were sampled occasionally over the last three decades in the Foxe Basin near Igloolik and off Baffin Island near Pond Inlet, respectively. Liver THg concentrations in walruses were mostly

< 3 µg g^{-1} ww (Fig. 6.31) and, in general, were considerably lower than in other marine mammals such as narwhals (Fig. 6.32), seals, and beluga. No temporal trends were observed for walruses between 1982 and 2009, although the number of monitoring years was relatively low during that period (Fig. 6.31). Liver THg concentrations of walruses were not related to age. Similarly, no temporal trends were evident for liver THg concentrations of narwhals near Pond Inlet between 1978 and 2004, although the number of monitoring years was again relatively low (Fig. 6.32). Narwhal length data are provided with liver THg concentrations rather than age data because an accurate method for age determination is not yet available for this species. Liver THg concentrations of narwhals were not related to length for animals of 350 to 485 cm, and therefore, concentrations were not length-corrected.

6.11.4 Polar bear

The polar bear is the apex predator of the Arctic marine ecosystem. Due to its position at the top of the marine food web, its levels of Hg are among the highest observed in the Arctic (Muir et al. 1999, Dietz et al. 2009). Polar bears are distributed throughout the circumpolar region and, thus, are an ideal sentinel or monitoring species for contaminants.

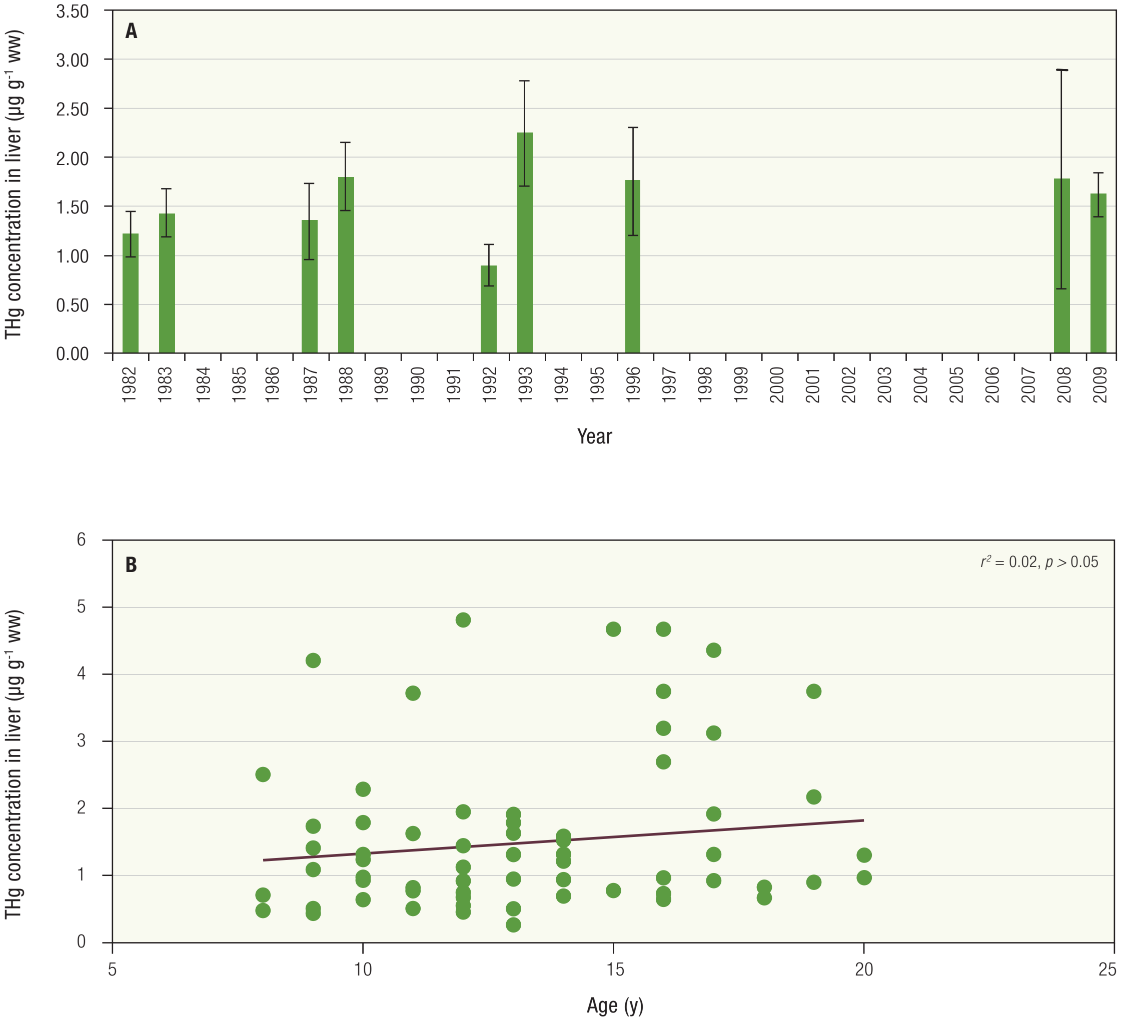

FIGURE 6.31

A) Temporal variation in concentrations of THg (mean ± standard error) in liver of Igloolik walruses ranging in age from 8–20 years.
B) Relationship between THg concentrations in walrus liver and the animal's age ($p > 0.05$). Data are from Stern (2011).

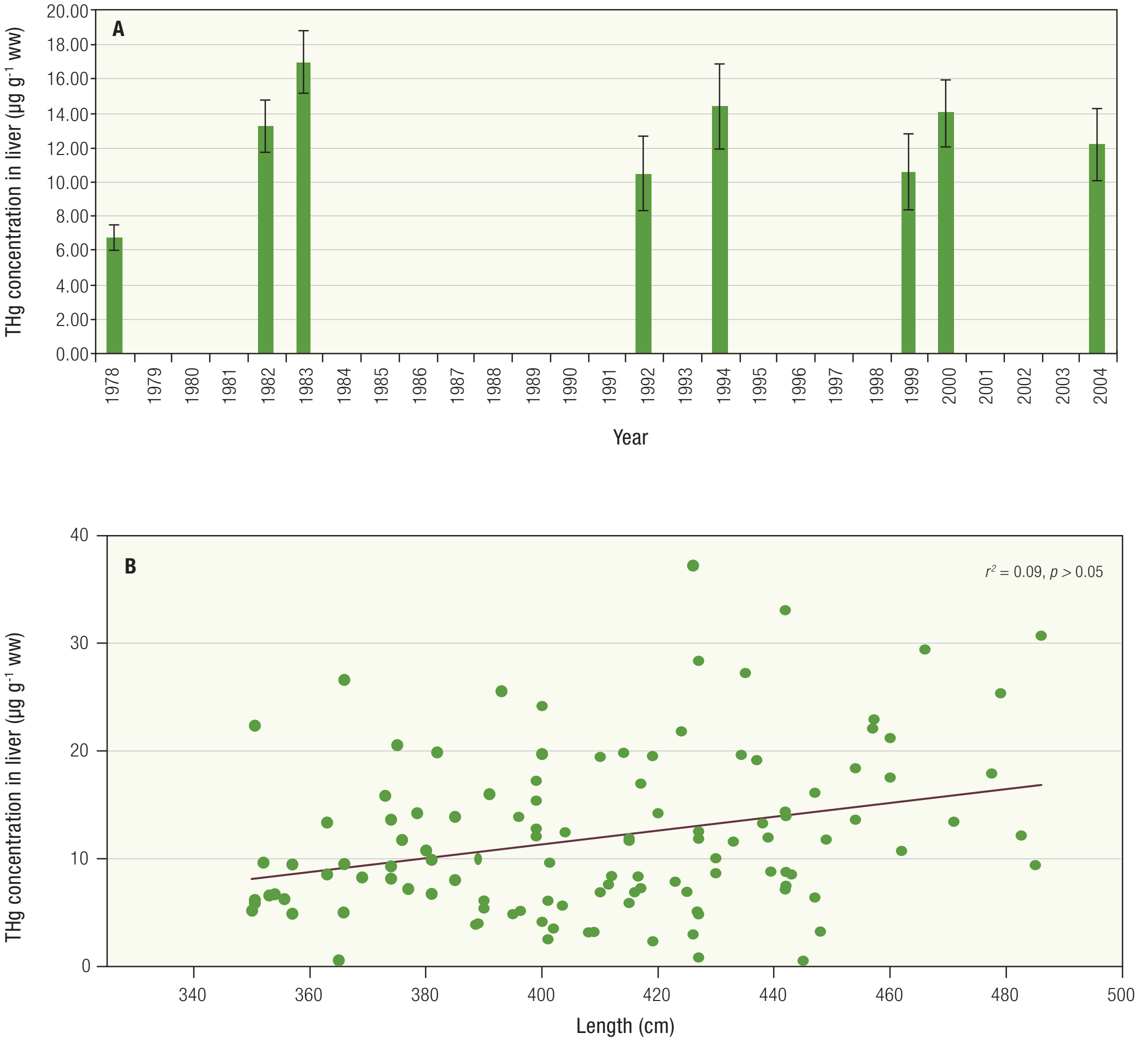

FIGURE 6.32

A) Temporal variation in liver concentrations of THg (mean ± standard error) of Pond Inlet narwhals ranging in length from 350–485 cm. B) Relationship between THg concentrations in narwhal liver and the animal's length ($p > 0.05$). Data are from Stern (2011).

Levels and spatial trends

Recent spatial trends of trace elements in polar bears were investigated with samples collected from 2005 to 2008 in Alaska (Chukchi and Bering seas), Canada, and Greenland (Routti et al. 2011). Liver concentrations of THg in polar bears from the northern and southern Beaufort Sea were higher than in any other areas except the adjacent Lancaster/Jones Sound and Gulf of Boothia (Fig. 6.33). Previous studies on polar bears from the Canadian Arctic similarly reported Hg concentrations being highest in the Beaufort Sea population and lowest in the Hudson Bay population (Norstrom et al. 1986, Braune et al. 1991, Rush et al. 2008). Bears from the Hudson Bay area had lower concentrations of THg compared to other areas except the adjacent Davis Strait. Concentrations of THg in bears from Alaska (Chukchi and Bering seas) were as low as those from Hudson Bay, which is consistent with the generally low levels of THg found in marine and terrestrial mammals from Alaska compared to other Arctic regions (Dehn et al. 2006).

The observed geographic pattern of Hg levels in these polar bear studies may be related to both abiotic and biotic factors. High Hg levels in marine mammals from the Beaufort Sea may potentially be a consequence of Hg transported from the Mackenzie River (Leitch et al. 2007). However, geographic patterns in Hg concentrations of beluga, narwhals, and ringed seals have also been related to geological formations (Wagemann et al. 1996, Rigét et al. 2005). In addition, recent studies

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have pointed to the importance of food web structure and feeding behaviour on Hg concentrations in polar bears (Cardona-Marek et al. 2009, Horton et al. 2009) and other marine mammals including beluga whales (Loseto et al. 2008a) and various seal species (Young et al. 2010).

Liver THg concentrations for a subset of the same Alaskan, Canadian, and East Greenland bears collected from 2005 to 2008 were investigated in relation to differences in food web structure among the subpopulations (estimated using $\delta^{15}N$, $\delta^{13}C$ and fatty acid tracers in muscle) (Routti et al. 2012). Regardless of subpopulation, liver THg concentrations were positively related to $\delta^{15}N$ ratios. This indicates that polar bears feeding at a higher trophic level accumulate more THg compared to polar bears feeding at a lower trophic level. This finding is consistent with previous studies that reported trophic biomagnification of Hg for other Arctic fish and wildlife (Muir et al. 2005, Loseto et al. 2008b, Gaden et al. 2009). Routti et al. (2012) found that adjustment for trophic position reduced the geographic variation in liver THg concentrations among polar bear subpopulations (Fig. 6.33), and many differences in unadjusted THg levels among subpopulations were no longer statistically significant after trophic adjustment. Mean THg concentrations in polar bears were also adjusted to food web structure using several tracers ($\delta^{15}N$, $\delta^{13}C$ and fatty acids) and the variables sex and age. Food web adjusted mean THg concentrations in southern and northern Beaufort Sea polar bears were not higher compared to any other subpopulation ($0.14 < p < 1$; $p = 0.06$ for southern Hudson Bay versus southern Beaufort Sea) except Chukchi Sea ($p < 0.008$). Food web adjusted THg concentrations in polar bears from western Hudson Bay were similar to all the subpopulations ($0.44 < p < 1$) and higher than in Chukchi Sea polar bears ($p = 0.006$). Food web adjusted THg concentrations for the southern Hudson Bay subpopulation were significantly lower compared to polar bears from Baffin Bay, Lancaster/Jones Sound, and East Greenland, although these differences were close to the level of significance ($0.014 < p < 0.040$). Interestingly, liver THg concentrations were negatively correlated with muscle $\delta^{13}C$ ratios. This suggests polar bears that feed in areas with higher river inputs of terrestrial carbon may accumulate more Hg than bears feeding in areas with lower freshwater input. Liver THg concentrations were also positively related to a fatty acid (20:1n-9) that is biosynthesized in large amounts in *Calanus* copepods. The hypothesis was proposed that *Calanus* are an important link in the uptake of Hg in the food web and ultimately in polar bears (Routti

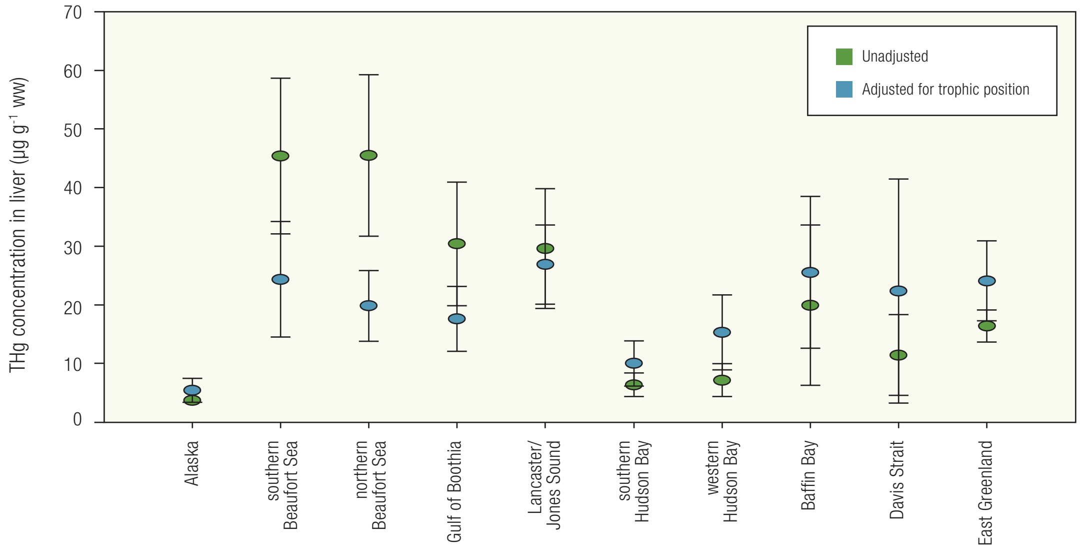

FIGURE 6.33

Geometric mean concentrations of THg (μg g^{-1} ww; ± 95% confidence intervals) in polar bear liver from ten subpopulations in Alaska (Chukchi and Bering seas), Canada, and Greenland collected from 2005 to 2008. All concentrations were adjusted for sex and age. In addition, THg concentrations were adjusted for trophic position using muscle δ^{15}N and compared to unadjusted concentrations (data from Routti et al. 2012).

et al. 2012). The findings of Routti et al. (2012) indicate that food web structure and dietary exposure have a significant influence on geographic trends of liver THg in polar bears.

St. Louis et al. (2011) similarly found that food web structure explained, in part, geographic variation in Hg bioaccumulation in polar bears. In that study, THg concentrations and nitrogen stable isotope ratios in polar bear hair indicated that southern Beaufort Sea bears bioaccumulated more Hg because they fed at a higher trophic position than bears from western Hudson Bay. Carbon stable isotope ratios suggested that Beaufort Sea bears fed more on pelagic than benthic carbon compared to Hudson Bay bears. McKinney et al. (2011) found carbon stable isotope evidence that, compared to other polar bear subpopulations, the diet of Hudson Bay polar bears may be more dependent on freshwater associated prey such as seals that inhabit rivers or estuaries. However, regional differences in polar bear Hg levels were also likely due to the pool of MeHg available for uptake into the food web because concentrations of methylated Hg mid-depth in the pelagic water column of the Canadian Arctic Archipelago were, on average, twice as high as in Hudson Bay (St. Louis et al. 2011). These findings indicate that food web length and structure combined with MeHg bioavailability are important factors in the interpretation of spatial Hg trends in polar bears, particularly in a changing Arctic.

Temporal trends

Recent temporal trends of THg levels in the livers of polar bears differed among populations in the circumpolar Arctic. In polar bears from the Beaufort Sea area, a slight increase in liver THg concentrations was observed between the 1980s and 2002 (Fig. 6.34) (Rush et al. 2008). However, Routti et al. (2011) reported that no further changes in THg levels were observed from 2002 to 2007 and 2008 for polar bears from the same Beaufort Sea subpopulations (Fig. 6.34). Results reported by Rush et al. (2008) indicated that a slight increase in THg concentrations occurred in bears from other subpopulations and areas of the Canadian Arctic sampled in 2002 relative to the 1980s. The most recent data show that THg levels in polar bears from Alaska (Chuckchi Sea) and Canadian Arctic subpopulations (except the Beaufort Sea) have declined relative to levels in 2002 (Fig. 6.34). In contrast, concentrations of THg in polar bear liver from East Greenland increased significantly in 2006 relative to measurements from 1999 and 2000 ($p < 0.05$) (Fig. 6.34). This finding was

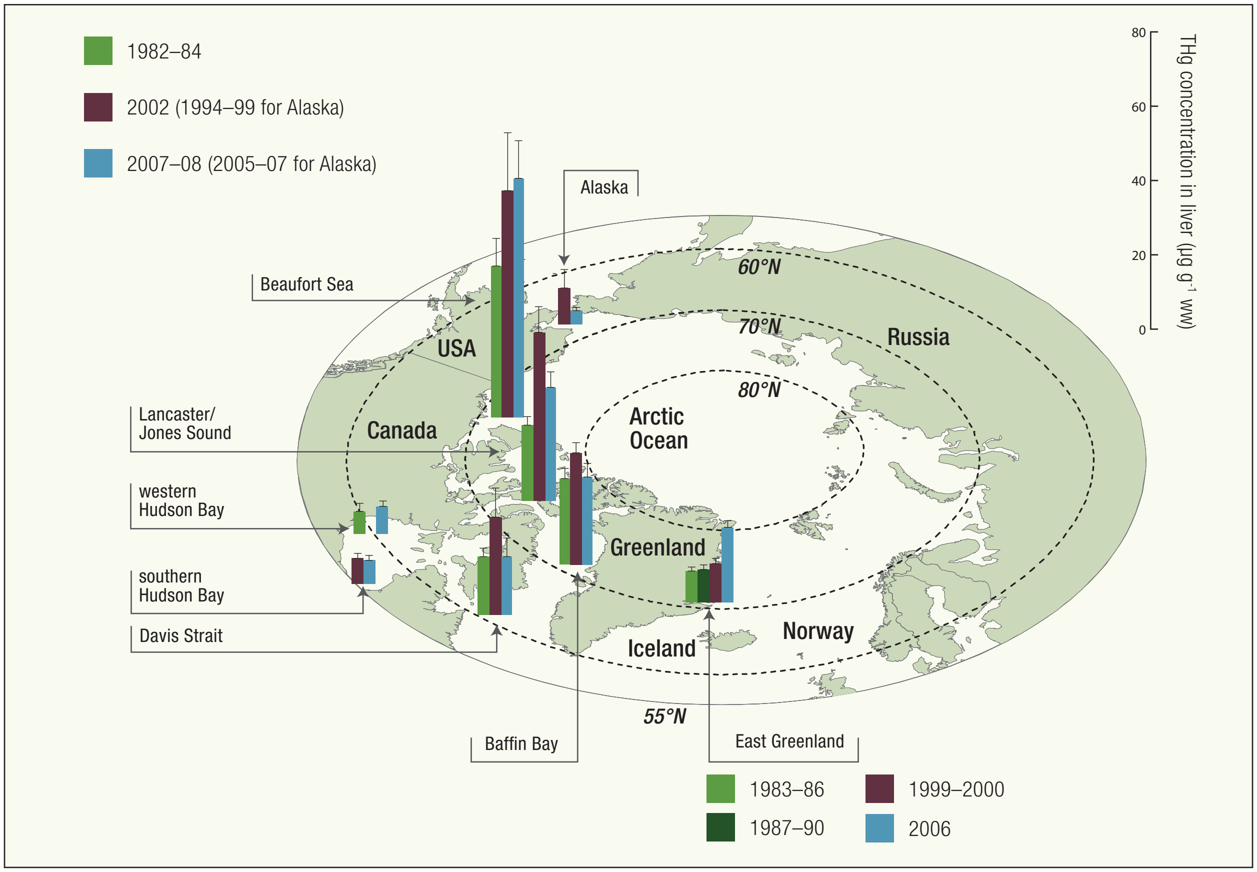

FIGURE 6.34

Concentrations (µg g^{-1} ww; geometric mean ± standard error) of THg in polar bear liver from Alaska (Chukchi and Bering seas) in 1994 to 2007, Canada in 1982 to 2008, and East Greenland in 1983 to 2006 (Routti et al. 2011 and references therein).

in agreement with increasing THg concentrations in polar bear hair from Northwest Greenland between 1992 and 2008 (Dietz et al. 2011), ringed seal teeth from Central, West, and East Greenland between 1994 and 2006 (Aubail et al. 2010), and ringed seal liver from central West Greenland during 1999 to 2004 (Rigét et al. 2007a). In contrast, Dietz et al. (2006) reported a decline in THg in polar bear hair from East Greenland between 1973 and 2001.

Overall, geographic trends of THg in polar bears appear to be related to Hg sources—potentially atmospheric deposition, river inputs, and/or local geology—and to polar bear diet. Temporal trends suggest that Hg concentrations in East and Northwest Greenland polar bears have recently increased. Future studies should focus on the role of changing climate and anthropogenic emissions on geographic and temporal trends of Hg in polar bears.

6.12 Summary

Much new information has been collected over the last decade on Hg concentrations in marine water, snow, and ice. In the seawater of the Arctic Archipelago and Hudson Bay, concentrations of methylated Hg were typically low in surface waters—often approaching the analytical detection limit—but increased with depth. This suggests that methylated Hg species are produced in deep Arctic marine waters and/or sediments. Ocean-atmosphere fluxes of GEM and Me_2Hg were high, and Arctic marine waters have been shown to be a substantial source of gaseous Hg to the atmosphere during the ice-free season. Inorganic Hg accumulation is enhanced in marine snow relative to the terrestrial environment. High Hg concentrations in marine snow may occur because AMDEs are a result of atmospheric photochemical reactions involving marine halogens and/or because of the effect of chloride on Hg(II) retention in snowpack. On Hudson Bay sea ice near Churchill, AMDEs were found to increase THg concentrations in surface snow but these concentrations declined rapidly within four or more days, likely resulting from re-emission of Hg to the atmosphere via photoreduction. In a study of ice in the Beaufort Sea, THg concentrations were consistently low in sea ice. However, brine in the sea ice was substantially more concentrated in THg, with higher levels found in more saline brines. Areal loads of THg in snow were estimated for Hudson Bay and the Arctic Archipelago, which suggest that snowmelt is likely a small flux of Hg to Arctic marine waters during spring.

The most extensive sampling of marine sediments in the Canadian Arctic was recently performed in Hudson Bay where sediment THg concentrations were low compared to other marine regions in the circumpolar Arctic. In most sediment cores, vertical profiles of sediment Hg fluxes increased during the early days of the Industrial Era (1870 to 1930), followed by stable or declining Hg inputs toward the end of the 20th Century. The flux patterns were consistent with general historical patterns of anthropogenic Hg emissions in North America. However, several other sediment cores in Hudson Bay displayed consistent increases in THg concentration in deep sediment accumulated before the Industrial Era. Natural changes in the materials incorporated into sediments were found to increase THg concentrations as much as those potentially caused by anthropogenic impact.

Mass balance models were developed for the Arctic Ocean and Hudson Bay to provide quantitative estimates of THg fluxes into and out of these ecosystems. For the Arctic Ocean as a whole, net atmospheric deposition had the largest estimated flux, followed by coastal erosion and oceanic transport. In contrast, rivers were identified as the largest single source for Hudson Bay followed in importance by the atmosphere and ocean inflow Hg fluxes. More work using a variety of methodological approaches is essential to resolve uncertainty in the atmospheric Hg contribution to Arctic ecosystems, especially since this flux would be most immediately affected by emission controls.

Biogeochemical processing of Hg in the Arctic marine environment remains poorly characterized. The first measurements of methylation rates in Arctic seawater suggest that the water column is an important site for MeHg formation from inorganic Hg. This observation challenges the paradigm that only anaerobic conditions support significant MeHg production. Production of Me_2Hg may occur in deep waters as suggested by recent water-column profiles in the Arctic Ocean, and its breakdown in the water column or in the air—after its evasion—may be a source of MeHg for biological uptake. Recent evidence from the Arctic suggests that microbes may be key players in altering Hg redox chemistry in salt water. The discovery of expressed Hg resistance genes suggests that microbial populations in the Arctic marine environment are sensitive to THg contamination and may be actively reducing this metal to its elemental form. These findings underscore the need for more fundamental research examining mechanistic aspects of Hg methylation and redox reactions in polar regions.

Biomagnification of Hg was observed in marine food webs from Cumberland Sound, Hudson Bay, and the Beaufort Sea and Amundsen Gulf. In addition to trophic position, habitat use and food web structure explained THg levels in beluga in the western Arctic. Furthermore, Hg bioaccumulation varied within the beluga population due to body size and sex segregation. Higher concentrations were observed in beluga and their associated food web in offshore waters compared to those feeding in an estuarine shelf near the Mackenzie River Delta. Although large amounts of Hg are transported by the Mackenzie River, Hg may not be readily bioavailable upon entering the estuary given the low Hg concentrations found in the estuarine food web.

The Hg content of teeth from Canadian Arctic marine mammals was used to estimate the long-term effect of global anthropogenic Hg emissions on Hg bioaccumulation. For ringed seals from the Amundsen Gulf and beluga from the Beaufort Sea and Somerset Island area, THg content increased several-fold in teeth collected in the early to mid-20th Century compared with teeth from the 19th Century and pre-industrial period. The greatest differences between historical and modern samples of seals and beluga were observed for older individuals. Walrus was the only species that did not show greater Hg in present-day teeth relative to pre-industrial samples.

Muscle of sea-run Arctic char consistently had very low concentrations of THg at locations across the Canadian Arctic. Little information exists for wholly marine fish species that may be important sources of dietary Hg for seabirds and large marine mammals such as seals and beluga. One of these keystone prey species, Arctic cod, has been sampled in recent food web studies where fish in offshore waters had higher THg concentrations than those in an estuarine shelf. The factors that determine geographic and habitat variation of Hg bioaccumulation in marine fish remain unresolved and warrant future investigation to better understand Hg transfer to top predator animals in the marine environment.

Spatial analyses of THg concentrations in marine biota indicated that ringed seals, beluga, and polar bears contained more Hg in the Beaufort Sea region than in other parts of the Canadian Arctic, particularly in comparison to Hudson Bay, Nunavik, and Nunatsiavut. Recent evidence indicates that higher THg in Beaufort Sea polar bears is due to their higher trophic position and higher concentrations of MeHg in water available for entry into the food web. Although the Beaufort Sea was consistently a region of high bioaccumulation, additional geographic variability was observed among species. Muscle THg concentrations in ringed seals were also elevated near some High Arctic communities. A latitudinal difference in THg levels was found among colonies of thick-billed murres, with birds breeding at the higher latitude colonies having higher THg concentrations than those breeding at sub-Arctic colonies. Inter-species differences in THg bioaccumulation were also observed among seabirds at a High Arctic site on Prince Leopold Island. Further research is needed to link biogeochemical and food web processes with geographic variation in Hg levels of Arctic biota.

Temporal trends of Hg in marine biota differed among species and locations in the Canadian Arctic. Monitored populations of ringed seal near five Arctic communities, narwhal from the Baffin Island region, and walrus from Igloolik showed no temporal change in THg concentration. For beluga from the western Canadian Arctic, Hg concentrations in muscle appear to have declined since the last *CACAR* evaluation in 2002. Significant increases in THg concentration were observed in recent years for sea-run char collected near three Arctic communities and since 1975 for eggs of three Arctic-breeding seabird species at Prince Leopold Island. Shifts in prey due to climate change are affecting the temporal THg trend in eggs of thick-billed murres breeding in northern Hudson Bay, which also showed an increase in THg over the last two decades after the concentrations were adjusted for trophic position. Similarly, inter-annual variation in ice conditions may be affecting the dietary exposure of Hg to seals. Recent temporal THg trends in polar bears differed among populations in the circumpolar Arctic including a decline in Alaska (Chukchi and Bering seas), stable concentrations over the last decade in Hudson Bay, and increases between the 1980s and post-2000 in the Beaufort Sea and Lancaster/Jones Sound. Overall, the different Hg trends reported for Arctic marine biota indicate that the drivers of such change may be regional or habitat-specific. Particularly at sites where increases were observed, further research is required to identify the underlying processes leading to changes over time in marine biota Hg bioaccumulation.

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CHAPTER 7

Biological Effects in Fish and Wildlife

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7.1 Introduction

From an environmental toxicology perspective, methylmercury (MeHg) is the most important of the different chemical forms of Hg. Methylmercury biomagnifies through food chains, is very efficiently absorbed from the diet, distributes efficiently into many organs of the body including the brain, and is highly toxic. The toxic effects of MeHg in wildlife have been reported and scientifically studied for over 50 years, during which time much has been learned about its food chain transfer, metabolism, and toxicity (Wiener et al. 2003). In recent years, increasingly subtle but important biological effects have been documented, including behavioural, neurochemical, hormonal, and reproductive changes in predatory fish and wildlife exposed to environmentally relevant levels of MeHg (Scheuhammer et al. 2007, Scheuhammer et al. 2012). Potential population-level impacts are now being assessed for some species, such as the common loon (Burgess and Meyer 2008). However, specifically for Arctic wildlife species, there is a general paucity of information regarding the effects of MeHg exposure.

Because MeHg biomagnifies through food webs, it is generally agreed that top predatory animals, particularly those linked to aquatic food chains, are at greatest risk for increased dietary MeHg exposure and potential Hg-related health effects. In the Arctic, species at greatest risk include polar bears, seals, toothed whales, various predatory seabirds, and large piscivorous fish such as lake trout, pike, and sharks. Conversely, Hg levels are generally far below those required to cause toxic effects in lower trophic level animals and in Arctic terrestrial

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animals not associated with aquatic food webs. Although there is evidence to suggest that non-aquatic birds, such as some forest passerine species, can experience elevated dietary MeHg exposure in sites near Hg-contaminated waterways (Brasso and Cristol 2008, Cristol et al. 2008), similar scenarios are less likely to occur in the Canadian Arctic where large-scale industrial activity is less prevalent. Data on Hg in insectivorous passerines in the Arctic are currently lacking, and concentrations in other terrestrial avian species (e.g., ptarmigan; section 4.8.3) are low compared to aquatic predatory species. In this chapter, we focus on freshwater and marine species and summarize findings from recent studies that have begun to investigate potential toxic effects of Hg in Arctic wildlife.

7.2 Mercury effects studies

7.2.1 Freshwater and marine fish

Studies published within the last decade have documented a range of toxic effects in fish at environmentally relevant levels of MeHg exposure. In a critical review of the recent literature, Sandheinrich and Wiener (2011) concluded that changes in biochemical processes, damage to cells and tissues, and reduced reproduction in fish begin to occur at concentrations of about 0.5–1.0 µg THg g^{-1} wet weight (ww) in axial muscle (> 90% of THg in muscle is MeHg) and, below this range, there is little if any indication of harm to fish from Hg exposure. Similarly, Dillon et al. (2010) conducted an assessment of numerous fish toxicology studies and estimated with a mathematical model a lowest observable adverse effects level (LOAEL) of about 0.3 µg THg g^{-1} ww in the whole body of fish—or about 0.5 µg THg g^{-1} ww in axial muscle. Using 0.5–1.0 µg THg g^{-1} ww in axial muscle as the estimated LOAEL range for fish, it is apparent that lake-averaged THg concentrations in northern Canadian freshwater fish species sampled since 2002 seldom enter or exceed this range (Fig. 7.1). However, average THg concentrations in landlocked char from Amituk Lake on Cornwallis Island and lake trout from Cli Lake in the Northwest Territories clearly exceeded the suggested LOAEL range. Concentrations were within the suggested threshold range for landlocked char from Char Lake (Nunavut), as well as for other fish species in several lakes in the Northwest Territories, specifically northern pike and whitefish from Narrow Lake, and lake trout from Kelly Lake and Lac Ste. Therese. A larger, older database of fish Hg levels measured in the Canadian Arctic from 1971 to 2001 (Lockhart et al. 2005a) showed that length-adjusted mean THg concentrations in highly predatory species—lake trout, walleye, northern pike and burbot—were greater than 0.5 µg g^{-1} in < 25% of the survey lakes (*n* = 29–94 lakes per species). Research is warranted to study possible reproductive or other toxic effects of Hg in fish in Arctic lakes where the LOAEL threshold is exceeded.

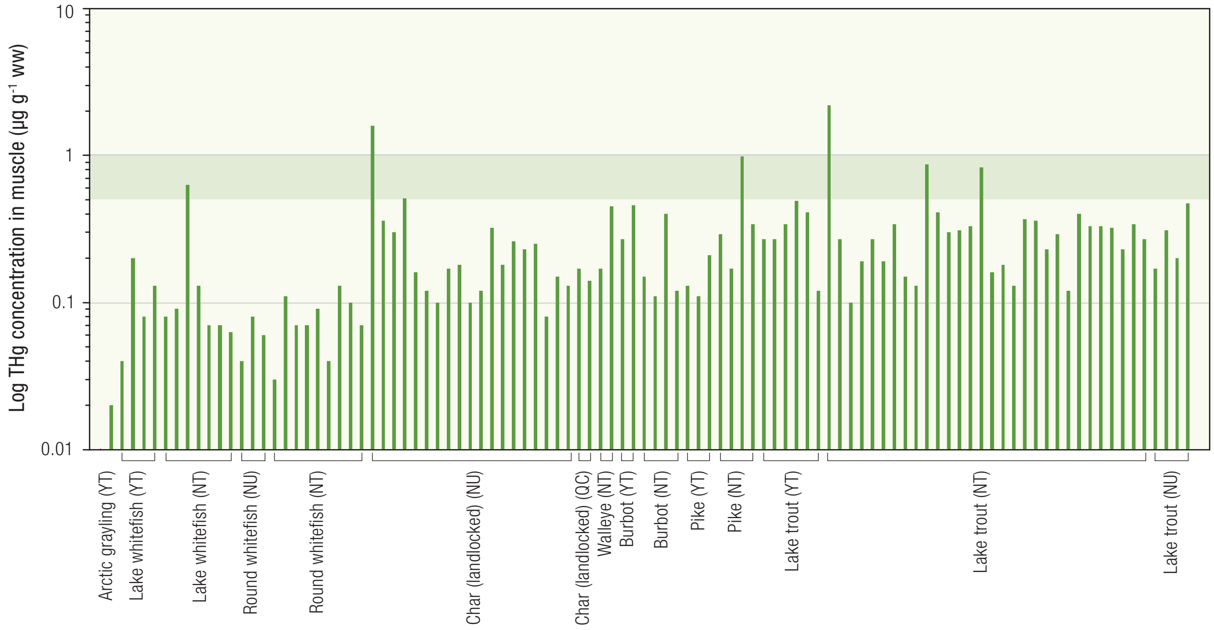

FIGURE 7.1

Mean concentrations of THg in muscle of selected freshwater fish species from various Canadian Arctic locations. The shaded area represents the range of LOAELs for fish toxicity as suggested by Sandheinrich and Wiener (2011). Data are from Depew (2011) and are presented in Annex Table A2.2.

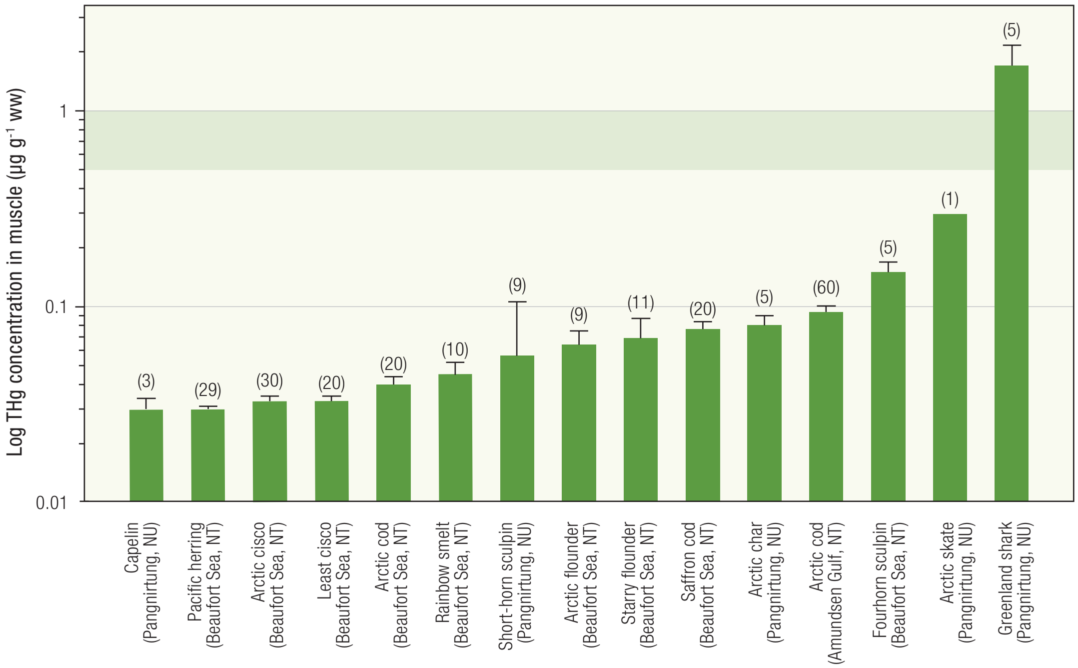

FIGURE 7.2

Mean concentrations of THg in muscle of selected marine fish species from various Canadian Arctic locations. Bracketed values are sample sizes. The shaded area represents the estimated range of LOAELs for freshwater fish toxicity as suggested by Sandheinrich and Wiener (2011). Data are from Loseto et al. (2008) for fourhorn sculpin, least cisco, Pacific herring, rainbow smelt, saffron cod and starry flounder; and from McMeans and Fisk (2010) for capelin, shorthorn sculpin and Greenland shark. Dry weight data from Loseto et al. (2008) were converted to ww based on an estimate of 75% water content for fish muscle.

Consistent with fish data reported in Dietz et al. (2011) for the circumpolar Arctic, muscle THg concentrations in many common marine fish tend to be substantially lower than in freshwater fish. As shown in Figure 7.2, mean muscle THg levels in most marine fish species from the Canadian Arctic were many-fold lower than the suggested LOAEL range. The single exception was the Greenland shark, a large long-lived predatory species for which greater bioaccumulation and thus higher THg concentrations are expected. Nevertheless, based on elevated muscle THg concentrations, studies are warranted to investigate possible toxic effects of Hg in sharks and other large carnivorous marine fish species in the Canadian Arctic.

7.2.2 Marine birds

Reproduction

Few effects studies have been undertaken on Arctic bird species. However, recent research on common loons serves to illustrate the kinds of population-level effects that environmentally relevant exposures to MeHg can have on wild fish-eating birds. A strong positive relationship between female blood Hg and egg Hg concentrations was used to demonstrate a link between blood Hg and various adverse reproductive effects in common loons (Evers et al. 2003). By integrating this and other relationships into a population matrix model, Evers et al. (2008) were able to effectively predict and identify North American loon populations that are experiencing reduced fledging success associated with elevated MeHg exposure. In Maine and New Hampshire, MeHg toxicity was the main factor identified as responsible for a 41% decline in average reproductive success over an 11-year period, with a LOAEL identified as 3.0 µg g^{-1} (ww) in blood (Evers et al. 2008, Scheuhammer et al. 2012). Based on comparable findings from two parallel studies using similar field protocols in New England (Evers et al. 2008) and in Wisconsin and the Canadian Maritimes (Burgess and Meyer 2008), maximum productivity for breeding loon pairs declined by at least 50% when whole-body concentrations of THg in prey fish exceeded about 0.2 µg g^{-1} ww.

Dietary MeHg is efficiently transferred to avian eggs in a dose-dependent manner, and reproduction is one of the most sensitive endpoints of Hg toxicity in birds

(Wolfe et al. 1998). Nearly 100% of the THg transferred to eggs is in the form of MeHg with the majority (about 85–95%) deposited into the albumen (Wiener et al. 2003). Mercury concentrations found in the egg are a good indicator of Hg risk to avian reproduction (Wolfe et al. 1998). Some of the documented effects of MeHg on avian reproduction leading to substantial overall reductions in productivity include aberrant reproductive behaviour, reduced clutch sizes, increased rates of embryonic deformity and mortality, and reduced hatchability (Thompson 1996, Wolfe et al. 1998). Dietary MeHg exposure, insufficient to cause obvious signs of Hg toxicity in adults, can decrease reproductive success by 35–50% in birds (Wolfe et al. 1998).

Embryotoxic thresholds for Hg have been determined for a limited number of species—primarily from captive breeding studies—and are often applied generically to all avian species. However, based on a study involving 26 avian species, Heinz et al. (2009) showed that there are significant interspecies differences in sensitivity to the embryotoxic effects of MeHg. Using estimated median lethal concentrations (LC_{50}) for 26 tested species, Heinz et al. (2009) grouped the sensitivity of avian embryos to MeHg into three categories with LC_{50} values ranging from ≥ 1 $\mu g\ g^{-1}$ ww in eggs of the low sensitivity group (e.g., Canada goose, hooded merganser, laughing gull, double-crested cormorant) to < 0.25 $\mu g\ g^{-1}$ ww in eggs of those species exhibiting the highest sensitivity (e.g., American kestrel, osprey, snowy egret, tri-coloured heron). Species, such as common, royal, and Caspian terns, as well as herring gulls, were categorized as having medium sensitivity to MeHg based on a calculated LC_{50} ranging between 0.25 and 1 $\mu g\ g^{-1}$ ww of Hg.

Using the egg-dosing protocol developed by Heinz et al. (2006), eggs of thick-billed murres and Arctic terns collected from the High Arctic were brought into the laboratory and injected with environmentally relevant, graded concentrations (0–6.4 $\mu g\ g^{-1}$ ww) of MeHg chloride (MeHgCl) to determine the relative sensitivity of the developing embryos to MeHg (Braune et al. 2012). Roughly half of the murre eggs (48%) and tern eggs (62%) reached the chosen endpoint of at least 90% development. To compare the murre and tern data with the results for 26 species reported by Heinz et al. (2009), the SAS version 9.2 probit procedure (SAS Institute, Cary, NC) was used to estimate a median lethal concentration (LC_{50}) and 95% confidence interval (CI) for each species with the survival data corrected for control mortality. The LC_{50} for the murre embryos was 0.48 $\mu g\ g^{-1}$ ww (95% CI: 0.26–0.99) based on MeHg injected into eggs uncorrected for maternally deposited MeHg. For the tern embryos, the LC_{50} was 0.95 $\mu g\ g^{-1}$

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ww (95% CI: 0.59–1.58) (Fig. 7.3). The wide 95% confidence intervals for the LC_{50} estimates suggest a lack of power in these tests. Nonetheless, based on these LC_{50} estimates, both the Arctic tern and thick-billed murre embryos would be categorized as having medium sensitivity to MeHg according to the sensitivity categories suggested by Heinz et al. (2009). This estimate places the Arctic tern in the same sensitivity category as three other tern species—common, royal, Caspian—included in the study by Heinz et al. (2009).

In comparison with eggs in the wild, the average THg concentration in thick-billed murre eggs collected from the Prince Leopold Island colony in 2009 was 0.40 µg g^{-1} ww, and for the Coats Island colony was 0.16 µg g^{-1} ww (Braune et al. 2012). Concentrations of THg measured in first-laid (early) Arctic tern eggs collected in 2008 from Tern Island, just north of Cornwallis Island, averaged 0.49 µg g^{-1} ww (Akearok et al. 2010). However, Heinz et al. (2009) cautioned that MeHg injected into the egg may be two to four times more embryotoxic than maternally deposited MeHg. In addition, it should be noted that results in Figure 7.3 have not yet been corrected for maternally deposited Hg, which would increase the Hg concentrations at which effects were observed.

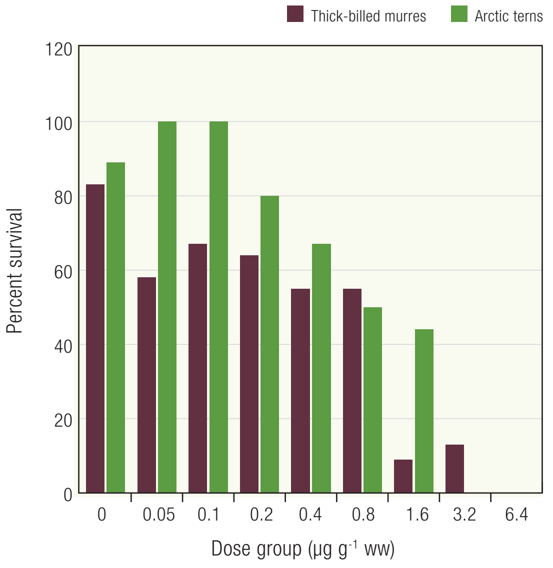

FIGURE 7.3

Percent survival to 90% of embryo development of thick-billed murre and Arctic tern eggs dosed with MeHgCl at 0.05–6.4 µg g^{-1} ww (Braune et al. 2012). Dose group concentrations are not corrected for maternally deposited Hg. For each species, 12 eggs were tested per dose group. The control eggs were injected with undosed safflower oil.

Neurochemical receptors

Concentrations of neurochemical receptors in the brain, such as for the neurotransmitters acetylcholine (muscarinic [mACh] receptor) and glutamate (N-methyl-D-aspartic acid [NMDA] receptor), can be significantly altered by low-level dietary exposure to MeHg in adult birds and mammals. Thus, specific neurochemical changes may potentially be used as biomarkers of MeHg exposure and effects in wildlife (Basu et al. 2006, 2007a, Scheuhammer et al. 2008). However, in the aforementioned egg injection study, brain tissue from thick-billed murre and Arctic tern embryos and hatchlings showed no significant correlations between Hg concentrations and densities of either the NMDA or mACh neuroreceptor for the control and dose groups assayed (Hg concentrations: 0–3.2 µg g^{-1} ww for the murre embryos and 0–1.6 µg g^{-1} ww for the tern embryos) (Braune et al. 2012). Reasons for this apparent lack of response of embryonic neuroreceptors to Hg exposure *in ovo* are currently unknown.

Oxidative stress

Exposure to contaminants, including MeHg, can lead to oxidative stress in wildlife, where the presence of excessive reactive oxygen species results in cellular damage (Kenow et al. 2008, Hoffman et al. 2011). Glutathione (GSH) is an important low molecular weight tripeptide involved in protecting cells from oxidative stress. A reduction in levels of the reduced form (GSH) and an increase in oxidized glutathione (GSSG) have been interpreted as indicators of greater risk for biological damage from oxidative stress (Hoffman et al. 2002). Wild diving ducks with higher liver THg concentrations also had higher GSSG:GSH ratios (Hoffman et al. 1998). Although no critical ratio of GSSG:GSH has been proposed as an unequivocal indicator of oxidative damage, Hoffman (2002) suggested that significant elevations of this ratio in contaminant-exposed animals, compared with unexposed or reference animals, are indicative of oxidative stress, including an increased risk of cellular lipid peroxidation. Wayland et al. (2010) examined relationships between THg levels and indicators of oxidative stress in livers of nestling glaucous gulls in Nunavut at Karrak Lake in the Queen Maud Gulf Bird Sanctuary and Devil Island. In that study, hepatic THg levels were relatively low (0.1–4 µg g^{-1} dry weight [dw]) compared with lower-latitude field studies reporting on Hg exposure and oxidative stress in birds.

In Arctic glaucous gull chicks, liver concentrations of both reduced GSH and oxidized GSSG were negatively correlated with THg concentrations (Fig. 7.4). This pattern was unexpected because decreased GSH is typically accompanied by increased GSSG, which is diagnostic of oxidative stress. Wayland et al. (2010) suggested that the observed decrease in GSSG may have resulted from low availability of precursor compounds such as cysteine or glutamate that could potentially occur if the animals were food-stressed. This explanation is consistent with measured levels of thiols (protein-bound and total sulfhydryls) in the gull livers, which were negatively correlated with THg concentrations. In addition, as suggested by a positive correlation between hepatic concentrations of THg and thiobarbituric acid-reactive substances, Hg-related oxidative stress in birds from Devil Island may have been high enough to cause increased lipid peroxidation. However, overall evidence for a shift in cellular status to a more oxidized state—decreased GSH, increased GSSG, or elevated GSSG:GSH ratio—was weak, leading to the conclusion that the glaucous gull nestlings at the two colonies were exposed to lower levels of oxidative stress than birds in more highly contaminated environments.

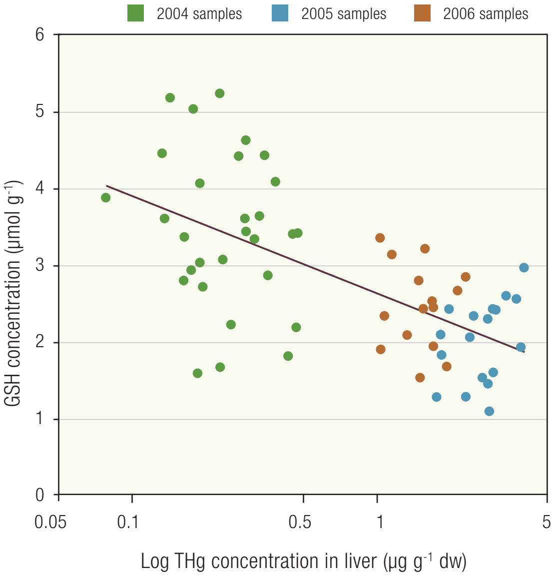

FIGURE 7.4

Relationship between log-transformed hepatic THg (µg g^{-1} dw) and reduced glutathione (GSH) concentrations (µmol g^{-1}) in glaucous gull chicks from two sites in the Canadian Arctic ($r^2 = 0.32$, $p < 0.001$). Hepatic GSH concentration was adjusted to a common measurement of body size and the relationship is shown across all years (source: Wayland et al. 2010).

Comparisons with Hg toxicity thresholds

Recent egg THg concentrations for marine birds from the Canadian Arctic were compared against suggested avian toxicity thresholds of 0.6 µg g^{-1} ww, proposed as an *indicative value* protective for most avian species, and to the species-specific range of 0.8–5.1 µg g^{-1} ww, at which adverse effects on reproduction occur in various avian species (Shore et al. 2011). The thresholds are based on a recent evaluation of published field and laboratory studies for non-marine birds (Shore et al. 2011) and are applied here because no evaluation of Hg threshold levels is available specifically for marine birds. Only the mean egg THg concentration for ivory gulls from Seymour Island exceeded the proposed indicative value (0.6 µg g^{-1} ww), and entered the threshold range of concentrations that are associated with adverse effects on reproduction (Fig. 7.5). Additional research to assess possible Hg-associated reproductive effects in this species is thus warranted. Some individual egg THg values reported for black guillemots, glaucous gulls, and Arctic terns also exceeded the proposed indicative value (Fig. 7.5).

Liver THg concentrations of Arctic marine birds were also compared to toxicity thresholds derived from the literature by Shore et al. (2011) for adverse effects on reproduction in non-marine birds (2–52 µg g^{-1} ww) and for lethality (18.4–127 µg g^{-1} ww). Levels of THg in some of the northern fulmars exceeded the minimum liver threshold for potential adverse effects on reproduction. However, all measured liver levels were well below the threshold range for lethality (Fig. 7.6). It should be noted that adverse-effects thresholds based on THg concentrations in liver implicitly assume that knowledge of the major chemical forms of Hg present in the tissue is largely unimportant—an assumption that may not be valid especially for long-lived species that can have substantial proportions of their liver Hg as a non-toxic complex of inorganic Hg associated with Se (section 7.3).

Burger et al. (2009) suggested that seabirds may be less vulnerable to Hg than birds that feed in other environments. Thompson (1996) suggested that pelagic seabirds have yet to be exposed to sufficiently high burdens of Hg to induce measurable effects on reproduction or survival. However, there is little published evidence that seabirds differ fundamentally from other birds with respect to the metabolism of MeHg, or in their sensitivity to MeHg toxicity. If seabirds are less sensitive to MeHg exposure, this may be related to more efficient demethylation of MeHg in these species.

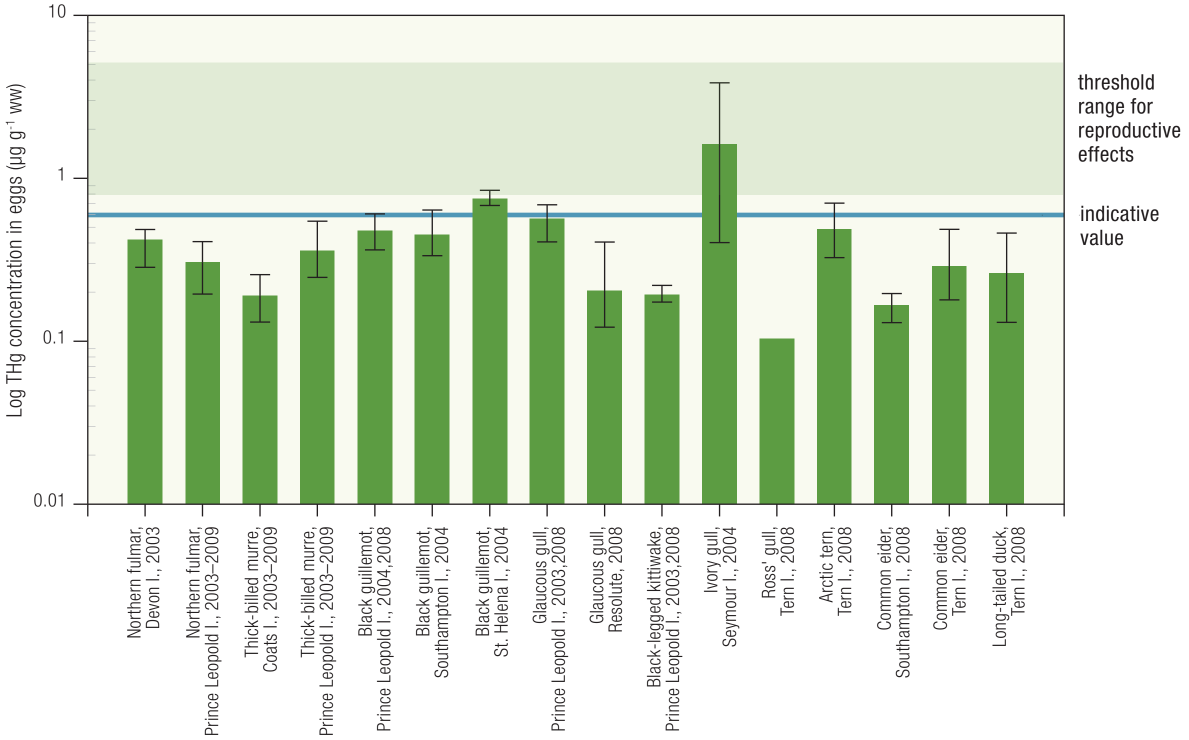

FIGURE 7.5

Mean concentrations and range bars of THg in marine bird eggs from the Canadian Arctic (data in Annex Table A2.5). The range of threshold THg concentrations associated with adverse effects on reproduction (indicated by the green horizontal stripe) is based on an evaluation of published studies for a variety of non-marine bird species (Shore et al. 2011). The *indicative value* (blue line) is defined as the egg THg concentration below which 95% of species are protected against Hg-induced reproductive toxicity (Shore et al. 2011).

However, this is largely a speculative statement rather than one based on direct evidence. Although the capacity for demethylation does appear to vary among bird species (Kim et al. 1996, Eagles-Smith et al. 2009), there is no direct evidence that seabirds demethylate MeHg more efficiently than other species. Indeed, common loons, which could be considered seabirds during the wintering season when they primarily occupy marine habitats, showed less apparent demethylation of MeHg in their brains than bald eagles (Scheuhammer et al. 2008). Breeding bald eagles experience greater Hg exposure through feeding in freshwater lakes and rivers than in marine or estuarine areas (Evers et al. 2005), and small marine fish often have lower Hg concentrations than freshwater species (see Figs. 7.1 and 7.2). This indicates that marine environments may not pose as great a risk for dietary MeHg exposure in fish-eating birds as some freshwater environments, especially Hg sensitive environments where hydrologic and chemical conditions favour microbial Hg methylation and biomagnification.

Numerous bird species that breed in Canada are experiencing population declines. The ivory gull has been listed as *endangered* and the Ross's gull has been listed as *threatened* in Canada (COSEWIC 2010). Both species are associated with polar environments, and the Ross's gull is the rarest breeding gull in North America (Mallory et al. 2006). There are reports that Arctic tern populations may also be experiencing declines (Hatch 2002). The THg concentration recorded for a single salvaged Ross's gull egg was relatively low. However, relative to other species, egg THg levels for ivory gulls and Arctic terns were high (Fig. 7.5). Although there is no conclusive evidence to date that Hg exposure is resulting in adverse biological effects in marine birds in the Canadian Arctic, it is generally acknowledged that it is difficult to attribute population-level impacts to single specific factors and that a multiple-stressor approach may be more appropriate. Additional research is needed to determine if, and to what extent, MeHg exposure, combined with other stressors such as climate change, is affecting ivory gulls or other Arctic marine bird populations.

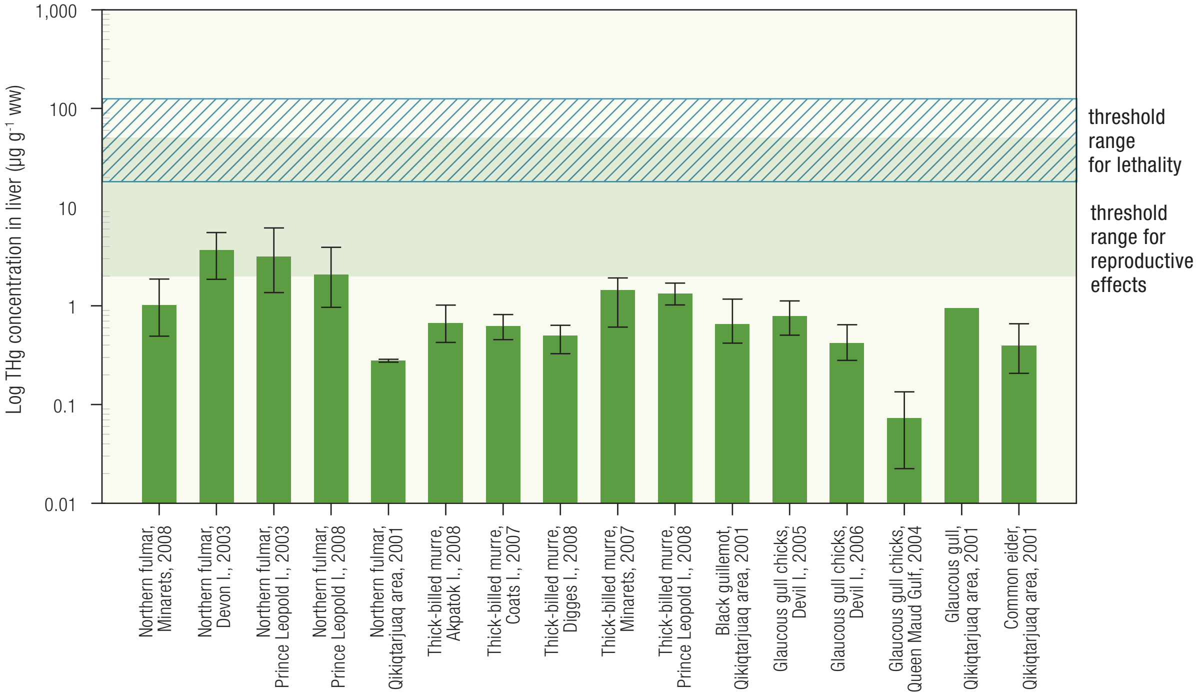

FIGURE 7.6

Mean concentrations and range bars of THg in marine bird livers from the Canadian Arctic (data in Annex Table A2.5). The range of threshold THg concentrations associated with adverse effects on reproduction (indicated by the green horizontal stripe) and range of thresholds associated with lethality (indicated by the diagonal blue lines) are based on evaluations of literature studies for non-marine birds (Shore et al. 2011). It should be noted that implicit in the concept of a toxic threshold concentration in liver is the notion that most of the liver Hg is present as MeHg. However, this assumption may not be correct, especially for older individuals.

7.2.3 Marine mammals

7.2.3.1 Neurotoxicity

The central and peripheral nervous systems are generally considered primary targets for MeHg toxicity because MeHg is efficiently absorbed from the diet and can readily pass the blood-brain barrier (Aschner and Aschner 1990). Characteristic lesions of MeHg poisoning in mammals include structural degeneration of the occipital cortex and the cerebellum, as well as degeneration of spinal cord and peripheral nerve fibers leading to ataxia (incoordination and loss of balance), weakness, tremors, convulsions, sensory impairment, and ultimately death (Heinz 1996, Wiener et al. 2003).

Mercury toxicity thresholds in brain tissue have not been specifically determined for most species of environmentally exposed wildlife and certainly not for Arctic species. However, a review of several published studies provides a range of THg and/or MeHg concentrations in brain tissue that can be associated with various degrees of neurotoxicity in different mammalian species exposed to controlled doses of MeHg via their diets. Concentrations of THg in brain tissue can be converted from a ww to dw value by multiplying the ww concentration by four. Various older reports of MeHg poisoning in free-living wild mammal species indicate that brain THg concentrations > 10 µg g^{-1} ww (or > 40 µg g^{-1} dw) are often associated with outright mortality (Wiener et al. 2003). Suzuki (1979) considered brain THg concentrations < 1.5 µg g^{-1} ww (or approximately 6 µg g^{-1} dw) as generally insufficient to cause MeHg neurotoxicity in mammals.

As a widely distributed predatory species which feeds mainly on fish and small mammals, mink are considered to be sensitive indicators of environmental Hg bioavailability (Kucera 1983, Wren et al. 1986). Captive mink fed diets dosed with MeHg (1.1, 1.8, 4.8, 8.3, or 15.0 µg g^{-1} Hg by analysis) over three months showed histopathological lesions in brain tissue, accompanied by manifestations of clinical toxicity including anorexia and loss of coordination (Wobeser et al. 1976). Brains of captive mink that died averaged 11.9 µg g^{-1}. Wobeser et al. (1976) concluded that brain THg concentrations > 5 µg g^{-1} ww (or > 20 µg g^{-1} dw) in mink were consistent with overt MeHg neurotoxicity in this species. It may be assumed that the majority of THg in brains of the captive mink fed MeHg-containing diets was in the form of MeHg because that is the dominant chemical form reported in wild mink (Haines et al. 2010). In adult rats chronically exposed to dietary MeHg (0.25 mg kg^{-1} day^{-1} dosage), THg concentrations in the cerebellum averaged 12 and 7.3 µg g^{-1} ww in males and females, respectively, and were accompanied by loss of balance, paralysis, and peripheral nerve damage (Munro et al. 1980). Similarly, brain Hg concentrations associated with neurotoxicity in cats dosed with MeHgCl or MeHg-contaminated fish were 16.3 µg g^{-1} ww (THg) and 10.9 µg g^{-1} ww (MeHg) in the cerebellum and 10.9 µg g^{-1} ww (THg) and 7.9 µg g^{-1} ww (MeHg) in the posterior cerebral cortex. These brain concentrations in cats were associated with movement disorders including a loss of balance and lack of coordination (ataxia) (Charbonneau et al. 1976). Taken together, dietary MeHg dosing studies on a variety of mammalian species indicate that brain Hg concentration > 10 µg g^{-1} ww can be associated with lethality (Shore et al. 2011), and that concentrations below approximately 5 µg g^{-1} ww, as suggested by earlier researchers (Wobeser et al. 1976, Suzuki 1979), are likely below thresholds of overt MeHg intoxication in most mammals.

A number of biochemical changes in the brain are associated with much lower concentrations of MeHg (< 10 µg g^{-1} dw or < 2.5 µg g^{-1} ww) than are required to produce overt signs of neurotoxicity. The following receptors and enzymes have been found to vary with Hg exposure in wild bird and/or mammal species:

- NMDA receptor levels decreased with increasing Hg levels (THg and MeHg) in polar bears from East Greenland collected between 1999 to 2001 (Basu et al. 2009), wild mink (Basu et al. 2007a), loons, and eagles (Scheuhammer et al. 2008);
- levels of mACh receptor increased with Hg levels in wild mink (Basu et al. 2005a), loons, and eagles (Scheuhammer et al. 2008) but the opposite relationship was found in river otters (Basu et al. 2005b);
- gamma-aminobutyric acid (GABA) receptor levels decreased with increasing Hg levels (THg and MeHg) in river otters (Basu et al. 2005a);
- dopamine D2-receptor levels were negatively correlated with THg levels in wild river otters and wild mink (Basu et al. 2005a, 2005b); and
- monoaminoxidase (MAO) and acetylcholinesterase (ChE) activities were negatively correlated to brain Hg in wild river otters (Basu et al. 2005b, 2007b).

Of these neurochemical markers, changes in NMDA and mACh receptor levels have proven to be among the most sensitive and robust correlates of brain Hg accumulation, and have thus been proposed as potential preclinical indicators of neurotoxic changes associated with MeHg exposure (Manzo et al. 1996, Basu et al. 2005b). Captive mink exposed to a range of dietary MeHg (0.1–2 µg g^{-1}) experienced subtle but significant changes in these neurochemical parameters at brain Hg concentrations between about 1–8 µg g^{-1} dw, levels that were not associated with overt signs of MeHg intoxication (Basu et al. 2006, 2007b, 2010). An average brain THg concentration as low as 1.5 ± 0.34 µg g^{-1} dw was associated with a significant decrease in NMDA receptor density in the cerebellum and occipital cortex of captive mink (Basu et al. 2007b). Although there are differences in sensitivity between rat, mink, mouse, and human with respect to Hg-induced neurochemical changes (Basu et al. 2005c), 1.5 µg g^{-1} dw may be considered a conservative mammalian LOAEL for Hg-induced neurochemical disruption. The average THg concentration reported by Gamberg et al. (2005) for brain tissue in wild mink from the Yukon (0.96 µg g^{-1} dw, or 0.22 µg g^{-1} ww) is below this proposed threshold for MeHg-induced neurochemical change. However, Basu et al. (2009) reported a significant negative association between brain THg and NMDA receptor concentrations in the lower brain stem of Greenlandic polar bears with an even lower range of THg (approximately 0.1–1.0 µg g^{-1} dw). Given the very low Hg concentrations that were associated with significant neurochemical effects in polar bears (Basu et al. 2009), it is suggested that additional studies be undertaken to test this finding.

In Canadian polar bears, no significant association was found between MAO activity or mACh receptor binding activity and THg (range: 0.12–0.47 µg g^{-1} dw; *n* = 36) or Se:Hg molar ratio in cerebellum (Krey and Chan 2010). This is consistent with the results of Basu et al. (2009) in brain stem tissue from Greenlandic polar bears. Although Basu et al. (2009) showed a significant association between THg and NMDA receptor density in brain stem, this biomarker was not measured in cerebellum of Canadian polar bears.

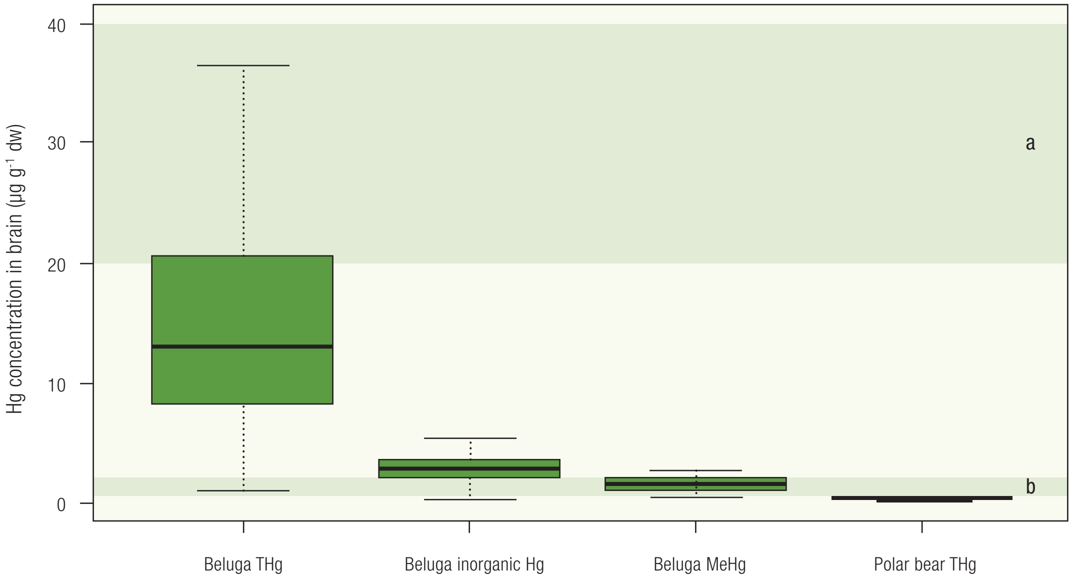

FIGURE 7.7

Concentrations of THg, soluble inorganic Hg (not including inorganic Hg that may be bound with Se in insoluble compounds), and MeHg in polar bear cerebellum from Nunavik (northern Quebec) harvested from 2000 to 2003 (*n* = 24) and in adult beluga whale cerebellum from the western Canadian Arctic harvested in 2008 (*n* = 21) (Krey and Chan 2010, Ostertag and Chan 2010). The concentrations are compared with effects levels identified in laboratory studies: a) approximate MeHg threshold range for overt MeHg poisoning and death; and, b) approximate MeHg threshold range for neurochemical change. The boxplots identify the 10th and 90th percentiles (error bars), the 25th and 75th percentiles (box) and the median concentration (bold line).

Concentrations of MeHg in polar bear cerebellum from the Canadian Arctic (range: 0.13–0.45 µg g^{-1} dw; *n* = 22) were lower than concentrations observed to be toxic in animal feeding trials. However, concentrations of MeHg in beluga whale cerebellum were sufficiently high to potentially cause significant neurochemical changes, but probably not high enough to cause overt MeHg neurotoxicity (Fig. 7.7). The formation of relatively inert Hg selenide compounds in the brains of beluga may provide a mechanism to reduce the risk of MeHg toxicity in these animals (see section 7.3). It should also be noted that different brain regions may react differently with respect to neurochemical changes in response to MeHg exposure (Basu et al. 2007a, 2010).

As liver is the most common tissue for monitoring environmental contaminant exposure in wildlife, researchers frequently attempt to establish toxicity thresholds for contaminants based on a consideration of the lowest liver concentrations that are associated with significant toxic effects in individual animals. For Hg in non-marine mammals, the most commonly cited threshold is probably that of Thompson (1996)—and recently updated by Shore et al. (2011)—of 25–30 µg g^{-1} ww THg in liver above which animals are likely to experience MeHg intoxication and death. Unfortunately, without additional information on the proportions of MeHg and inorganic Hg in the liver, a toxicity threshold expressed solely on a THg basis is insufficient for making confident toxicological assessments. The studies reviewed by Thompson (1996) and Shore et al. (2011) to estimate a Hg toxicity threshold based on THg in liver are all studies in which animals were exposed to relatively high doses of MeHg through their diets until they died or showed clinical signs of MeHg neurointoxication. Therefore, there is an implicit assumption—probably valid—that all or most of the Hg in the dosed animals' tissues was present as MeHg. But for liver tissue from free-living mammals or birds, many species of which demonstrate variable proportions of inorganic Hg and MeHg, this assumption is not valid. There are many examples of apparently healthy free-living mammals and birds with very high (> 100 µg g^{-1} ww) hepatic THg concentrations (Smith and Armstrong 1975, Norstrom et al. 1986, Thompson and Furness 1989,

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Dietz et al. 1990). As THg concentrations in liver increase in long-lived aquatic predatory wildlife, a progressively lower proportion is typically present as MeHg (Wagemann et al. 2000). Inorganic Hg resulting from demethylation in liver is often found in close association with selenium (Se), especially at higher Hg concentrations, and the Hg-Se complex is generally considered to be relatively non-toxic (e.g., Ikemoto et al. 2004). Without knowledge of the proportions of THg in the liver that are present as MeHg versus inorganic Hg, plus an estimate of corresponding Se concentrations, confident toxicological assessments cannot be made (see section 7.3 for additional discussion of Hg-Se interactions). Indeed, in their recent review, O'Hara et al. (2011) refrained from suggesting Hg toxicity thresholds for marine mammals, based largely on a recognition that much of the Hg in livers of these species may be inorganic Hg bound with Se, and on a lack of clear empirical evidence of toxicity in marine mammals with elevated liver Hg concentrations.

In the absence of information on the proportion of THg in liver that is present as MeHg, it is useful to have data on THg concentrations in certain other tissues, such as skeletal muscle. There is little or no evidence of demethylation in muscle tissue, thus THg and MeHg concentrations are approximately equal in this tissue. In some of the dosing studies examined by Thompson (1996) and Shore et al. (2011) to estimate a THg concentration in liver above which MeHg poisoning occurred in mammals, THg in both liver and muscle were reported. In these MeHg dosing studies, THg in both liver and muscle were highly elevated in animals suffering from MeHg poisoning, with muscle THg concentrations reaching about 1/3 to 1/2 of liver concentrations (e.g., Aulerich et al. 1974, Wobeser et al. 1976, O'Connor and Nielsen 1980). Thus, for a concentration of THg in liver of 25–30 µg g^{-1} ww (Shore et al. 2011) to be accepted as a valid threshold for Hg intoxication, THg in skeletal muscle tissue should be in the range of about 8–15 µg g^{-1} ww or greater. Conversely, if muscle THg concentrations are found to be low (≤ 1 µg g^{-1} ww) in animals with elevated liver THg, it is unlikely that such individuals will be suffering from Hg toxicity. An examination of available data on THg in liver and muscle of Canadian Arctic beluga and seal samples confirmed that, although some animals had THg concentrations in liver exceeding the putative 30 µg g^{-1} ww toxicity threshold, muscle THg concentrations were generally low which indicates that these species are unlikely to be experiencing significant MeHg toxicity (Fig. 7.8). Only harbour seals from western Hudson Bay had highly elevated mean liver THg along with muscle THg substantially higher than 1 µg g^{-1} ww. Therefore, further study on possible health effects from MeHg exposure on this subpopulation of seals is warranted.

7.2.3.2 Methylmercury toxicity in other organs

Although it is generally accepted that the central and peripheral nervous system is the primary target for MeHg toxicity in mammals, other tissues can also show varying degrees of cellular effects in response to Hg accumulation. In a review by Sonne (2010), a liver THg concentration of 11 µg g^{-1} ww was suggested as a threshold for hepatic effects in polar bears based on studies relating Hg concentrations with liver lesions in East Greenland polar bears. In the same review, a threshold THg concentration of 14 µg g^{-1} ww was suggested for kidney lesions in adult male East Greenland polar bears. It is not known how debilitating such relatively minor hepatic or renal lesions may be for individual bears. However, it should be noted that much higher liver THg concentrations have been observed in apparently healthy polar bears (e.g., Norstrom et al. 1986). Nevertheless, additional research is warranted to better characterize sublethal toxic effects of Hg accumulation in organs other than the brain in polar bears and other Arctic marine mammals.

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BOX 7.1 Integrating Observational Studies with Contaminant Monitoring

To achieve a better understanding of the effects of contaminants on animal function and health in the Arctic, there is an overall need to involve local hunters and knowledge-holders. Behavioural changes associated with Hg exposure are challenging to observe in free-living wildlife. However, hunters have the potential to obtain unique and valuable information about animal behaviour during their harvesting activities. When possible, future studies should be designed to involve hunters and local knowledge-holders to ensure that observations regarding changes or abnormalities in animal behaviour and health that may be related to Hg or other contaminant exposure are documented. Integrating observational studies with contaminant monitoring data has the potential to aid our understanding of sublethal effects of Hg on animal function in the Arctic.

7.2.3.3 Toxicogenomics and immunotoxicity in beluga

With a minimum estimate of 40,000 individuals (COSEWIC 2004), the Beaufort beluga whale population is one of Canada's largest, and there is no indication of a population decline. Mercury monitoring from 1981 to 2002 revealed high levels in Beaufort Sea beluga whales relative to other Canadian populations and an increasing temporal trend (Lockhart et al. 2005b). This led to several more recent studies aimed at identifying the main sources of Hg to the region. As part of the beluga sampling program at Hendrickson Island, a health assessment of contaminant effects on known toxicological endpoints was conducted.

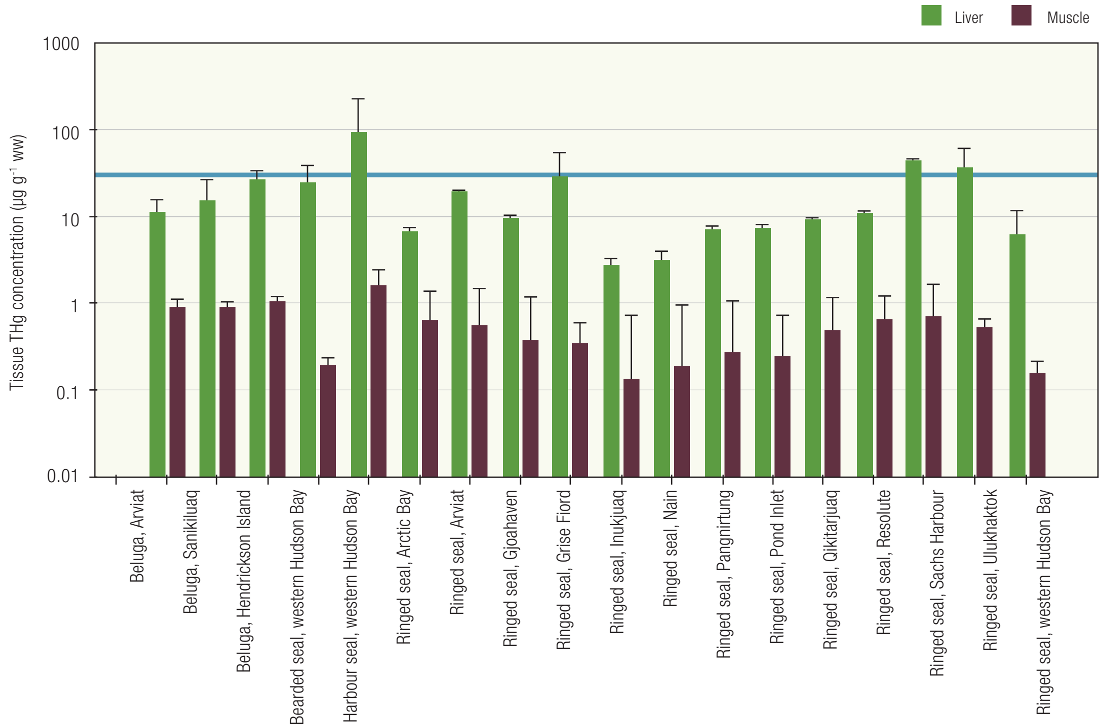

FIGURE 7.8

Concentrations of THg in liver and muscle (on a logarithmic scale, mean ± standard deviation) of Arctic beluga and seal compared to a toxic threshold of 30 µg g^{-1} ww (blue line) suggested for terrestrial mammals (Thompson 1996). Beluga data are from Gaden and Stern (2010) while seal data are from Muir (2010) and Young et al. (2010). Comparisons between toxicity thresholds and liver THg concentrations should be interpreted with caution because a large portion of elevated liver Hg in these marine mammals is likely in a less toxic inorganic form—as corroborated by relatively low (≤ 1 µg g^{-1}) muscle THg concentrations in most samples.

The impacts of Hg on gene expression in beluga are being investigated (Ross et al. 2010). The long-term goal of this research is to develop a highly sensitive technique that will provide a useful early warning indicator of the effects of Hg and other contaminant exposure on the health of the western Arctic beluga population. Metallothioneins (MTs) are low molecular weight sulfhydryl-rich proteins that are able to bind to group II metals, especially the essential trace metals copper and zinc, and the toxic non-essential metals cadmium and inorganic Hg. It is generally accepted that MT synthesis and binding of toxic metals provide protection against the cytotoxic action of these metals. In beluga liver, no correlation was found between MT1 gene expression and THg levels (Fig. 7.9), which suggests that MTs may not be the primary means of Hg detoxification in liver. This result is consistent with previous work showing that only 5% of Hg is bound to MTs in liver (Wagemann and Muir 1984). In skin, a positive relationship between MT1 expression and THg levels was observed ($r^2 = 0.51$; $p = 0.017$), which suggests that MTs might play a significant role in the binding of inorganic Hg in the skin of beluga. However, Hg in the skin of cetaceans and pinnipeds is predominately in the form of MeHg (90%) rather than inorganic Hg (Wagemann et al. 1998, Dehn et al. 2006).

To investigate the effects of inorganic Hg and MeHg on the immune system of beluga whales, laboratory experiments were conducted on peripheral blood collected from four captive beluga held at the Vancouver Aquarium (Frouin et al. 2012). Lymphocyte suspensions were exposed *in vitro* for 66 hours to 0.1–10 µM of Hg chloride ($HgCl_2$) and 0.033–10 µM of MeHgCl. Relationships between Hg concentration and the proliferation of concanavalin A (Con-A)-stimulated lymphocytes

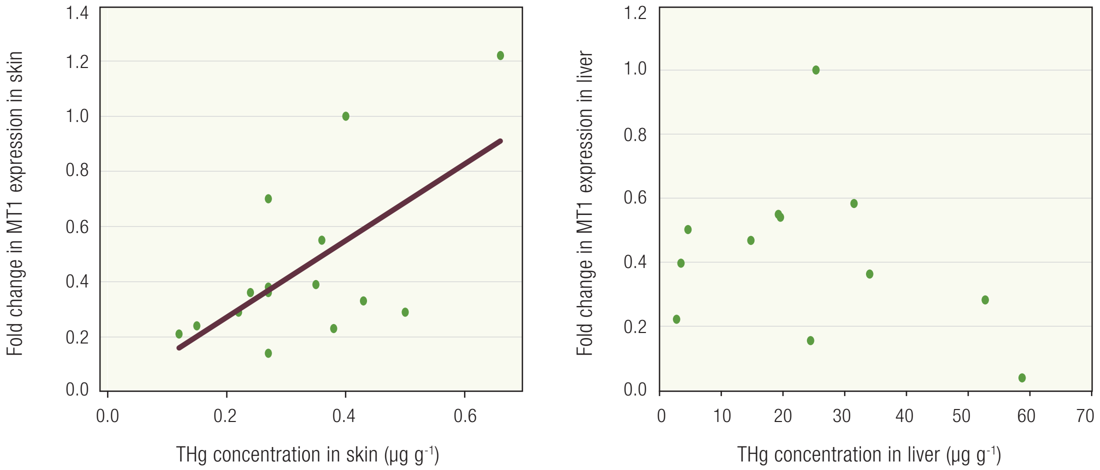

FIGURE 7.9

Relationships between THg concentrations and metallothionein (MT1) gene expression in skin ($r^2 = 0.51$, $p = 0.017$) and liver (not significant) of male beluga whales sampled in 2008 (Ross et al. 2010). The MT1 expression was first normalized to the expression of the gene encoding the ribosomal protein L8 and further normalized to the individual showing average contaminant concentrations.

were then evaluated. Cell viability was also measured. A significant reduction in T-lymphocyte proliferation was observed at ≥ 1 µM $HgCl_2$ and ≥ 0.33 µM MeHgCl (Fig. 7.10). Cell viability decreased only at the highest concentrations of $HgCl_2$ (10 µM), and at ≥ 1 µM of MeHgCl. Calculated doses for 50% inhibition of the proliferation response (ID_{50}) indicate that MeHgCl (ID_{50} = 0.24 µM) suppressed proliferation of beluga lymphocytes at concentrations ten times lower than $HgCl_2$ (ID_{50} = 2.62 µM). These results agreed with previous observations that MeHg is a more potent suppressor of splenocyte proliferation than inorganic Hg (De Guise et al. 1996), and they suggest that Hg, particularly MeHg, may be toxic to beluga whale immune cells at the range of concentrations (1–100 µg g^{-1}) observed in the liver of some free-ranging populations of Arctic beluga whales (Lockhart et al. 2005b). However, *in vivo*, most MeHg in blood is bound to proteins such as haemoglobin within red blood cells and may not be as available to interact with lymphocytes compared to *in vitro* studies using isolated lymphocytes. Additional research to investigate the possible *in vivo* immunotoxic effects of MeHg exposure in Arctic marine mammals is warranted.

7.3 Mercury-selenium interactions

In some tissues of fish-eating and other predatory aquatic mammals and birds, MeHg from the diet is demethylated and the resulting inorganic Hg combines with Se. This Hg-Se sequestration occurs especially in liver and to some extent in kidney and brain. At higher concentrations of liver Hg in such animals, it is common to find an increasingly large proportion of THg present as inorganic Hg associated with Se, and less THg present as MeHg (e.g., Scheuhammer et al. 1998, Henny et al. 2002). A liver MeHg concentration of about 8–10 µg g^{-1} dw has been suggested as a threshold above which demethylation is activated in a number of wildlife species (Dietz et al. 1990, Scheuhammer et al. 2008, Eagles-Smith et al. 2009). At high THg concentrations (> 50 µg g^{-1} dw), the molar ratio of Hg:Se in the liver is often approximately 1:1 (Koeman et al. 1975, Dietz et al. 2000). Demethylation of MeHg and subsequent sequestration of inorganic Hg with Se has frequently been suggested as a probable detoxification mechanism for animals exposed to relatively high levels of dietary MeHg (Bjorkman et al. 1995, Palmisano et al. 1995, Caurant et al. 1996). Ralston et al. (2008) reported that the molar ratio of Hg:Se was critical to the expression of MeHg toxicity. This suggests that as long as molar excesses of Se over Hg are maintained, cells may be largely protected from the toxicity of Hg by the presence of Se.

Mercury has an even greater binding affinity for Se than it does for sulphur (Sugiura et al. 1978). Therefore, tissue accumulation of Hg can affect the amount of Se that is biologically available for maintaining adequate seleno-enzyme levels essential for protecting the brain and other tissues from oxidative stress (Ralston et al.

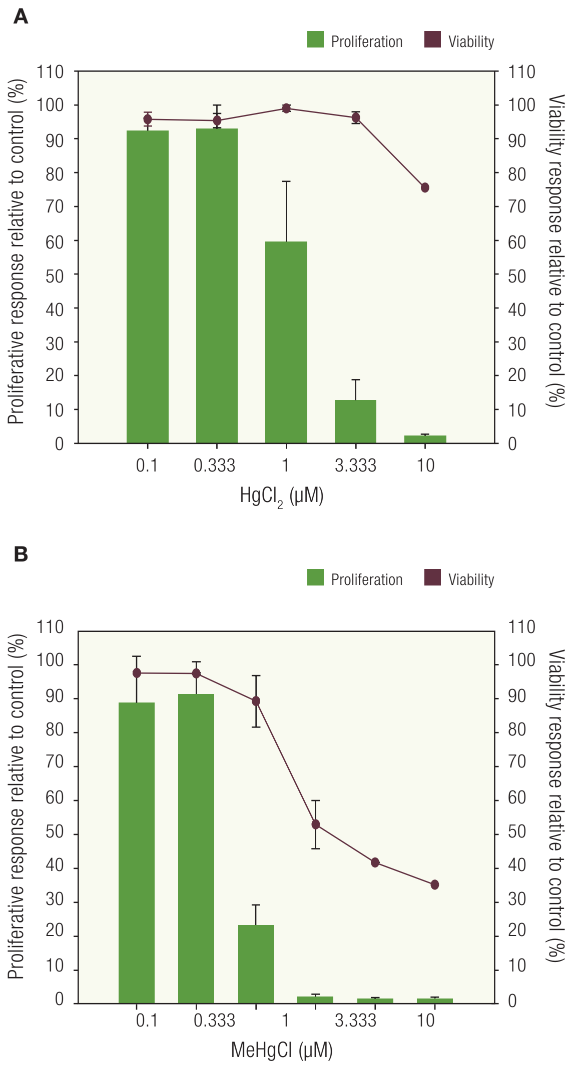

FIGURE 7.10

The proliferation of T lymphocytes (bars) and their viability (lines) following different exposures to HgCl$_2$ (A) or MeHgCl (B). Results are expressed as a percentage (%) of the control response (unexposed cells) ($n = 4$ per dose group; mean ± standard error) (source: Frouin et al. 2012).

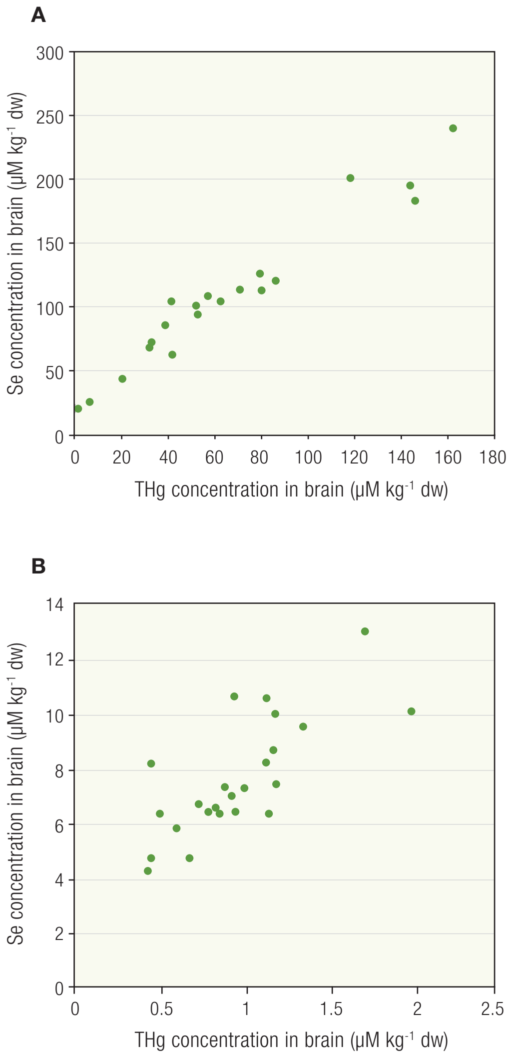

FIGURE 7.11

Relationship between molar concentrations of Se and THg (µM kg^{-1} dw) in the cerebellum of beluga whales (A) harvested in the western Canadian Arctic ($n = 20$) and polar bears (B) harvested in the eastern Canadian Arctic ($n = 24$) (Krey and Chan 2010, Ostertag and Chan 2010).

2008). Certain seleno-enzymes (e.g., thioredoxin reductase) are highly sensitive to inhibition by low nanomolar concentrations of Hg and may be primary targets of MeHg toxicity at the molecular level (Carvalho et al. 2008). From a toxicological perspective, demethylation of MeHg in liver and the interaction between Hg and Se have important implications. Chief among these is a realization that confident toxicological assessments cannot be made based solely on THg concentrations commonly reported in tissues such as liver or kidney. Rather than estimating LOAEL or similar threshold toxicity values based solely on THg concentrations in brain, liver, or other tissues that are prone to exhibit variable proportions of MeHg and inorganic Hg, it is preferable that concentrations of THg and MeHg as well as Hg:Se molar ratios be considered together to more confidently assess whether tissue Hg concentrations are sufficiently high to impair health and reproduction.

The relationship between Hg and Se co-accumulation has recently been investigated in the cerebellum of Arctic beluga whales and polar bears (Krey and Chan 2010, Ostertag and Chan 2010). Concentrations of Se and THg were positively correlated for both beluga whales ($r = 0.97$, $p < 0.01$) and polar bears ($r = 0.75$, $p < 0.01$) (Fig. 7.11).

The molar ratio of Hg:Se was higher in beluga whale cerebellum than in polar bear cerebellum ($p < 0.01$) (Fig. 7.12) (Krey and Chan 2010, Ostertag and Chan 2010). This finding may be of concern, given that beluga have much greater brain Hg concentrations than polar bears. However, a molar excess of Se over Hg was nevertheless observed in both polar bear and beluga brains, which indicates that beluga may, in general, be protected from MeHg toxicity in spite of relatively high brain Hg concentrations. Because of the protective effect of Se on Hg toxicity, a high tissue Hg concentration accompanied by a high (> 1) Hg:Se molar ratio is of greater concern than a high tissue Hg concentration accompanied by a low (< 1) Hg:Se molar ratio. However, more research is necessary to better understand the role of Se in protecting the brains of Arctic predatory mammals and birds from Hg toxicity. Further discussion of Hg-Se interactions in marine mammals can be found in a recent review by O'Hara et al. (2011).

Data from the 1980s indicated that THg concentrations in livers of Canadian polar bears were highest in western Arctic areas bordering the Beaufort Sea (mean approximately 100–200 µg g^{-1} dw). However, the Hg:Se molar ratio for these animals was approximately 1:1 indicating

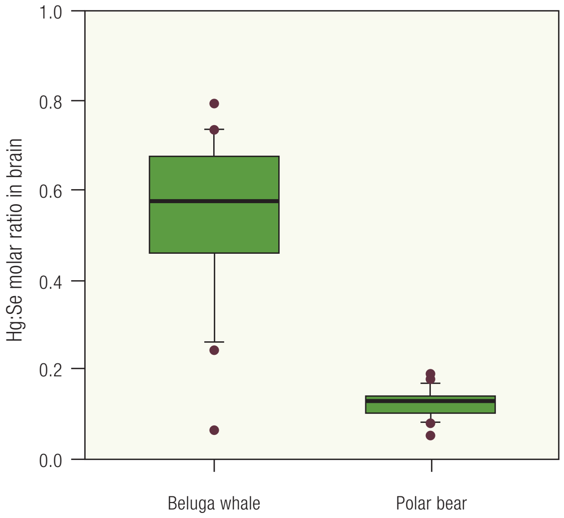

FIGURE 7.12

Molar ratio of Hg:Se in beluga whale (*n* = 21) and polar bear (*n* = 24) cerebellum (Krey and Chan 2010, Ostertag and Chan 2010). The boxplots identify the 10th and 90th percentiles (error bars), the 25th and 75th percentiles (box) and the median concentration (middle line).

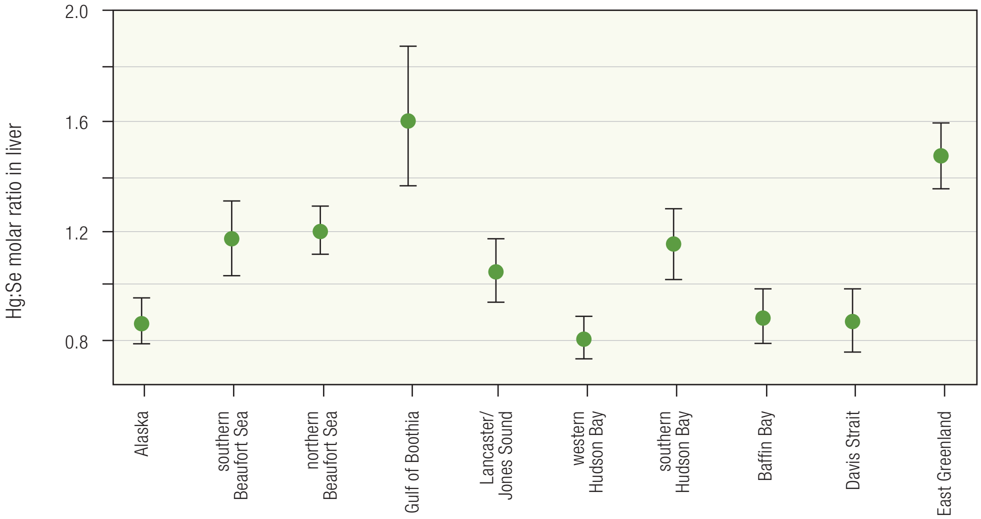

FIGURE 7.13

Molar ratio of Hg:Se concentrations in polar bear livers collected from Alaska, Canada, and Greenland in 2005 to 2008. Reprinted with permission from Routti et al. (2011). © The Royal Society of Chemistry.

sufficient Se to sequester all Hg (Braune et al. 1991). More recently, Routti et al. (2011) reported that THg concentrations were higher in the livers of polar bears sampled from 2006 to 2008 from the Beaufort Sea compared to animals sampled in the 1980s. However, Se concentrations increased concurrently with Hg and resulted in stable Hg:Se molar ratios over time for this population. Geographic, and perhaps temporal, differences in THg concentrations in polar bears are largely explained by differences in trophic position (section 6.12.3). The highest mean molar ratio of Hg:Se (1.6) in polar bear liver—indicating a molar excess of Hg over Se—was observed in bears from the Gulf of Boothia compared with several other northern Canadian locations sampled from 2006 to 2008 (Fig. 7.13; Routti et al. 2011). Total Hg concentrations in Gulf of Boothia bears, although lower than in Beaufort Sea bears, were not trivial (> 20 µg g^{-1}), thus, there is currently a basis for concern that some bears from Gulf of Boothia may be at risk for Hg-mediated toxicity. In addition, assuming concentrations of THg in liver are high (> 80 µg g^{-1} dw), polar bears from other locations where the Hg:Se molar ratio exceeds unity should also be further assessed for possible Hg toxicity.

7.4 Summary

Although recent advances have been made through investigations of toxicological effects on certain seabirds and marine mammals in the Canadian Arctic, there is insufficient information to confidently determine whether Hg exposure is currently impacting the health of Arctic fish and wildlife. Studies on MeHg accumulation and toxicity in temperate species, including information gleaned from controlled feeding studies, offer additional useful information that can be applied, with caution, to an assessment of Hg concentrations in tissues of Arctic species. Nevertheless, there is an explicit need for Hg effects information for Arctic species. Therefore, bioeffects studies should comprise a major focus of future Hg research in the Canadian Arctic.

An assessment of potential toxicological effects on Arctic fish from freshwater and marine environments was limited to comparisons with a muscle Hg threshold derived from laboratory and field studies of non-Arctic species. Freshwater fish in northern Canada mostly had lower THg concentrations than a threshold range of 0.5–1.0 µg g^{-1} ww, above which adverse biological effects are more likely to occur. However, a few lakes in the Northwest Territories and Nunavut contained fish species that exceeded this threshold. Marine fish species generally had substantially lower THg concentrations than freshwater fish. One exception was a long-lived predatory species, the Greenland shark, which had THg levels that exceeded the lowest observable adverse effects threshold. Future study of toxicological effects is warranted on those Arctic fish with elevated Hg levels.

Seabirds were investigated for overt toxicological effects and more subtle biochemical changes resulting from Hg exposure. A laboratory egg injection study indicated that embryos of thick-billed murres appeared to be slightly more sensitive to MeHg than those of Arctic terns and had a higher embryo mortality at a lower MeHg dose. However, both species had LC_{50} concentrations that indicated a medium sensitivity to MeHg relative to a variety of non-Arctic birds. Neurochemical receptors were measured in the brains of thick-billed murre and Arctic tern embryos from the egg injection study, and receptor levels were not related to MeHg exposure. The effect of Hg on cellular oxidative stress was investigated in livers of nestling glaucous gulls from two Canadian Arctic sites. Although significant correlations were found between concentrations of THg and oxidized and reduced glutathione (a biochemical that regulates oxidative stress), overall evidence for an effect was weak, leading to the conclusion that glaucous gull nestlings at the two colonies were exposed to lower levels of oxidative stress than birds in more highly contaminated environments. In addition, the potential for toxicological effects in marine birds from the Canadian Arctic was assessed by comparing THg concentrations in eggs and livers with threshold levels derived from published field and laboratory studies. Individuals from several species including ivory gulls, black guillemots, glaucous gulls, Arctic terns, and northern fulmars had egg or liver Hg concentrations that exceeded the threshold for possible reproductive effects. Additional research is needed to determine if, and to what extent, Hg exposure, combined with other stressors such as climate change, are affecting Arctic marine bird populations.

The potential for toxicological effects in beluga and seals from the Canadian Arctic was assessed by comparing THg concentrations in liver and muscle with threshold levels derived from published field and laboratory studies. While liver in marine mammals can have variable proportions of inorganic Hg and MeHg, muscle primarily contains MeHg. Some beluga and seals had THg concentrations in liver exceeding the putative 30 µg g^{-1} ww toxicity threshold. However, muscle THg concentrations were generally low which indicates that these species are unlikely to be experiencing significant MeHg toxicity. Only harbour seals from western Hudson Bay had highly elevated mean liver THg along with comparatively high muscle THg. Therefore, further study on possible health effects from MeHg exposure on this subpopulation of seals is warranted.

Recent and ongoing investigations have examined neurotoxic and immunotoxic effects of Hg on polar bears and beluga whales. In polar bears from the Canadian Arctic, no significant association was found between THg concentrations and the activity of a neurochemical enzyme (MAO), the density of a neurotransmitter receptor (mACh), nor Se:Hg molar ratios in cerebellum. Concentrations of MeHg in polar bear cerebellum were lower than levels observed to be toxic in laboratory feeding trials of non-Arctic mammals. Concentrations in beluga whale cerebellum were sufficiently high to potentially cause significant neurochemical changes, but probably not high enough to cause overt MeHg neurotoxicity. Preliminary research on the sub-lethal effects of Hg on beluga whales suggests a link between THg concentration and MT gene expression in their skin tissue. In addition, laboratory experiments were conducted on peripheral blood collected from four captive beluga held at the Vancouver Aquarium. Lymphocyte suspensions exposed to inorganic Hg and MeHg showed suppressed growth and reduced cell viability at environmentally relevant concentrations. Additional research is warranted to investigate possible neurotoxic and immunotoxic effects of MeHg exposure in Arctic marine mammals.

In some tissues of predatory mammals and birds, MeHg from the diet is demethylated and sequestered as a Hg-Se complex—most likely as a detoxification mechanism against exposure to relatively high levels of MeHg. At higher concentrations of liver THg in those animals, it is common to find an increasingly large proportion of THg present as inorganic Hg associated with Se, and less THg present as MeHg. The relationship between Hg and Se co-accumulation was investigated in the cerebellum of beluga whales and polar bears from the Canadian Arctic. Concentrations of Se and THg were positively correlated for beluga whales and polar bears, with the molar ratio of Hg:Se higher in beluga. Despite the higher ratio, there was still a molar excess of Se over Hg in beluga brains, and beluga may, in general, be protected from MeHg toxicity in spite of relatively high brain Hg concentrations. Ratios of Hg:Se were examined in livers of polar bears from across the Canadian Arctic and, unrelated to Hg concentration, were found to vary considerably among regions. Liver Hg:Se ratios were especially high in some polar bears from the Gulf of Boothia, indicating a deficit of Se relative to Hg. More research is needed to better understand the role of Se in protecting Arctic predatory mammals and birds against the toxicological effects of Hg.

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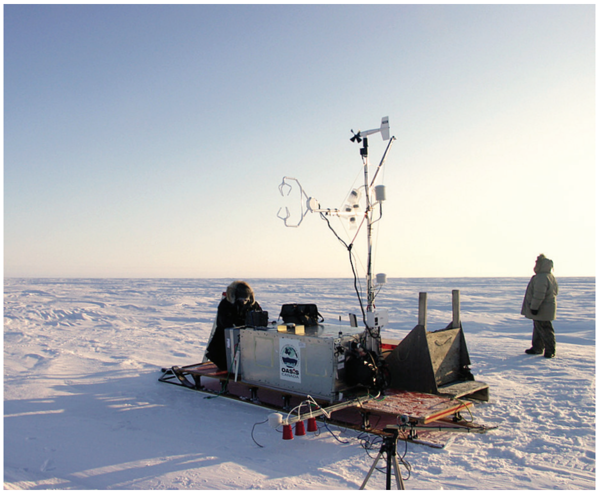
Chapter 8

Conclusions and Recommendations

Authors: John Chételat, Birgit Braune

8.1 Introduction

Important scientific advances have been made in identifying the environmental fate of Hg and the processes that control its movement in the Canadian Arctic. The *CACAR III: Mercury in Canada's North* provides much new information on the sources and long-range transport of Hg to the Arctic, its cycling within marine, freshwater, and terrestrial environments, and its bioaccumulation in, and effects on, the biota that live there. While these accomplishments are significant, the complex nature of the Hg cycle continues to provide challenges in characterizing and quantifying the relationships of Hg sources and transport processes with Hg levels in biota and biological effects of Hg exposure. These gaps in our knowledge of how global anthropogenic emissions of Hg are delivered to, and accumulate in, Arctic environments have implications for policy development related to the risk management of Hg. Of particular concern are large uncertainties in our understanding of the processes that are contributing to increasing Hg concentrations in some Arctic fish and wildlife. Specific recommendations are provided for future research and monitoring of the environmental impacts of anthropogenic Hg emissions, climate change, and the effectiveness of mitigation strategies for Hg in the Canadian Arctic.



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8.2 Main findings, knowledge gaps and recommendations

8.2.1 Atmospheric Hg

Since the last *CACAR*, research and monitoring of atmospheric Hg has improved our understanding of this key transport pathway to and in the Canadian Arctic. The chemical processes of atmospheric mercury depletion events (AMDEs) have been better characterized. More field measurements on atmospheric Hg speciation are now available, and changes in speciation that occur during AMDEs have been more fully documented. Automated measurements of air Hg concentrations over the last decade have provided new insights into temporal trends at different time scales. The geographic coverage of air measurements has been expanded, and data are now available for several new sites in the Arctic. In addition, models of atmospheric processes associated with the long-range atmospheric transport of Hg from various source regions around the world and Hg deposition in the Arctic have been refined.

Key findings:

- The speciation of atmospheric Hg changes during springtime AMDEs in the Arctic where the oxidation of gaseous elemental Hg is accompanied by large increases in reactive gaseous Hg and particulate Hg concentrations.
- The timing of AMDEs at the High Arctic monitoring station at Alert has changed over the last two decades, with the month of maximum AMDE activity shifting from May to April. The reason for the shift in the timing of depletion events is not yet understood, but may be related to changes in air temperature.
- Air Hg concentrations have declined from 2000 to 2009 at Alert in the High Arctic and Kuujjuarapik in the sub-Arctic. The rate of decline at Kuujjuarapik was comparable to non-Arctic monitoring sites at lower latitudes while a slower rate of decline was observed at Alert.

- Model simulations of air Hg concentrations and deposition for the Canadian High Arctic suggest that atmospheric Hg trends during the last two decades were related to meteorology and global changes in anthropogenic Hg emissions.
- Model simulations suggest that long-range atmospheric transport from Asia likely contributes the most Hg (from anthropogenic, natural, and re-emission sources) to the High Arctic and western Arctic, followed by contributions from North America, Russia, and Europe.
- For the Canadian Arctic, if anthropogenic Hg emissions were to increase along a *status quo* scenario, model simulations project an average increase in atmospheric Hg deposition of about 5% between 2005 and 2020. Maximum declines expected under the implementation of best control technologies are projected at nearly 20% by 2020.

Knowledge gaps and recommendations:

- Considerable uncertainty remains in the quantification of atmospheric Hg contributions to Arctic ecosystems. Direct measurements of wet and dry deposition in the Canadian Arctic are extremely limited due, in part, to technical challenges. Therefore, more research using a variety of methodological approaches is essential to resolve this knowledge gap, especially since any changes in emission controls would most immediately affect the atmospheric Hg flux.
- Better characterization of geographic variation in atmospheric Hg is warranted, particularly over the Arctic Ocean, to determine the influence of unique marine conditions on deposition, including climate-driven changes in ice cover. Continued monitoring is recommended for temporal trends in air Hg concentrations and speciation.
- Laboratory and field research should focus on further characterizing the mechanisms of atmospheric Hg deposition. More information is needed on oxidized Hg species in air as well as chemical reaction rates and products in gaseous and aqueous phases in the Arctic environment. Rate constants in air and Hg reduction chemistry in snow are particularly important for atmospheric modelling.
- Atmospheric models of long-range transport and temporal trends of Hg in Arctic air depend on the reliability of data for anthropogenic emissions and the speciation of Hg emissions. Further improvements of global Hg emission inventories would help to refine model estimates.

8.2.2 Biogeochemical cycling of Hg

Some of the most significant advances since the last *CACAR* have been made in the area of Hg cycling in the Arctic environment. Large amounts of new data are now available for water, snow, ice, and sediment, which provide more information on the concentrations, fluxes, and transformations of Hg in these abiotic matrices. This information was virtually absent for the Arctic a decade ago. Investigations of Hg biogeochemistry have focussed on a range of environments including tundra, lakes, rivers, glaciers, and the Arctic Ocean.

Key findings:

- Recent studies on snow Hg concentrations indicate that much of the Hg deposited during AMDEs is quickly reduced and emitted to the atmosphere. The level of evasion is dependant on proximity to the ocean because halogen compounds from the marine environment promote the stabilization and accumulation of Hg in snowpack. Areal loads of Hg in snow were estimated for Hudson Bay and the Arctic Archipelago, which suggested that snowmelt probably constitutes a small flux of Hg to Arctic marine waters during spring.
- The atmospheric flux of Hg to glacier snow and ice increased from north to south in the eastern Arctic, likely due to latitudinal change in wet deposition and/or particulate scavenging. Mercury accumulated in glacier snow primarily during the autumn when AMDEs do not occur. Contributions to Hg accumulation from wet deposition are probably more important at lower Arctic latitudes.
- A peak atmospheric flux of Hg was observed between the mid-1990s and 2005 in two glacier archives from the High Arctic. This temporal trend was not found in direct monitoring of air Hg concentrations at nearby Alert and indicates discrepancies between temporal trends measured in air and estimates of recent net deposition from environmental archives.
- Mercury fluxes to lake sediments have increased several-fold since the onset of the Industrial Era at a large number of sub-Arctic and Arctic sites. While all evidence points to increasing Hg fluxes in the lakes, it is unclear to what extent Hg from anthropogenic sources is being deposited in greater amounts or whether other factors, such as higher algal productivity or increased sedimentation, are attenuating Hg inputs from the atmosphere and lake catchment.

- In the High Arctic, snowmelt was found to be an important source of Hg to lake waters. Higher levels of water MeHg were found in some fresh waters, such as shallow ponds, where conditions are probably more suitable for *in situ* methylation.
- The first measurements of methylation rates in Arctic seawater indicate that the water column is an important site for MeHg formation from inorganic Hg. Production of dimethylmercury (Me_2Hg) also occurs in the water column, though generally at a slower rate. The breakdown of Me_2Hg in the water column or in the air after evasion may be a source of MeHg available for biological uptake. These findings challenge the paradigm that only anaerobic conditions support significant MeHg production.
- Arctic marine waters were found to be a substantial source of gaseous Hg to the atmosphere during the ice-free season.
- A mass balance model for the Arctic Ocean estimated that net atmospheric deposition was the largest Hg source, followed by coastal erosion and oceanic transport. In contrast, mass balance modelling identified rivers as the largest single Hg source in Hudson Bay followed in importance by Hg fluxes from the atmosphere and ocean inflow.

Knowledge gaps and recommendations:

- Little information currently exists on Hg methylation in marine, freshwater, and terrestrial environments in the Canadian Arctic. It is also unclear what processes lead to the occurrence of MeHg in the cryosphere. More fundamental research is recommended on the mechanistic aspects of Hg methylation and degradation to better understand the processes regulating MeHg entry into Arctic food webs.
- Oxidation and reduction cycles of inorganic Hg remain poorly characterized in Arctic environments. Further research is recommended to develop a quantitative and mechanistic understanding of abiotic and microbially driven Hg fluxes between air and various surfaces (soil, water, snow, and vegetation). Biogeochemical models of air-surface Hg exchanges are important for the modelling of atmospheric Hg inputs and long-term trends on Hg pollution in the Arctic.
- The bioavailability of Hg, as divalent inorganic Hg and as MeHg, is poorly understood in the Arctic environment, and research is needed to determine how much of the atmospherically-deposited Hg is readily available for microbial processing and uptake in food webs.

Rodd Laing

- Arctic soils, especially, have received little attention with respect to Hg levels and cycling. Further investigation is recommended to determine the extent of soil Hg processing and its role in local and regional Hg cycles, particularly since climate change may potentially alter the fluxes of Hg from thawing soils to the atmosphere and aquatic ecosystems.
- Uncertainties remain in the use of environmental archives as chronologies of atmospheric Hg deposition in the Arctic. Further research is recommended to explain increasing Hg fluxes to Arctic lake sediments, specifically the roles of anthropogenic Hg inputs and other processes in the lakes and their catchments.

8.2.3 Monitoring of Arctic biota

An important advance during Phase III of the NCP was the establishment of more frequent monitoring for key Arctic biota. In addition, supporting biological measurements such as stable isotopes and age are now routinely taken during monitoring to aid with trend interpretation. Since the last *CACAR*, a number of food web studies have been conducted in freshwater, marine, and terrestrial environments across the Canadian Arctic. These detailed investigations have provided a better understanding of the roles of Hg uptake, trophic transfer, and diet choice for Hg bioaccumulation in Arctic food webs.

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Key findings:

- The Hg content of teeth from ringed seals and beluga in the Canadian Arctic were several-fold higher in samples collected in the early to mid-20th Century compared with teeth from the 19th Century and pre-industrial period. These findings indicate a long-term effect of global anthropogenic Hg emissions on Hg bioaccumulation in the Arctic.
- A power analysis of temporal trend datasets for Arctic char and seabirds indicated that more frequent monitoring, initiated during Phase III of the NCP, provided important increases in the ability to detect changes in biotic Hg levels.
- Over the last two to four decades, Hg concentrations have increased in marine and freshwater animals, specifically in several species of Arctic-breeding seabirds, some polar bear subpopulations, and some freshwater fish from the Mackenzie River Basin. Other monitored populations, including caribou, beluga, ringed seal, and freshwater fish from the Yukon and Nunavut showed no change or a slight decline. The different Hg trends reported for Arctic biota indicate that the drivers of temporal change may be regional or habitat-specific.
- In general, terrestrial animals in the Canadian Arctic had low levels of Hg, with caribou having the highest concentrations among the species studied. The importance of lichen in the diet of caribou, which contains more Hg than other Arctic plant types, may explain their higher Hg burdens.
- No geographic patterns or regional hotspots were evident for Hg in common freshwater fish species that were sampled across the Canadian Arctic between 2002 and 2009. Average Hg concentrations in fish muscle did not exceed the Health Canada consumption guideline for commercial sale of fish (0.5 $\mu g\ g^{-1}$ wet weight) at the majority of sites surveyed.
- Food web processes as well as Hg supply from the catchment both affect the availability of Hg for uptake by Arctic freshwater fish. Preliminary work in the emerging field of Hg stable isotopes shows a future potential for differentiating sources of Hg to Arctic food webs.

- An extensive survey of sea-run Arctic char across the Canadian Arctic showed consistently very low concentrations of Hg. Lake-dwelling char are distinctly smaller than sea-run char and have substantially higher Hg concentrations that are, in some cases, due to slower growth rates and poorer body condition.
- Habitat use and food web structure factored into Hg levels in Beaufort Sea beluga, which varied within the population according to size and sex segregation.
- Geographic variation in Hg concentrations was observed for marine biota. Compared to other regions of the Canadian Arctic, the Beaufort Sea remains an area of higher Hg contamination for beluga, ringed seals, and polar bears. Elevated concentrations in polar bears from the Beaufort Sea relative to Hudson Bay were related to their higher trophic level and higher water concentrations of MeHg available for entry into the food web. Latitudinal trends were also found for ringed seals and seabirds, with greater concentrations in the High Arctic than at sub-Arctic sites.

Knowledge gaps and recommendations:

- Continued annual monitoring of Hg levels is recommended for key Arctic species to detect temporal trends. To determine the influence of food web and trophic level differences, ancillary measurements such as stable isotopes, age, sex, and size must continue to be included to facilitate the interpretation of trends.
- While strong evidence exists for increasing Hg bioaccumulation in some monitored fish and wildlife, the driver(s) of these temporal trends remain unclear. Further research is required to identify the underlying processes leading to changes over time in biotic Hg concentrations, particularly at sites where increases have been observed. Multiple factors may be implicated and should be assessed, including the roles of Hg delivery (e.g., atmospheric deposition, Hg methylation), shifts in food web structure, and climate change.
- Geographic patterns of Hg bioaccumulation in marine biota deserve further investigation because current observations suggest that regional factors may affect the vulnerability of biota to Hg exposure within the Arctic.
- Few recent data exist for lake trout or lake-dwelling Arctic char within their distribution ranges in the Kivalliq region (south-central Nunavut) and Nunavik. Given the recent increases observed in fish Hg concentrations in the Northwest Territories, renewed investigation of fish Hg bioaccumulation in the central and eastern Arctic is warranted.
- Little information exists on Hg levels in wholly marine fish species that may be important sources of dietary Hg for large marine mammals and seabirds. The factors that determine geographic variation and effects of habitat (e.g., nearshore versus offshore feeding) on Hg bioaccumulation in marine fish remain unresolved and warrant future investigation to better understand Hg transfer to top predator animals in the marine environment.

8.2.4 Climate change

Widespread evidence indicates that the Arctic environment is undergoing profound change and at a rapid rate. New research since the last *CACAR* indicates that this environmental change is altering the cycling and bioaccumulation of Hg through effects on Hg fluxes and food web structure. However, large uncertainties remain in this emerging and complex field.

Key findings:

- In the Mackenzie River basin, slumping of permafrost soils has increased the transport of inorganic material to lakes, but preliminary findings indicate that these large inputs have not increased Hg concentrations in sediment.
- Recent increases in algal primary production and catchment inputs may be enhancing Hg fluxes to Arctic lakes.
- Climate warming may be affecting the local biogeochemical processing of Hg and its resulting mobilization may have increased exposure to fish in the Mackenzie River, where a nearly two-fold increase in Hg concentration has occurred in burbot muscle since 1985.
- Climate change appears to be affecting the structure of food webs in certain areas, which is resulting in dietary shifts and changes in Hg bioaccumulation in some marine biota such as thick-billed murres and ringed seals.

Knowledge gaps and recommendations:

- A myriad of complex environmental changes are occurring in the Arctic and detailed, process-focussed investigations are needed to more precisely identify how these changes will alter the fate of Hg. Many aspects of the Hg cycle may be impacted including how Hg is delivered to Arctic ecosystems, its biogeochemical transformations, and its trophic transfer in food webs.

8.2.5 Biological effects

Modest progress has been made since the last *CACAR* in identifying the effects of MeHg exposure specifically for Arctic wildlife. These investigations have focussed on adverse reproductive effects in seabirds, neurotoxicity in marine mammals, immunotoxicity in beluga, and the role of selenium (Se) in Hg detoxification.

Key findings:

- An assessment of potential toxicological effects was conducted for Arctic fish from freshwater and marine environments, seabirds, and marine mammals by comparing tissue Hg concentrations with Hg toxicity thresholds derived from laboratory and field studies for non-Arctic species. In general, Arctic biota had Hg concentrations that were below threshold levels of potential concern, although exceedances were observed for some populations of freshwater fish, Greenland shark, and seabirds.
- A laboratory study, involving the injection of environmentally relevant levels of MeHg into Arctic seabird eggs, identified differences in sensitivity of the developing embryos to MeHg between thick-billed murres and Arctic terns. Neurochemical receptor levels in the brains of thick-billed murre and Arctic tern embryos from the egg injection study were not related to the level of MeHg exposure.

Ed Struzik

- Glaucous gulls from two colonies in the Canadian Arctic showed lower levels of cellular oxidative stress than would be expected for birds in more highly contaminated environments at lower latitudes.
- Selenium is known to play an important role in sequestering and detoxifying Hg. The amount of Se present in Arctic polar bears and beluga whales relative to Hg was variable, which may suggest differences in protection from MeHg toxicity.
- The activity of two neurochemical receptors was not related to cerebellum Hg concentrations in polar bears from the Canadian Arctic. In beluga whale cerebellum, Hg concentrations were sufficiently high to potentially cause significant neurochemical changes, but probably not high enough to cause overt MeHg neurotoxicity. The formation of relatively inert Hg–Se compounds in beluga brains may reduce their risk of Hg toxicity.
- Preliminary research on sub-lethal effects of Hg on beluga whales suggests a link between Hg concentration and gene expression in their skin tissue. Environmentally relevant concentrations of Hg were also found to suppress growth and reduce cell viability of lymphocytes obtained from peripheral blood of beluga.

Knowledge gaps and recommendations:

- There is currently insufficient information to adequately assess the effects of Hg exposure specifically for Arctic wildlife species, and further research should determine if and to what extent Hg exposure, combined with other stressors such as climate change, is affecting populations of Arctic fish, seabirds, and marine mammals that have elevated Hg levels.
- Additional research is recommended on possible neurotoxic and immunotoxic effects of MeHg exposure in Arctic marine mammals.
- The role of Se in protecting Arctic predatory mammals and birds against toxicological effects of Hg warrants further study.

8.2 Future directions

Over the last two decades, the NCP has played a pivotal role in supporting research and monitoring of Hg in the Canadian Arctic environment. Phases I and II of the NCP provided important new knowledge on the Arctic Hg cycle, which, in turn, resulted in the development and refinement of program activities related to this contaminant. Recent scientific advances during Phase III of the NCP, which are documented in this assessment, confirm the importance of continuing Hg research and monitoring in core areas of the program and highlight the need for increased efforts to address unresolved knowledge gaps that are central to the issue of Hg pollution in the Arctic. Key recommendations of the *CACAR III: Mercury in Canada's North* are as follows:

- Continue research and monitoring of atmospheric Hg, with an enhanced focus on deposition measurements to facilitate quantification of atmospheric contributions of Hg to Arctic ecosystems.
- Continue temporal trend monitoring of Hg in Arctic biota, and identify the processes that are changing Hg concentrations in some species.
- Further characterize the key processes acting on Hg after atmospheric deposition and their effects on the fate of Hg in the Arctic environment.
- Better characterize the processes that link climate change with Hg transport, cycling, and bioaccumulation.
- Increase efforts to determine the biological effects of MeHg exposure on Arctic fish, marine mammals, and seabirds.

ANNEX 1

NCP III Quality Assurance and Quality Control Program for Mercury

Co-authors: Rania Farag, Eric Reiner and Sathi Selliah

A1.1 Introduction

An integral component of the Northern Contaminants Program (NCP), the Quality Assurance/Quality Control (QA/QC) Program ensures that analytical data of defined and consistent quality are being generated from monitoring and research activities. Private, government, and academic laboratories are involved in the provision of analytical data to the NCP. During Phase III of the NCP, the focus of the QA/QC Program was on evaluating the precision and bias of data produced by these analytical laboratories. These criteria were evaluated through the organization of a series of Interlaboratory Studies (ILSs).

The ILSs were conducted to routinely assess the analytical performances of laboratories for a broad range of contaminants, which included trace metals and persistent organic pollutants (POPs). This chapter focusses on the total mercury (THg) analysis component of the ILSs. Performances were evaluated based on the analysis of spiked standard solutions and natural-matrix samples of biological tissue.

A1.2 Participant profile

Formal invitations to participate in the interlaboratory studies were distributed to a wide range of national and international laboratories. Invitees included past participants of the NCP QA/QC Program, laboratories that participate in the Arctic Monitoring and Assessment Programme (AMAP) of the Arctic Council, and additional laboratories that have participated in other known proficiency testing or evaluation studies unrelated to the NCP or AMAP programs (Table A1.1). The latter laboratories were included to obtain more confidence in the statistics obtained and also to identify potential

TABLE A1.1 List of participating laboratories in the analysis of THg during the NCP III QA/QC Program. Laboratories that provided services to either the NCP or AMAP are identified.

Participating Laboratory	Country	ILSs Participation*	AMAP/NCP
ALS Laboratory Group-Environtmental Division (Edmonton)	Canada	Ph 2	
Centre de toxicologie du Québec/INSPQ	Canada	Ph 3, Ph 4	NCP
Centre for Environmental Chemistry of SPA "Typhoon"	Russia	Ph 4	AMAP
Estonian Environmental Centre	Estonia	Ph 5	AMAP
Flett Research Ltd.	Canada	Ph 2, Ph 3, Ph 4, Ph 5	
FNIHB Laboratory, Health Canada	Canada	Ph 2, Ph 3, Ph 4	
Great Lakes Institute for Environmental Research (GLIER), University of Windsor	Canada	Ph 3, Ph 4, Ph 5	NCP
Institut des sciences de la mer de Rimouski (ISMER)	Canada	Ph 3, Ph 4	
Maxxam Analytics	Canada	Ph 2, Ph 3, Ph 4	
National Environmental Research Institute, Aarhus University, Department of Marine Ecology	Denmark	Ph 4, Ph 5	AMAP
National Wildlife Research Centre, Environment Canada, Carleton University	Canada	Ph 2, Ph 3	NCP
Nunavik Research Centre, Makivik Corporation	Canada	Ph 2, Ph 3	NCP
Ontario Ministry of the Environment, Laboratory Services Branch	Canada	Ph 2, Ph 3, Ph 4, Ph 5	
Research Centre for Environmental Chemistry and Ecotoxicology (RECETOX), Masaryk University	Czech Republic	Ph 5	AMAP
SRC Analytical Laboratories	Canada	Ph 2, Ph 3, Ph 4, Ph 5	NCP
Umweltbundesamt Air Pollution Monitoring Network	Germany	Ph 5	AMAP
West Finland Regional Environment Centre	Finland	Ph 4, Ph 5	AMAP

* Note: Ph refers to the phase of the ILSs.

labs for future NCP work. There were approximately 30 participants per ILS and, on average, 37% of those participants performed THg analyses. The participating laboratories were provided with an identification code to maintain their confidentiality throughout the studies.

A1.3 Study design

Five rounds or phases of ILSs were conducted between 2005 and 2010. One year was dedicated to the completion of each phase. However, Hg was not included in Phase 1. The studies for Hg involved the analysis of THg in an injection-ready solution and in biological tissue. In Phases 2 to 5, participating laboratories performed analyses of an injection-ready commercial standard of inorganic Hg chloride spiked in acidified, deionized water to assess calibration and instrument performance. The inorganic Hg solution was acidified to a pH < 2 with nitric acid, coloured yellow with potassium dichromate solution, and homogenized for distribution to the laboratories. Phases 2 to 5 of the ILSs also involved analyses of natural-matrix certified reference material (CRM) and non-certified material used for Arctic research. The natural-matrix samples consisted mainly of fish tissue CRM's obtained from the National Research Council (NRC) Canada, the National Institute of Standards and Technology (NIST), and the Ontario Ministry of the Environment (MOE) (Table A1.2). In addition, samples of non-certified material—Arctic char and seal—used for Arctic research were obtained from Environment Canada. Laboratories were instructed to use their routine analytical methods for the sample analyses.

TABLE A1.2 Samples used for THg analysis in the series of ILSs.

Samples	Amount Provided	Producer	Phase 2	Phase 3	Phase 4	Phase 5	Reference
DOLT-3 *(Dogfish Liver)*	2.5 g	NRC	✖				NRC (2003a)
TORT-2 *(Lobster Hepatopancreas)*	2.5 g	NRC		✖			NRC (2003b)
DORM-3 *(Fish Protein CRM)*	2.5 g	NRC			✖		NRC (2008)
SRM 1566b *(Oyster Tissue Freeze Dried)*	2.5 g	NIST	✖		✖		NIST (2001)
SRM 1946 *(Lake Superior Fish Tissue)*	8 g	NIST		✖			NIST (2004)
SRM 1947 *(Lake Michigan Fish Tissue)*	8 g	NIST			✖	✖	NIST (2007)
WMF-01 *(Freeze Dried Fish Tissue)*	5 g	MOE	✖				OME (2002)
Arctic Char *(Non-Certified Material)*	10 g	Environment Canada		✖		✖	
Ringed Seal Muscle *Non-Certified Material)*	12 g	Environment Canada		✖			
Spiking Standard	40 mL	MOE	✖*	✖*	✖	✖	

* Note: The design of the spiking standards used for Phases 2 and 3 did not include Hg.

A1.4 Data evaluation methods

Analytical results were evaluated against the certified values of THg concentration for all CRMs. For non-certified material where reference values were not available, a statistical method outlined by the Royal Society of Chemistry was employed (AMC 2001). By this procedure, the median of all laboratory values was used to estimate the *true* value of the matrix, and the standard deviation was estimated as 1.5 times the Median Absolute Difference (MAD). The MAD was determined by first calculating the absolute differences between each result and the median value, and then taking the median of the absolute differences. The standard deviation estimated by the MAD was then used to calculate the percent relative standard deviation (% RSD). This method offered a robust approach for characterizing data from a small sample size without the removal of questionable data and for extreme results to have no impact on the final statistic. A statistical analysis was performed when results from at least three laboratories were received. Any data reported as not analyzed, not detected, or with a qualifier symbol (e.g., < 0.08) were omitted from the analysis.

A preliminary report of results submitted from all laboratories was distributed to the participants for verification of their reported data. Only a few laboratories submitted data corrections for their results and these were implemented before the final data analysis.

Upon completion of each ILS phase, a final report containing the data analysis with a graphical presentation of results was provided to all participating laboratories and NCP management.

A1.5 Study results

The laboratories as a group showed no significant bias for the analysis of THg (Fig. A1.1). Study averages were within 8% of the reference values for all the samples with known Hg concentrations. There was lower variability in the analysis of the natural-matrix samples—especially for CRMs—than for the injection-ready spiked solutions. The % RSDs for natural-matrix CRMs were less than 10%, with the exception of Ph 2–DOLT-3 which was 19% (Fig. A1.2). The aqueous samples in Phase 2 (Ph 2–TMStd) and Phase 3 (Ph 3–TMStd) did not contain any known values of Hg but a few laboratories reported values ranging from 0.00118–0.912 mg L^{-1} and 0.000420–0.00390 mg L^{-1}, respectively. As a result, relatively high % RSD values were observed in those samples (Fig. A1.2). For the aqueous samples in Phase 4 (Ph 4–Hg-1, Ph 4–Hg-2) and Phase 5 (Ph 5–Hg-1), the % RSD values ranged from 19–52%. The cause of this high variation is unknown and should be investigated.

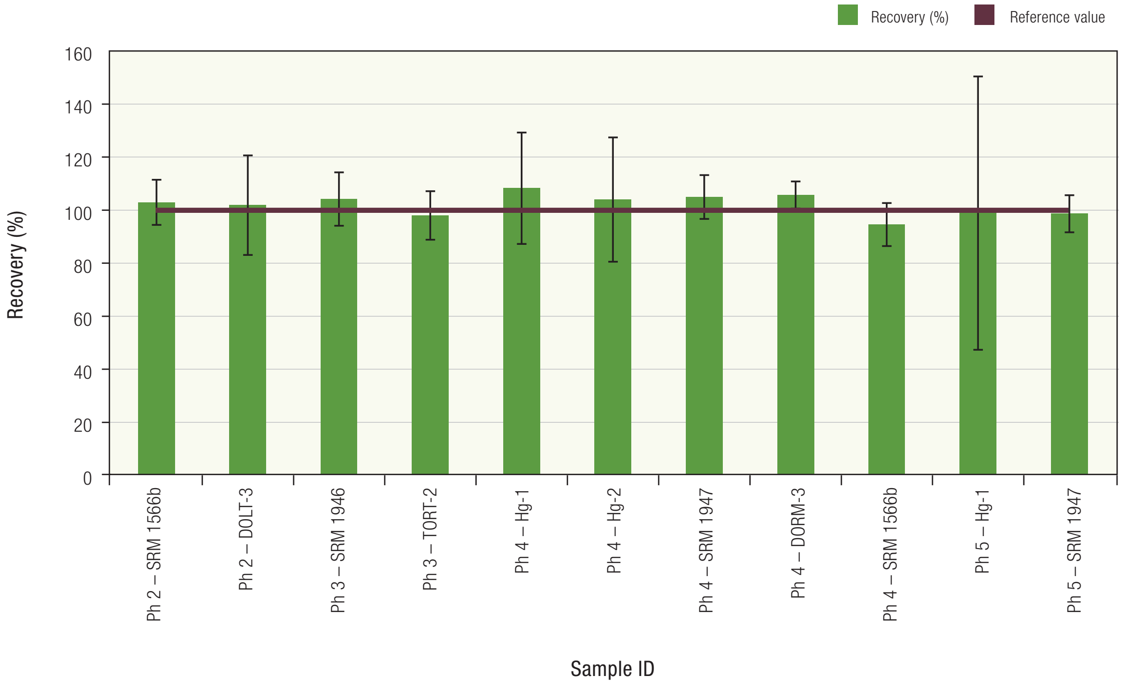

FIGURE A1.1

Average percent recoveries and RSDs of THg concentrations in CRMs and spiked solutions analyzed by participating laboratories in the QA/QC Program.

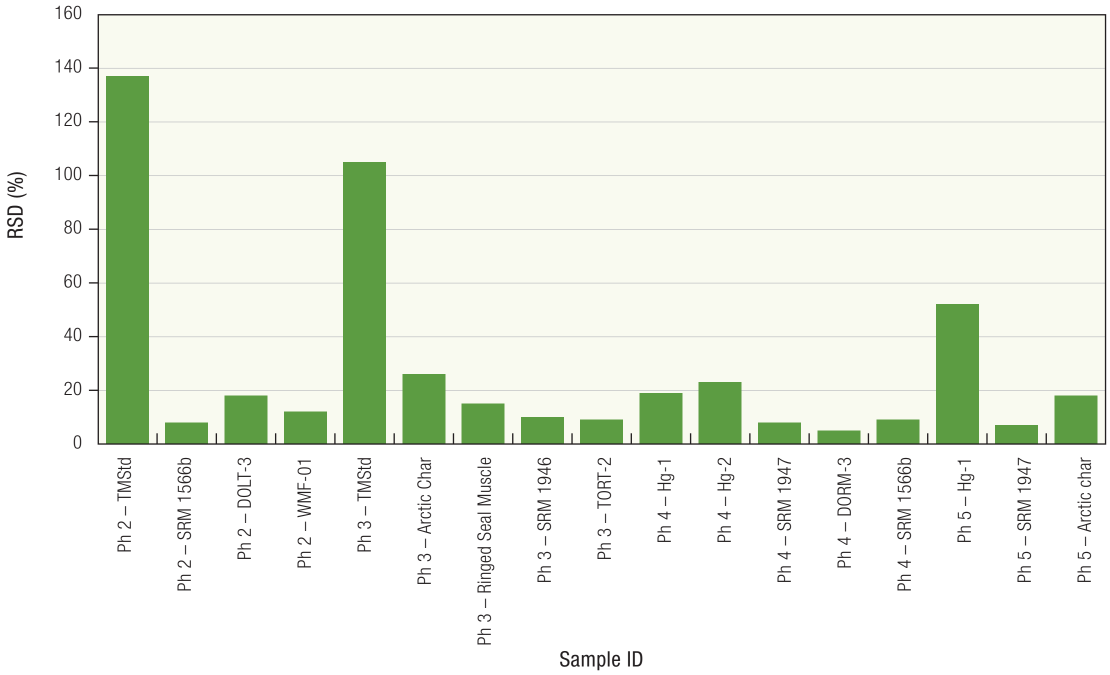

FIGURE A1.2

Percent RSDs of THg concentrations in natural-matrix samples and spiked solutions analyzed by participating laboratories in the QA/QC Program.

A1.6 Laboratory performances

The overall performance of each laboratory was evaluated as the proportion of analytical results within 20% of the median result measured by participating laboratories for each matrix and each study phase (Table A1.3). A result within 20% of the median value for the study was deemed an acceptable level of variability. Most laboratories that participated in more than one study phase showed acceptable performance in the analysis for THg (i.e. the majority of their results were within 20% of study median values). Five NCP laboratories participated in the QA/QC Program for the analysis of THg, although two of those participated in only two study rounds. Over the four study rounds, four out of the five NCP laboratories showed an overall improvement in their performance in THg analysis from round to round.

A1.7 Summary and recommendations

The QA/QC Program demonstrated that laboratories who participated in Phase III of the NCP were capable of producing satisfactory results for THg analysis of biological tissue. It also showed that, on average, there was no significant bias in the THg results.

The QA/QC Program focussed on the analysis of THg in biological material, mainly fish tissue, because it was extensively analysed by the majority of laboratories. However, monitoring and research activities conducted under the NCP are increasingly focussing on the analysis of other forms of Hg as well as abiotic matrices. Future QA/QC activities under the NCP should consider including other analyses that are routinely performed such as for methylmercury as well as other matrices such as sediment or water.

TABLE A1.3 Summary of laboratory performances in the analysis of THg, evaluated as the number of results that were within 20% of study median values per total number of analyses reported.

Lab Participants*	Number of Results Within ± 20% Study Median/Number of Results Reported			
	Phase 2	Phase 3	Phase 4	Phase 5
Lab 1		3/5	1/5	
Lab 2	3/4	4/4	5/5	2/3
Lab 3	1/3			
Lab 4 (N)	0/1	0/3	1/4	3/3
Lab 5 (N)	2/4	5/5		
Lab 6	2/4	3/4	4/4	2/3
Lab 7	2/4	4/5	2/5	
Lab 8 (N)	3/3	4/5	4/5	2/2
Lab 9 (N)		4/5	5/5	
Lab 10	2/3	5/5	5/5	
Lab 11 (N)		3/4	2/5	2/3
Lab 12 (A)			2/5	
Lab 13 (A)			2/5	1/3
Lab 14 (A)			5/5	1/2
Lab 15 (A)				0/1
Lab 16 (A)				0/1
Lab 17 (A)				3/3

* NCP laboratories are designated with (N), AMAP laboratories are designated with (A).

A1.8 References

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NIST. SRM 1947, Lake Michigan Fish Tissue. National Institute of Standards and Technology (NIST), Gaithersburg, Maryland, 2007.

NRC. DOLT-3, Dogfish Liver Certified Reference Material for Trace Metals. National Research Council (NRC) Canada, Ottawa, Canada, 2003a.

NRC. TORT-2, Lobster Hepatopancreas Reference Material for Trace Metals. National Research Council (NRC) Canada, Ottawa, Canada, 2003b.

NRC. DORM-3, Fish Protein Certified Reference Material for Trace Metals. National Research Council (NRC) Canada, Ottawa, Canada, 2008.

OME. WMF-01, Reference Fish Tissue for Organic Contaminant Analysis. Laboratory Services Branch, Ontario Ministry of the Environment (OME), Etobicoke, Canada, 2002.

ANNEX 2

Annex 2: Data Tables

TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Vegetation															
Water sedge	*Carex aquatilis*	Chapman Lake, YT		2008				dry	6 ± 2	ng g^{-1}	5	0.25 ± 0.10	ng g^{-1}	5	1
Water sedge	*Carex aquatilis*	North Slope, YT		2008				dry	5 ± 1	ng g^{-1}	3	0.60 ± 0.51	ng g^{-1}	3	1
Common cottongrass	*Eriophorum angustifolium*	North Slope, YT		2008				dry	5 ± 1	ng g^{-1}	4	0.30 ± 0.14	ng g^{-1}	4	1
Tussock cottongrass	*Eriophorum vaginatum*	Chapman Lake, YT		2008				dry	11 ± 11	ng g^{-1}	5	0.20 ± 0.21	ng g^{-1}	5	1
Tussock cottongrass	*Eriophorum vaginatum*	North Slope, YT		2008				dry	11 ± 4	ng g^{-1}	4	0.31 ± 0.17	ng g^{-1}	4	1
Purple saxifrage	*Saxifraga oppositifolia*	Cape Herschel, Devon Island, NU		2005–2007				dry	35	ng g^{-1}	1				2
Purple saxifrage	*Saxifraga oppositifolia*	Cape Vera, Devon Island, NU		2005–2007				dry	48 ± 39	ng g^{-1}	15				2
Tufted saxifrage	*Saxifraga caespitosa*	Cape Herschel, Devon Island, NU		2005–2007				dry	15	ng g^{-1}	1				2
Tufted saxifrage	*Saxifraga caespitosa*	Cape Vera, Devon Island, NU		2005–2007				dry	33 ± 13	ng g^{-1}	15				2
Grayleaf willow	*Salix glauca*	North Slope, YT		2008				dry	5 ± 2	ng g^{-1}	4	1.10 ± 0.54	ng g^{-1}	4	1
Tealeaf willow	*Salix pulchra*	Chapman Lake, YT		2008				dry	6 ± 1	ng g^{-1}	5	0.31 ± 0.25	ng g^{-1}	5	1
Crinkled snow lichen	*Flavocetraria nivalis*	North Slope, YT		2008				dry	49 ± 5	ng g^{-1}	1	2.53 ± 0.02	ng g^{-1}	1	1
Curled snow lichen	*Flavocetraria cucullata*	Chapman Lake, YT		2008				dry	72 ± 86	ng g^{-1}	5	1.79 ± 1.96	ng g^{-1}	5	1
Jewel lichen	*Xanthoria elegans*	Cape Herschel, Devon Island, NU		2005–2007				dry	199 ± 18	ng g^{-1}	3				2
Jewel lichen	*Xanthoria elegans*	Cape Vera, Devon Island, NU		2005–2007				dry	231 ± 74	ng g^{-1}	15				2
Reindeer lichen	*Cladina mitis*	Chapman Lake, YT		2008				dry	107 ± 123	ng g^{-1}	5	2.07 ± 2.63	ng g^{-1}	5	1
Worm lichen	*Thamnolia vermicularis*	Cape Herschel, Devon Island, NU		2005–2007				dry	93 ± 19	ng g^{-1}	3				2
Worm lichen	*Thamnolia vermicularis*	Cape Vera, Devon Island, NU		2005–2007				dry	176 ± 78	ng g^{-1}	14				2

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TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Birds															
Snow bunting	*Plectrophenax nivalis*	Devon Island, NU		2006–2007			whole body	dry	0.18 ± 0.13	μg g^{-1}	25				2
Small mammals															
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	juvenile		muscle	dry	0.002	μg g^{-1}	7				3
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	juvenile		liver	dry	0.02 ± 0.01	μg g^{-1}	7				3
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	juvenile		kidney	dry	0.20 ± 0.14	μg g^{-1}	7				3
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	adult		muscle	dry	0.002 ± 0.0003	μg g^{-1}	9				3
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	adult		liver	dry	0.17 ± 0.10	μg g^{-1}	9				3
Arctic hare	*Lepus arctica*	Baker Lake, NU		2003	adult		kidney	dry	0.54 ± 0.29	μg g^{-1}	9				3
Muskrat	*Ondatra zibethicus*	Old Crow, YT		2007–2009			muscle	wet	0.001 ± 0.0006	μg g^{-1}	31				4
Northern collared lemming	*Dicrostonyx groenlandicus*	Devon Island, NU		2006–2007			whole body	dry	0.10 ± 0.14	μg g^{-1}	5				2
Common shrew	*Sorex cinereus*	Faro, YT		2004–2005			whole body	dry	0.14 ± 0.08	μg g^{-1}	5				5
Hoary marmot	*Marmota caligata*	Faro, YT		2005		M	kidney	dry	0.049	μg g^{-1}	1				5
Hoary marmot	*Marmota caligata*	Faro, YT		2005		M	muscle	dry	< 0.01	μg g^{-1}	1				5
Hoary marmot	*Marmota caligata*	Faro, YT		2005		M	liver	dry	< 0.01	μg g^{-1}	1				5
Ground squirrel	*Tamias minimus*	Faro, YT		2005	adult	M	kidney	dry	0.089	μg g^{-1}	1				5
Ground squirrel	*Tamias minimus*	Faro, YT		2005	adult	M	muscle	dry	< 0.01	μg g^{-1}	1				5
Ground squirrel	*Tamias minimus*	Faro, YT		2005	adult	M	liver	dry	0.05	μg g^{-1}	1				5
Northern red-backed vole	*Clethrionomys rutilus*	Faro, YT		2004			muscle	dry	0.14 ± 0.06	μg g^{-1}	4				5
Northern red-backed vole	*Clethrionomys rutilus*	Faro, YT		2004			liver	dry	0.44 ± 0.13	μg g^{-1}	4				5
Northern red-backed vole	*Clethrionomys rutilus*	Faro, YT		2004			kidney	dry	2.94	μg g^{-1}	4				5
Deer mouse	*Peromyscus maniculatus*	Faro, YT		2005			muscle	dry	< 0.05	μg g^{-1}	9				5
Deer mouse	*Peromyscus maniculatus*	Faro, YT		2004–2005			liver	dry	< 0.02	μg g^{-1}	10				5
Deer mouse	*Peromyscus maniculatus*	Faro, YT		2004–2005			kidney	dry	< 0.05	μg g^{-1}	10				5

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TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Least chipmunk	*Neotamias minimus*	Faro, YT		2005			muscle	dry	< 0.05	μg g^{-1}	2				5
Least chipmunk	*Neotamias minimus*	Faro, YT		2005			liver	dry	< 0.02	μg g^{-1}	2				5
Least chipmunk	*Neotamias minimus*	Faro, YT		2005			kidney	dry	< 0.05	μg g^{-1}	2				5
Ermine	*Mustela erminea arctica*	Devon Island, NU		2007			whole body	dry	0.20 ± 0.04	μg g^{-1}	2				2
Mink	*Mustela vison*	Watson Lake, YT		2002	1.5	F	kidney	wet	0.81 ± 0.42	μg g^{-1}	19	0.75 ± 0.29	μg g^{-1}	10	6
Mink	*Mustela vison*	Watson Lake, YT		2002	1.5	M	kidney	wet	0.54 ± 0.48	μg g^{-1}	48	0.89 ± 0.50	μg g^{-1}	10	6
Mink	*Mustela vison*	Watson Lake, YT		2002	1.5	F	liver	wet	1.18 ± 0.71	μg g^{-1}	19	1.05 ± 0.47	μg g^{-1}	10	6
Mink	*Mustela vison*	Watson Lake, YT		2002	1.5	M	liver	wet	0.70 ± 0.74	μg g^{-1}	48	1.13 ± 0.72	μg g^{-1}	10	6
Mink	*Mustela vison*	Watson Lake, YT		2002	1.4	F	brain	wet	0.29 ± 0.16	μg g^{-1}	10	0.27 ± 0.24	μg g^{-1}	5	6
Mink	*Mustela vison*	Watson Lake, YT		2002	1.4	M	brain	wet	0.18 ± 0.16	μg g^{-1}	10	0.21 ± 0.10	μg g^{-1}	5	6
Ungulates															
Caribou	*Rangifer tarandus*	Haines Junction, YT		2002–2003	4.9	M	kidney	dry	1.46 ± 0.54	μg g^{-1}	7				7
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2005		M	kidney	dry	1.77 ± 0.55	μg g^{-1}	22				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2006		F	kidney	dry	5.30 ± 1.82	μg g^{-1}	25				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2007	3.0	F	kidney	dry	1.40 ± 0.49	μg g^{-1}	5				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2007	2.7	M	kidney	dry	1.41 ± 0.97	μg g^{-1}	11				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2008	7.2	F	kidney	dry	4.15 ± 1.47	μg g^{-1}	33				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2008	6.3	M	kidney	dry	2.35 ± 0.86	μg g^{-1}	11				8
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2009	5.9	F	kidney	dry	4.86 ± 1.81	μg g^{-1}	18				7
Caribou	*Rangifer tarandus*	Bathurst Inlet, NT		2009	2.0	M	kidney	dry	4.06 ± 0.38	μg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Fort Smith, NT	Beverly	2008	6.3	F	kidney	dry	5.71 ± 1.46	μg g^{-1}	10				8
Caribou	*Rangifer tarandus*	Fort Smith, NT	Beverly	2008	4.0	M	kidney	dry	7.22	μg g^{-1}	1				8
Caribou	*Rangifer tarandus*	Sahtu, NT		2005	6.8	F	kidney	dry	4.49 ± 2.02	μg g^{-1}	5				8

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TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Caribou	*Rangifer tarandus*	Sahtu, NT		2006	4.6	M	kidney	dry	4.67 ± 0.97	µg g^{-1}	15				8
Caribou	*Rangifer tarandus*	Carcoss, YT		2002		F	kidney	dry	5.61 ± 1.10	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Dawson, YT	Clear Creek	2006	3.8	M	kidney	dry	1.61 ± 0.16	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Watson Lake, YT	Coal River	2002	5.0	M	kidney	dry	1.86	µg g^{-1}	1				7
Caribou	*Rangifer tarandus*	Dawson, YT		2006	5.8	F	kidney	dry	3.80 ± 1.50	µg g^{-1}	17				9
Caribou	*Rangifer tarandus*	Dawson, YT		2006	3.7	M	kidney	dry	2.70 ± 0.20	µg g^{-1}	3				9
Caribou	*Rangifer tarandus*	Whitehorse, YT		2005			kidney	dry	0.10	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Nain, NL	George River	2008	4.9	F	kidney	dry	5.15 ± 1.31	µg g^{-1}	15				8
Caribou	*Rangifer tarandus*	Nain, NL	George River	2008	2.7	M	kidney	dry	4.40 ± 0.82	µg g^{-1}	6				8
Caribou	*Rangifer tarandus*	Dawson, YT	Hart River	2006	3.0	M	kidney	dry	1.73	µg g^{-1}	1				7
Caribou	*Rangifer tarandus*	Carmacks, YT	Klaza	2002–2005	4.4	M	kidney	dry	0.88 ± 0.13	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Watson Lake, YT	Little	2002	5.0	M	kidney	dry	2.69	µg g^{-1}	1				7
Caribou	*Rangifer tarandus*	Watson Lake, YT		2003	4.0	M	kidney	dry	1.04	µg g^{-1}	1				7
Caribou	*Rangifer tarandus*	Faro, YT	Pelly	2003	5.3	M	kidney	dry	2.73 ± 1.41	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Dawson, YT		2002	5.6	F	kidney	dry	1.64 ± 0.33	µg g^{-1}	12				10
Caribou	*Rangifer tarandus*	Dawson, YT		2002	5.5	M	kidney	dry	1.39 ± 0.27	µg g^{-1}	9				10
Caribou	*Rangifer tarandus*	Dawson, YT		2003	6.0	F	kidney	dry	2.52 ± 1.00	µg g^{-1}	23				10
Caribou	*Rangifer tarandus*	Dawson, YT		2003	5.7	M	kidney	dry	1.19 ± 0.25	µg g^{-1}	23				10
Caribou	*Rangifer tarandus*	Dawson, YT		2004	5.5	F	kidney	dry	2.02 ± 0.30	µg g^{-1}	4				7
Caribou	*Rangifer tarandus*	Dawson, YT		2004	4.8	M	kidney	dry	1.62 ± 0.59	µg g^{-1}	16				7

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TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Caribou	*Rangifer tarandus*	Dawson, YT		2005	7.3	F	kidney	dry	3.28 ± 1.17	$\mu g\ g^{-1}$	4				8
Caribou	*Rangifer tarandus*	Dawson, YT		2005	3.8	M	kidney	dry	1.81 ± 0.33	$\mu g\ g^{-1}$	14				8
Caribou	*Rangifer tarandus*	Dawson, YT		2006	4.0	F	kidney	dry	3.21 ± 1.72	$\mu g\ g^{-1}$	3				8
Caribou	*Rangifer tarandus*	Dawson, YT		2006	6.1	M	kidney	dry	2.12 ± 0.57	$\mu g\ g^{-1}$	9				8
Caribou	*Rangifer tarandus*	Dawson, YT		2007	7.0	F	kidney	dry	1.25	$\mu g\ g^{-1}$	1				8
Caribou	*Rangifer tarandus*	Dawson, YT		2007	4.3	M	kidney	dry	1.72 ± 0.51	$\mu g\ g^{-1}$	6				8
Caribou	*Rangifer tarandus*	Dawson, YT		2008	6.1	M	kidney	dry	1.34 ± 0.60	$\mu g\ g^{-1}$	20				11
Caribou	*Rangifer tarandus*	Dawson, YT		2009	6.3	M	kidney	dry	0.94 ± 0.39	$\mu g\ g^{-1}$	20				7
Caribou	*Rangifer tarandus*	Arviat, NU		2006	7.3	F	kidney	dry	3.37 ± 0.96	$\mu g\ g^{-1}$	7				8
Caribou	*Rangifer tarandus*	Arviat, NU		2006	5.8	M	kidney	dry	2.58 ± 0.86	$\mu g\ g^{-1}$	14				8
Caribou	*Rangifer tarandus*	Arviat, NU		2007	5.1	F	kidney	dry	5.57 ± 2.33	$\mu g\ g^{-1}$	10				8
Caribou	*Rangifer tarandus*	Arviat, NU		2007	4.0	M	kidney	dry	4.23 ± 1.62	$\mu g\ g^{-1}$	8				8
Caribou	*Rangifer tarandus*	Arviat, NU		2008	8.1	F	kidney	dry	5.11 ± 1.85	$\mu g\ g^{-1}$	7				11
Caribou	*Rangifer tarandus*	Arviat, NU		2008	5.4	M	kidney	dry	3.28 ± 1.53	$\mu g\ g^{-1}$	10				11
Caribou	*Rangifer tarandus*	Dawson, YT		2003	3.3	M	kidney	dry	1.85 ± 0.68	$\mu g\ g^{-1}$	2				7
Caribou	*Rangifer tarandus*	Ross River, YT	Tay	2002–2004	3.2	M	kidney	dry	1.10 ± 0.48	$\mu g\ g^{-1}$	10				7
Caribou	*Rangifer tarandus*	Ross River, YT	Wolf Lake	2002–2003	5.4	M	kidney	dry	1.25 ± 0.39	$\mu g\ g^{-1}$	2				7
Caribou	*Rangifer tarandus*	Carmacks, YT	Klaza	2002–2004	4.4	M	liver	dry	0.09 ± 0.01	$\mu g\ g^{-1}$	3				7
Caribou	*Rangifer tarandus*	Dawson, YT		2005	3.2	M	liver	dry	0.31 ± 0.06	$\mu g\ g^{-1}$	5				7
Caribou	*Rangifer tarandus*	Ross River, YT	Tay	2004	2.6	M	liver	dry	0.11 ± 0.10	$\mu g\ g^{-1}$	4				7

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TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Caribou	*Rangifer tarandus*	Haines Junction, YT	Aishihik	2002–2006	4.2	M	muscle	wet	3.66 ± 1.76	ng g^{-1}	12				7
Caribou	*Rangifer tarandus*	Carcoss, YT		2002–2006	4.9	F	muscle	wet	6.14 ± 2.00	ng g^{-1}	9				7
Caribou	*Rangifer tarandus*	Dawson, YT	Clear Creek	2004–2006	3.8	M	muscle	wet	2.60 ± 0.90	ng g^{-1}	3				7
Caribou	*Rangifer tarandus*	Watson Lake, YT	Coal River	2000–2005	4.6	M	muscle	wet	3.58 ± 0.30	ng g^{-1}	3				7
Caribou	*Rangifer tarandus*	Carmacks, YT	Klaza	2002–2004	5.0	M	muscle	dry	0.02 ± 0.01	µg g^{-1}	2				7
Caribou	*Rangifer tarandus*	Watson Lake, YT		2003–2004	3.3	M	muscle	wet	2.70 ± 0.10	ng g^{-1}	2				7
Caribou	*Rangifer tarandus*	Dawson, YT		2003	6.4	M	muscle	wet	1.78 ± 0.80	ng g^{-1}	4				7
Caribou	*Rangifer tarandus*	Dawson, YT		2004	5.4	M	muscle	wet	1.75 ± 1.10	ng g^{-1}	2				7
Caribou	*Rangifer tarandus*	Dawson, YT		2005	4.0	M	muscle	wet	1.90 ± 0.50	ng g^{-1}	7				7
Caribou	*Rangifer tarandus*	Dawson, YT		2009	6.0	M	muscle	dry				5.63 ± 1.78	ng g^{-1}	9	7
Caribou	*Rangifer tarandus*	Ross River, YT		2003	2.9	M	muscle	wet	2.50 ± 1.80	ng g^{-1}	2				7
Caribou	*Rangifer tarandus*	Whitehorse, YT		2004	8.0	F	muscle	wet	6.60	ng g^{-1}	1				7
Caribou	*Rangifer tarandus*	Ross River, YT	Tay	2002–2006	3.9	M	muscle	wet	4.40 ± 2.40	ng g^{-1}	7				7
Dall sheep	*Ovis dalli*	Whitehorse, YT		2004	9.0	M	kidney	dry	0.25	µg g^{-1}	1				7
Dall sheep	*Ovis dalli*	Whitehorse, YT		2004	10.0	M	liver	dry	< 0.05	µg g^{-1}	1				7
Elk	*Cervus elaphus*	YT		2002	5.0	M	kidney	dry	0.21	µg g^{-1}	1				7
Moose	*Alces alces*	YT		2002	5.1	M	kidney	dry	0.06 ± 0.04	µg g^{-1}	59				10
Moose	*Alces alces*	YT		2002	3.7	M	liver	dry	< 0.05	µg g^{-1}	5				7
Moose	*Alces alces*	YT		2002	4.8	M	muscle	wet	0.88 ± 0.96	ng g^{-1}	20				7
Moose	*Alces alces*	YT		2003	4.7	M	kidney	dry	0.06 ± 0.04	µg g^{-1}	42				10
Moose	*Alces alces*	YT		2003	4.4	M	muscle	wet	1.10 ± 1.14	ng g^{-1}	20				7
Moose	*Alces alces*	YT		2004	4.8	M	kidney	dry	0.03	µg g^{-1}	3				7
Moose	*Alces alces*	YT		2004	3.8	M	liver	dry	< 0.05	µg g^{-1}	3				7
Moose	*Alces alces*	YT		2004	5.4	M	muscle	wet	0.57 ± 0.78	ng g^{-1}	9				7
Moose	*Alces alces*	YT		2005	1.9	M	kidney	dry	0.26 ± 0.16	µg g^{-1}	2				7

Continued on next page

TABLE A2.1 Mercury concentrations in terrestrial vegetation, birds, small mammals, and ungulates in the Canadian Arctic.

Common Name	Species	Location	Herd	Sampling Year	Average Age (years)	Sex	Tissue	Tissue Weight	THg (mean ± SD)	THg Units	Sample Size	MeHg (mean ± SD)	MeHg Units	Sample Size	Reference
Moose	*Alces alces*	YT		2005	2.9	M	liver	dry	< 0.05	μg g^{-1}	4				7
Moose	*Alces alces*	YT		2005	5.9	M	muscle	wet	1.78 ± 1.32	ng g^{-1}	7				7
Moose	*Alces alces*	Mackenzie Valley, NT		2006	5.9	F	kidney	dry	0.09 ± 0.06	μg g^{-1}	14				11
Moose	*Alces alces*	Mackenzie Valley, NT		2006	5.9	F	liver	dry	0.04 ± 0.04	μg g^{-1}	14				11
Moose	*Alces alces*	Mackenzie Valley, NT		2006	3.6	M	kidney	dry	0.12 ± 0.11	μg g^{-1}	29				11
Moose	*Alces alces*	Mackenzie Mountains, NT		2006	7.4	M	kidney	dry	0.18 ± 0.08	μg g^{-1}	18				11
Moose	*Alces alces*	Mackenzie Valley, NT		2006	3.6	M	liver	dry	0.04 ± 0.04	μg g^{-1}	29				11
Moose	*Alces alces*	Mackenzie Mountains, NT		2006	7.4	M	liver	dry	0.12 ± 0.07	μg g^{-1}	18				11
Moose	*Alces alces*	YT		2006	5.5	M	muscle	wet	0.55 ± 0.63	ng g^{-1}	15				7
Moose	*Alces alces*	YT		2008	6.6	M	kidney	dry	0.01 ± 0.03	μg g^{-1}	21				11
Moose	*Alces alces*	YT		2009	4.8	M	muscle	dry				4.30 ± 1.74	ng g^{-1}	5	7
Mule deer	*Odocoileus hemionus*	YT		2002	3.6	F	kidney	dry	0.12	μg g^{-1}	1				7
Mule deer	*Odocoileus hemionus*	YT		2002	3.8	M	kidney	dry	0.24 ± 0.18	μg g^{-1}	2				7

Notes:

YT = Yukon Territory, NU = Nunavut, NT = Northwest Territories, F = female, M = male, THg = total mercury, SD = standard deviation, MeHg = methylmercury

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TABLE A2.2 Mercury concentrations in freshwater fish in the Canadian Arctic.

Common Name	Species	Location	Latitude (° N)	Longitude (° W)	Years Sampled	Sample Size	Length-Adjusted Mean THg (µg g^{-1} ww)	Length Used to Adjust THg (cm)	Unadjusted Mean THg (µg g^{-1} ww)	Site Mean Length (cm)	Reference
Arctic char (landlocked)	*Salvelinus alpinus*	Amituk Lake, NU	75.04	-93.78	2006, 2007	10	1.08	37.71	1.59	39.96	1
Arctic char (landlocked)	*Salvelinus alpinus*	Angajurjualuk Lake, NU	71.13	-79	2006, 2007	28	0.17	37.71	0.33	40.79	2
Arctic char (landlocked)	*Salvelinus alpinus*	Aquiatusuk Lake, NU	74.69	-94.23	2002, 2005	10	0.12	37.71	0.36	44.07	1
Arctic char (landlocked)	*Salvelinus alpinus*	Boomerang Lake, NU	73.94	-92.89	2007	8	0.08	37.71	0.30	46.13	1
Arctic char (landlocked)	*Salvelinus alpinus*	Camp Lake, NU	71.32	-79.4	2006	10	0.15	37.71	0.15	35.19	2
Arctic char (landlocked)	*Salvelinus alpinus*	Char Lake, NU	74.7	-94.89	2005, 2007	10	0.45	37.71	0.51	38.68	1
Arctic char (landlocked)	*Salvelinus alpinus*	Gavia Faeces, NU	68.34	-107.73	2006	8	0.25	37.71	0.16	27.45	1
Arctic char (landlocked)	*Salvelinus alpinus*	Keyhole Lake, NU	69.38	-106.24	2006	7	0.06	37.71	0.12	42.74	1
Arctic char (landlocked)	*Salvelinus alpinus*	Laflamme Lake, QC	61.32	-73.71	2007	7	0.11	37.71	0.14	51.71	1
Arctic char (landlocked)	*Salvelinus alpinus*	Lake D, NU	82.11	-67.48	2007	7	0.09	37.71	0.10	33.27	1
Arctic char (landlocked)	*Salvelinus alpinus*	Lake F, NU	81.81	-69.35	2007	8	0.29	37.71	0.17	26.21	1
Arctic char (landlocked)	*Salvelinus alpinus*	Lake G, NU	81.82	-69.18	2007	10	0.16	37.71	0.18	28.45	1
Arctic char (landlocked)	*Salvelinus alpinus*	Lake Hazen, NU	81.83	-70.42	2006	10	0.05	37.71	0.10	32.97	1
Arctic char (landlocked)	*Salvelinus alpinus*	Little Nauyuk Lake, NU	68.34	-107.75	2006	9	0.12	37.71	0.12	34.80	1
Arctic char (landlocked)	*Salvelinus alpinus*	Mary Lake, NU	71.24	-79.48	2006, 2007	32	0.14	37.71	0.28	41.08	2
Arctic char (landlocked)	*Salvelinus alpinus*	Meretta Lake, NU	74.68	-94.92	2006	10	0.22	37.71	0.32	45.82	1
Arctic char (landlocked)	*Salvelinus alpinus*	Nine Mile Lake, NU	74.81	-95.2	2006	10	0.19	37.71	0.18	31.37	1
Arctic char (landlocked)	*Salvelinus alpinus*	North Lake, NU	74.77	-95.09	2005, 2006	10	0.32	37.71	0.26	34.98	1
Arctic char (landlocked)	*Salvelinus alpinus*	Notgordie Lake, NU	68.35	-107.66	2006	5	0.37	37.71	0.23	44.24	1
Arctic char (landlocked)	*Salvelinus alpinus*	Pingaluk Lake, QC	61.28	-73.66	2007	10	0.16	37.71	0.17	38.28	1
Arctic char (landlocked)	*Salvelinus alpinus*	Resolute Lake, NU	74.68	-94.93	2006	10	0.23	37.71	0.25	38.24	1
Arctic char (landlocked)	*Salvelinus alpinus*	Second Portage Lake, NU	65.01	-96.02	2008	15	0.08	37.71	0.08	35.38	3
Arctic char (landlocked)	*Salvelinus alpinus*	Sheardown Lake, NU	71.31	-79.31	2006, 2007	26	0.12	37.71	0.16	35.57	2
Arctic char (landlocked)	*Salvelinus alpinus*	Small Lake, NU	74.76	-95.06	2006, 2007	10	0.11	37.71	0.15	38.31	1
Arctic char (landlocked)	*Salvelinus alpinus*	Twelve Mile Lake, NU	74.82	-95.34	2006	10	0.16	37.71	0.13	33.37	1
Burbot	*Lota lota*	Great Slave Lake (All), NT	61.5	-114	2002, 2003, 2004	5	0.02	59.46	0.07	68.85	4
Burbot	*Lota lota*	Great Slave Lake (Fort Resolution), NT	61.1	-113.8	2002, 2003	10	0.13	59.46	0.15	63.90	5

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TABLE A2.2 Mercury concentrations in freshwater fish in the Canadian Arctic.

Common Name	Species	Location	Latitude (° N)	Longitude (° W)	Years Sampled	Sample Size	Length-Adjusted Mean THg (µg g^{-1} ww)	Length Used to Adjust THg (cm)	Unadjusted Mean THg (µg g^{-1} ww)	Site Mean Length (cm)	Reference
Burbot	*Lota lota*	Great Slave Lake (Lustel K'e), NT	62.41	-110.75	2002	10	0.12	59.46	0.11	53.17	5
Burbot	*Lota lota*	Kusawa Lake, YT	60.34	-136.35	2002-2009	22	0.34	59.46	0.27	48.13	6
Burbot	*Lota lota*	Lake Laberge, YT	61.18	-135.2	2002-2009	56	0.48	59.46	0.47	55.92	6, 7
Burbot	*Lota lota*	Mackenzie River (Fort Good Hope), NT	66.28	-128.63	2009	39	0.38	59.46	0.40	70.35	7
Burbot	*Lota lota*	Slave River (Fort Smith), NT	60.01	-115.88	2002	10	0.11	59.46	0.12	61.76	5
Lake trout	*Salvelinus namaycush*	Cigar Lake, NU	66	-111.67	2005	9	0.30	54.41	0.17	41.54	8
Lake trout	*Salvelinus namaycush*	Cli Lake, NT	61.97	-123.31	2006	17	0.90	54.41	2.20	66.77	5
Lake trout	*Salvelinus namaycush*	Colville Lake, NT	67.18	-126	2005	20	0.26	54.41	0.27	56.63	5
Lake trout	*Salvelinus namaycush*	Counts Lake, NT	64.64	-110.28	2007	9	0.11	54.41	0.10	48.50	9
Lake trout	*Salvelinus namaycush*	Cujo Lake, NT	64.58	-110.18	2007	4	0.43	54.41	0.19	41.05	9
Lake trout	*Salvelinus namaycush*	Fox Lake, YT	61.23	-135.46	2007	5	0.50	54.41	0.27	38.49	7
Lake trout	*Salvelinus namaycush*	Fox Lake (below Laberge), YT	60.65	-134.07	2007	5	0.44	54.41	0.27	41.30	6
Lake trout	*Salvelinus namaycush*	Gagnon Lake (east), NT	61.91	-110.1	2004	7	0.27	54.41	0.27	47.35	10
Lake trout	*Salvelinus namaycush*	Gagnon Lake (west), NT	61.97	-110.58	2004	28	0.27	54.41	0.19	35.39	10
Lake trout	*Salvelinus namaycush*	Great Bear Lake, NT	65.18	-122.3	2002	10	0.13	54.41	0.34	67.41	5
Lake trout	*Salvelinus namaycush*	Great Slave Lake (All), NT	61.5	-114	2002, 2003, 2004	24	0.08	54.41	0.15	64.16	4
Lake trout	*Salvelinus namaycush*	Great Slave Lake (Hay ’River), NT	61.1	-115.83	2002	10	0.13	54.41	0.15	57.73	5
Lake trout	*Salvelinus namaycush*	Great Slave Lake (Lustel K'e), NT	62.41	-110.75	2002	10	0.11	54.41	0.13	54.22	5
Lake trout	*Salvelinus namaycush*	Jericho Lake, NU	66.03	-111.48	2005	10	0.41	54.41	0.31	48.07	8
Lake trout	*Salvelinus namaycush*	Kelly Lake, NT	65.38	-126.09	2007	19	0.75	54.41	0.87	57.78	5
Lake trout	*Salvelinus namaycush*	Kennady Lake, NT	63.43	-109.21	2004, 2006	21	0.19	54.41	0.26	56.31	11
Lake trout	*Salvelinus namaycush*	Kirk Lake, NT	63.7	-109.12	2005	25	0.54	54.41	0.60	55.18	11
Lake trout	*Salvelinus namaycush*	Kodiak Lake, NT	64.7	-110.62	2007	6	0.48	54.41	0.41	47.96	9
Lake trout	*Salvelinus namaycush*	Kusawa Lake, YT	60.34	-136.35	2002–2009	97	0.35	54.41	0.30	40.65	6, 7, 12
Lake trout	*Salvelinus namaycush*	Lac Belot, NT	66.93	-126.32	2007	19	0.17	54.41	0.30	60.88	5
Lake trout	*Salvelinus namaycush*	Lac de Gras, NT	64.5	-110.2	2008, 2009	30	0.16	54.41	0.33	60.13	13
Lake trout	*Salvelinus namaycush*	Lac du Sauvage, NT	64.57	-110.03	2008	20	0.17	54.41	0.33	64.92	13

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TABLE A2.2 Mercury concentrations in freshwater fish in the Canadian Arctic.

Common Name	Species	Location	Latitude (° N)	Longitude (° W)	Years Sampled	Sample Size	Length-Adjusted Mean THg (µg g^{-1} ww)	Length Used to Adjust THg (cm)	Unadjusted Mean THg (µg g^{-1} ww)	Site Mean Length (cm)	Reference
Lake trout	*Salvelinus namaycush*	Lac Ste Therese, NT	64.63	-121.58	2002–2004	10	0.14	54.41	0.83	70.03	5
Lake trout	*Salvelinus namaycush*	Lake 410, NT	63.56	-109.06	2004	24	0.30	54.41	0.29	52.12	11
Lake trout	*Salvelinus namaycush*	Lake Laberge, YT	61.18	-135.2	2002–2009	57	0.38	54.41	0.40	50.09	6, 7
Lake trout	*Salvelinus namaycush*	Lake N16, NT	63.46	-109.32	2004	25	0.26	54.41	0.30	54.55	11
Lake trout	*Salvelinus namaycush*	Leslie Lake, NT	64.68	-110.66	2007–2008	19	0.15	54.41	0.14	54.80	9, 14
Lake trout	*Salvelinus namaycush*	Moose Lake, NT	64.68	-110.64	2007	7	0.23	54.41	0.18	45.68	9
Lake trout	*Salvelinus namaycush*	Nanuq Lake, NT	64.91	-110.29	2007, 2008	25	0.15	54.41	0.14	51.40	9, 14
Lake trout	*Salvelinus namaycush*	Nema Lake, NT	64.66	-110.71	2007	6	0.43	54.41	0.37	49.63	9
Lake trout	*Salvelinus namaycush*	Nonacho Lake, NT	61.75	-109.5	2003, 2004	61	0.33	54.41	0.30	53.31	10, 15
Lake trout	*Salvelinus namaycush*	Quiet Lake, YT	61.08	-133.08	2002, 2003	23	0.29	54.41	0.43	62.11	6, 7
Lake trout	*Salvelinus namaycush*	Roberts Lake, NU	68.16	-106.53	2002	30	0.12	54.41	0.20	62.92	16
Lake trout	*Salvelinus namaycush*	Rutledge Lake, NT	61.61	-110.78	2003, 2004	71	0.23	54.41	0.21	53.78	10, 15
Lake trout	*Salvelinus namaycush*	Second Portage Lake, NU	65.01	-96.02	2008	18	0.34	54.41	0.47	57.36	3
Lake trout	*Salvelinus namaycush*	Slipper Lake, NT	64.61	-110.85	2007	15	0.32	54.41	0.29	47.81	9
Lake trout	*Salvelinus namaycush*	Sparks Lake, NT	61.16	-109.75	2004	28	0.13	54.41	0.12	45.85	10
Lake trout	*Salvelinus namaycush*	Stark Lake, NT	62.46	-110.33	2003	26	0.19	54.41	0.40	67.51	5
Lake trout	*Salvelinus namaycush*	Talston River (Methlaka Lake), NT	60.45	-111.12	2003, 2004	3	0.36	54.41	0.33	50.90	10
Lake trout	*Salvelinus namaycush*	Talston River (Nonacho Lake Dam), NT	61.66	-109.94	2003, 2004	20	0.27	54.41	0.23	58.02	10, 15
Lake trout	*Salvelinus namaycush*	Taltson Lake, NT	61.49	-110.27	2004	67	0.27	54.41	0.33	56.80	10, 15
Lake trout	*Salvelinus namaycush*	Teslin Lake, YT	60.17	-132.83	2008	10	0.10	54.41	0.12	58.90	6
Lake trout	*Salvelinus namaycush*	Travaillant Lake, NT	67.68	-131.68	2006	33	0.16	54.41	0.23	59.66	5
Lake trout	*Salvelinus namaycush*	Trout Lake, NT	60.52	-121.17	2003	27	0.36	54.41	0.34	61.80	5
Lake trout	*Salvelinus namaycush*	Upper Exeter Lake (Fay Bay), NT	64.75	-110.69	2008	20	0.33	54.41	0.27	47.83	17
Lake trout	*Salvelinus namaycush*	Vulture Lake, NT	64.75	-110.53	2007	17	0.07	54.41	0.11	46.03	9
Lake whitefish	*Coregonus clupeaformis*	Earn Lake, YT	62.83	-134.28	2002, 2004	13	0.03	39.53	0.04	27.61	6
Lake whitefish	*Coregonus clupeaformis*	Fox Lake (below Laberge), YT	60.65	-134.07	2007	4	0.33	39.53	0.20	50.62	6
Lake whitefish	*Coregonus clupeaformis*	Gagnon Lake (east), NT	61.91	-110.1	2004	28	0.07	39.53	0.08	31.30	10
Lake whitefish	*Coregonus clupeaformis*	Gagnon Lake (west), NT	61.97	-110.58	2004	7	0.09	39.53	0.09	39.55	10

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TABLE A2.2 Mercury concentrations in freshwater fish in the Canadian Arctic.

Common Name	Species	Location	Latitude (° N)	Longitude (° W)	Years Sampled	Sample Size	Length-Adjusted Mean THg (µg g^{-1} ww)	Length Used to Adjust THg (cm)	Unadjusted Mean THg (µg g^{-1} ww)	Site Mean Length (cm)	Reference
Lake whitefish	*Coregonus clupeaformis*	Great Slave Lake (All), NT	61.5	-114	2002, 2003, 2004	5	0.05	39.53	0.05	41.42	4
Lake whitefish	*Coregonus clupeaformis*	Lake Laberge, YT	61.18	-135.2	2002–2009	4	0.11	39.53	0.13	39.62	6
Lake whitefish	*Coregonus clupeaformis*	Narrow Lake, NT	63.16	-113.94	2005	8	0.51	39.53	0.63	35.32	18
Lake whitefish	*Coregonus clupeaformis*	Nonacho Lake, NT	61.75	-109.5	2003, 2004	62	0.10	39.53	0.13	50.74	10, 15
Lake whitefish	*Coregonus clupeaformis*	Rutledge Lake, NT	61.61	-110.78	2003, 2004	70	0.05	39.53	0.06	42.02	10, 15
Lake whitefish	*Coregonus clupeaformis*	Sparks Lake, NT	61.16	-109.75	2004	35	0.05	39.53	0.07	44.23	10
Lake whitefish	*Coregonus clupeaformis*	Tadru Lake, YT	62.47	-135.69	2002	6	0.08	39.53	0.08	38.08	6
Lake whitefish	*Coregonus clupeaformis*	Talston River (Methlaka Lake), NT	60.45	-111.12	2003, 2004	24	0.09	39.53	0.10	37.15	10, 15
Lake whitefish	*Coregonus clupeaformis*	Talston River (Natla Falls), NT	60.51	-111.61	2003	3	0.07	39.53	0.10	44.76	15
Lake whitefish	*Coregonus clupeaformis*	Talston River (Nonacho Lake Dam), NT	61.66	-109.94	2003, 2004	4	0.04	39.53	0.09	50.93	10, 15
Lake whitefish	*Coregonus clupeaformis*	Talston River (Oracha Falls), NT	61.06	-112.56	2003, 2004	8	0.05	39.53	0.07	41.56	10, 15
Lake whitefish	*Coregonus clupeaformis*	Talston River (Rat River), NT	61.1	-112.58	2003, 2004	22	0.04	39.53	0.03	38.40	10, 15
Lake whitefish	*Coregonus clupeaformis*	Talston River (Twin Gorges Forebay 1), NT	60.42	-111.38	2004	18	0.23	39.53	0.09	16.63	10
Lake whitefish	*Coregonus clupeaformis*	Talston River (Twin Gorges Forebay 2), NT	60.47	-111.29	2004	17	0.17	39.53	0.11	22.14	10
Lake whitefish	*Coregonus clupeaformis*	Taltson Lake, NT	61.49	-110.27	2003, 2004	70	0.05	39.53	0.05	40.93	10, 15
Northern pike	*Exos lucius*	Cadzow Lake, YT	67.55	-138.98	2006	5	0.44	59.89	0.13	45.60	6
Northern pike	*Exos lucius*	Earn Lake, YT	62.83	-134.28	2002, 2004	13	0.15	59.89	0.11	53.46	6
Northern pike	*Exos lucius*	Great Slave Lake (All), NT	61.5	-114	2002, 2003, 2004	22	0.22	59.89	0.23	61.16	4
Northern pike	*Exos lucius*	Great Slave Lake (Fort Resolution), NT	61.1	-113.8	2002, 2003	10	0.19	59.89	0.29	70.77	5
Northern pike	*Exos lucius*	Great Slave Lake (Lustel K'e), NT	62.41	-110.75	2002	10	0.16	59.89	0.17	61.58	5
Northern pike	*Exos lucius*	Hanson Lake, YT	64.02	-135.35	2007	10	0.11	59.89	0.21	67.15	6
Northern pike	*Exos lucius*	Kakisa Lake, NT	60.93	-117.71	2002, 2003	6	0.33	59.89	0.33	60.75	4
Northern pike	*Exos lucius*	Narrow Lake, NT	63.16	-113.94	2005	5	0.95	59.89	0.98	59.26	18
Northern pike	*Exos lucius*	Winter Lake, NT	63.16	-113.91	2005	5	0.48	59.89	0.34	45.30	18
Round whitefish	*Prosopium cylindraceum*	Cigar Lake, NU	66	-111.67	2005	5	0.03	32.71	0.04	38.32	8
Round whitefish	*Prosopium cylindraceum*	Counts Lake, NT	64.64	-110.28	2007	18	0.03	32.71	0.03	32.81	9

Continued on next page

TABLE A2.2 Mercury concentrations in freshwater fish in the Canadian Arctic.

Common Name	Species	Location	Latitude (° N)	Longitude (° W)	Years Sampled	Sample Size	Length-Adjusted Mean THg (µg g^{-1} ww)	Length Used to Adjust THg (cm)	Unadjusted Mean THg (µg g^{-1} ww)	Site Mean Length (cm)	Reference
Round whitefish	*Prosopium cylindraceum*	Cujo Lake, NT	64.58	-110.18	2007	20	0.11	32.71	0.11	34.36	9
Round whitefish	*Prosopium cylindraceum*	Jericho Lake, NU	66.03	-111.48	2005	4	0.03	32.71	0.08	40.72	8
Round whitefish	*Prosopium cylindraceum*	Kennady Lake, NT	63.43	-109.21	2004, 2006	3	0.06	32.71	0.06	34.20	11
Round whitefish	*Prosopium cylindraceum*	Kodiak Lake, NT	64.7	-110.62	2007	19	0.08	32.71	0.07	28.16	9
Round whitefish	*Prosopium cylindraceum*	Leslie Lake, NT	64.68	-110.66	2007–2008	20	0.06	32.71	0.07	36.28	9
Round whitefish	*Prosopium cylindraceum*	Moose Lake, NT	64.68	-110.64	2007	10	0.08	32.71	0.09	35.25	9
Round whitefish	*Prosopium cylindraceum*	Nanuq Lake, NT	64.91	-110.29	2007, 2008	3	0.01	32.71	0.04	38.13	9
Round whitefish	*Prosopium cylindraceum*	Nema Lake, NT	64.66	-110.71	2007	19	0.11	32.71	0.13	34.35	9
Round whitefish	*Prosopium cylindraceum*	Second Portage Lake, NU	65.01	-96.02	2008	6	0.05	32.71	0.06	29.70	3
Round whitefish	*Prosopium cylindraceum*	Slipper Lake, NT	64.61	-110.85	2007	20	0.11	32.71	0.10	28.56	9
Round whitefish	*Prosopium cylindraceum*	Upper Exeter Lake (Fay Bay), NT	64.75	-110.69	2008	20	0.08	32.71	0.07	29.90	17
Walleye	*Sander vitreus*	Kakisa Lake, NT	60.93	-117.71	2002, 2003	12	0.20	41.31	0.21	43.07	4
Walleye	*Sander vitreus*	Talston River (Natla Falls), NT	60.51	-111.61	2003	4	0.18	41.31	0.17	38.52	15
Walleye	*Sander vitreus*	Talston River (Oracha Falls), NT	61.06	-112.56	2003, 2004	2	0.56	41.31	0.45	36.30	15

Notes:

YT = Yukon Territory, NU = Nunavut, NT = Northwest Territories, THg = total mercury, ww = wet weight

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TABLE A2.3 Mercury concentrations in freshwater invertebrates in the Canadian Arctic.

Invertebrate	Study Area	Number of Water Bodies	THg Range ($ng\ g^{-1}$ dw)	MeHg Range ($ng\ g^{-1}$ dw)	References
Benthic invertebrates	Mackenzie River Basin	5	7–414		1
Caddisflies	Kent Peninsula	1	271	133	2
Caddisflies	Victoria Island	1	38	20	2
Chironomids	Cornwallis Island	12	86–503	27–319	2, 3
Chironomids	Devon Island	23	26–360	10–268	3, 4
Chironomids	Ellesmere Island	2	94–263	24–51	2
Chironomids	Kent Peninsula	1		7	5
Chironomids	Melville Sound	5		5–13	5
Daphnia	Cornwallis Island	1	153	127–293	2, 6
Daphnia	Devon Island	2		264–302	6
Fairy shrimp	Cornwallis Island	1		113	2
Fairy shrimp	Devon Island	4		113–137	6
Isopods	Kent Peninsula	1		30	5
Isopods	Melville Sound	5		43–82	5
Mysids	Kent Peninsula	1		33	5
Mysids	Melville Sound	5		17–88	5
Zooplankton	Cornwallis Island	11	27–297	3–269	2, 6
Zooplankton	Devon Island	19	30–262	11–170	4, 6
Zooplankton	Ellesmere Island	4	32–113	1–35	2
Zooplankton	Kent Peninsula	4	22–89	2–35	2, 5
Zooplankton	Mackenzie River Basin	5	15–105		1
Zooplankton	Melville Sound	5		4–21	5
Zooplankton	Victoria Island	1	133	56	2

Notes:

THg = total mercury, MeHg = methylmercury, dw = dry weight

References:

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TABLE A2.4 Mercury concentrations in marine waters of the Arctic Archipelago and Hudson Bay. Data are from Kirk et al. (2008).

Site #	Latitude (° N)	Longitude (° W)	Depth (m)	THg (ng L^{-1})	GEM (pg L^{-1})	Methylated Hg (pg L^{-1})	Me$_2$Hg (pg L^{-1})	% Methylated Hg
North Open Water Polynya								
1	76.300	71.407	0	0.14	19.5	21.4	4.7	15.1
			250	0.16	8	81.2	86.2	51.4
			660	0.19	3.1	87.1	110	46.4
2	76.255	74.594	0	0.37	14.5	30.5	2.8	8
			200	0.19	49.0	85.1	67.6	44
			439	0.23	133	103	76.7	45
3	76.383	77.382	0	0.18	22.4	40.3	10.9	23
			200	0.23	27.7	64.1	56.7	28
			347	0.17	28.3	89.3	60.7	53
4	77.335	76.021	0	0.30	25.6	30.8	15.2	10
			180	0.36	43.4	85.9	42.2	24
			522	1.21	31.7	146.0	71.0	12
5	78.328	74.356	0	0.32	48.9	28.5	10.0	9
			140	0.20	47.8	77.6	35.3	39
			487	0.22	85.9	106	37.8	49
Northwest Passage								
6	75.240	74.982	0	1.26	25.1	15.0	4.5	1
			200	0.55	33.1	81.8	54.4	15
			470	0.31	24.1	112	76.1	36
7	74.051	79.921	0	2.08	20.0	15.0	4.0	1
			200	0.14	12.0	62.6	88.3	45
			808	0.27	9.3	99.7	92.8	37
8	74.267	91.197	0	0.45	39.8	31.6	14.3	7
			200	0.18	50.3	86.4	50.2	48
			322	0.26	12.2	90.7	83.5	35
9	73.654	96.284	0	0.38	20.3	68.0	52.3	18
			150	0.27	43.1	178	161	65
			240	0.24	13.7	158	170	67
10	69.167	100.700	0	0.53	25.6	15.0	5.8	3
			35	0.23	30.1	31.4	24.0	14
			54	0.23	23.2	48.4	36.5	22
11	68.678	103.869	0	0.44	17.3	15.0	6.7	3
			50	0.23	11.3	43.9	53.5	19
			102	0.26		46.3	67.0	18
Hudson Strait								
12	61.153	64.825	0	0.21	22.5	24.4	32.0	11.6
			75	0.16	73.9	55.6	3.9	35.2
			435	0.24	27.2	96.8	43.4	40.5
13	62.275	71.978	0					
			150	0.14	24.7	26.7	41.5	19
			330	0.20	25.7	60.8	49.1	31

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TABLE A2.4 Mercury concentrations in marine waters of the Arctic Archipelago and Hudson Bay. Data are from Kirk et al. (2008).

Site #	Latitude (° N)	Longitude (° W)	Depth (m)	THg (ng L^{-1})	GEM (pg L^{-1})	Methylated Hg (pg L^{-1})	Me$_2$Hg (pg L^{-1})	% Methylated Hg
14	64.029	79.215	0	0.15	24.2	23.0	3.1	15
			200	1.70	11.0	37.4	22.1	2
			304	0.30	5.6	30.1	24.4	10
Hudson Bay								
15	62.138	78.714	0	0.15	33.0	24.5	1.9	16
			70	0.19	18.2	34.5	54.8	18
			141	0.14	5.0	42.8	48.3	31
16	60.125	79.166	0	0.23	23.3	15.0	1.4	7
			75	0.19	ND	61.0	93.9	33
			128	0.24	29.5	107	82.0	45
17	58.419	78.332	0	0.34	41.2	15.9	3.4	5
			30	0.72	39.6	15.0	4.6	2
			104	0.52	ND	78.4	91.1	15
18	55.285	77.899	0	2.90	33.3	30.2	2.1	1
			45	0.26	31.8	19.3	32.4	7
			82	0.22	1.9	32.1	43.9	15
19	54.720	80.752	0	0.35	33.2	15.0	2.4	4
			50	0.26	22.3	39.9	40.6	15
			87	0.26	ND	64.0	81.4	25
20	55.429	80.487	0	0.24	29.0	15.0	1.4	6
			50	0.43	31.3	41.3	36.5	10
			84	0.39	9.7	55.8	58.5	14
21	58.398	83.292	0	0.95	25.6	15.0	2.6	2
			75	0.21	25.9	44.2	76.5	21
			168	0.27	12.0	126	129	46
22	57.574	91.612	0	0.28	32.6	41.9	0.1	15
			20	0.22	31.5	15.0	0.4	7
			60	1.36	32.4	24.5	1.5	2
23	59.060	87.482	0	0.20	28.0	22.2	0.1	11
			100	0.28	85.9	71.1	38.0	26
			188	0.26	81.9	63.5	22.9	25
24	59.041	94.001	0	0.35	30.5	18.7	0.5	5
			20	0.25	27.0	44.8	1.1	18
			45	0.22	41.0	27.6	3.2	12

Notes:

ND = concentration below the method detection limit, THg = total mercury, MeHg = methylmercury, Me$_2$Hg = dimethylmercury, GEM = gaseous elemental mercury

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Kirk JL, St. Louis VL, Hintelmann H, Lehnherr I, Else B, Poissant L. Methylated mercury species in marine waters of the Canadian high and sub Arctic. Environmental Science & Technology 2008; 42: 8367–8373.

TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Invertebrates												
Amphipod	*Themisto libellula*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE			whole body	dry	0.087 ± 0.007	(40)	1
Amphipod	*Themisto libellula*	Amundsen Gulf, NT	2002–2006		mean ± SE			whole body	dry	0.127 ± 0.019	(57)	1
Amphipod	*Anonyx* spp.	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE			whole body	dry	0.291 ± 0.050	(6)	1
Amphipod	*Acanthostepheia malmgreni*	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE			whole body	dry	0.116 ± 0.011	(2)	1
Amphipod	mixed	Nuvuk Islands, NU	2009		one measured value			whole body	dry	0.054	(1)	2
Clam	*Mya truncata*	Pangnirtung, NU	2007		mean ± SD			muscle	wet	0.009 ± 0.003	(6)	3
Copepod	*Calanus* spp.	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE			whole body	dry	0.025 ± 0.003	(8)	1
Copepod	*Calanus* spp.	Amundsen Gulf, NT	2002–2006		mean ± SE			whole body	dry	0.032 ± 0.002	(33)	1
Euphausiid		Nuvuk Islands, NU	2009		one measured value			whole body	dry	0.235	(1)	2
Jellyfish	mixed	Nuvuk Islands, NU	2009		mean ± SD			whole body	dry	0.014 ± 0.005	(2)	2
Mysids	mixed	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE			whole body	dry	0.081 ± 0.009	(15)	1
Sea angel	*Clione limacina*	Nuvuk Islands, NU	2009		one measured value			whole body	dry	0.087	(1)	2
Shrimp	*Eualus* spp., *Bythocaris* spp.	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE			whole body	dry	0.316 ± 0.061	(14)	1
Squid	*Gonatus* sp.	Coats Island, NU	2009		one measured value			whole body	dry	0.060	1	2
Zooplankton		Pangnirtung, NU	2007		mean ± SD			whole body	wet	0.027	(1)	3
Zooplankton		Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE			whole body	dry	0.035 ± 0.005	(8)	1
Fish												
Arctic char	*Salvelinus alpinus*	Pangnirtung, NU	2007		mean ± SD		43 ± 3	muscle	wet	0.017 ± 0.009	5	3
Arctic cisco	*Coregonus autumnalis*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	5.6 ± 0.5	31.2 ± 0.6	muscle	dry	0.133 ± 0.009	30	1
Arctic cod	*Boreogadus saida*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	2.6 ± 0.2	14.3 ± 0.3	muscle	dry	0.163 ± 0.016	20	1
Arctic cod	*Boreogadus saida*	Amundsen Gulf, NT	2002–2006		mean ± SE	3.4 ± 0.1	15.8 ± 0.01	muscle	dry	0.377 ± 0.029	60	1
Arctic cod	*Boreogadus saida*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.097 ± 0.055	4	2
Arctic flounder	*Pleuronectes glacialis*	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE	9.1 ± 0.6	22.6 ± 1.1	muscle	dry	0.255 ± 0.044	9	1
Arctic shanny	*Stichaeus punctatus*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.201 ± 0.033	2	2
Arctic skate	*Amblyraja hyperborea*	Pangnirtung, NU	2007		mean ± SD		53 ± 0	muscle	wet	0.295	1	3
Arctic staghorn sculpin	*Gymnocanthus tricuspis*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.101 ± 0.015	2	2
Banded gunnel	*Pholis fasciata*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.100 ± 0.004	3	2
Capelin	*Mallotus villosus*	Pangnirtung, NU	2007		mean ± SD		15 ± 1	muscle	wet	0.03 ± 0.015	3	3

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Capelin	*Mallotus villosus*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.071 ± 0.009	3	2
Daubed shanny	*Leptoclinus maculatus*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.118 ± 0.000	2	2
Eelpout	*Gymnelus viridis*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.158 ± 0.085	2	2
Fourhorn sculpin	*Myoxocephalus quadricornis*	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE		22.0 ± 3.2	muscle	dry	0.587 ± 0.076	5	1
Fourline snakeblenny	*Eumesogrammus praecisus*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.327 ± 0.115	4	2
Greenland shark	*Somniosus microcephalus*	Pangnirtung, NU	2007		mean ± SD		293 ± 11	muscle	wet	1.715 ± 0.457	5	3
Least cisco	*Coregonus sardinella*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	7.6 ± 0.8	28.5 ± 0.8	muscle	dry	0.133 ± 0.009	20	1
Pacific herring	*Clupea palasii*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	5.1 ± 0.6	20.7 ± 0.2	muscle	dry	0.123 ± 0.005	29	1
Rainbow smelt	*Osmerus mordax*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	6.1 ± 0.9	20.4 ± 1.6	muscle	dry	0.181 ± 0.028	10	1
Saffron cod	*Eliginus gracilis*	Beaufort Sea, estuarine shelf, NT	2002–2006		mean ± SE	5.8 ± 0.5	28.0 ± 1.1	muscle	dry	0.308 ± 0.029	20	1
Sandlance	*Ammodytes hexapterus*	Coats Island, NU	2009		mean ± SD			whole body	dry	0.101 ± 0.016	5	2
Short-horn sculpin	*Myoxocephalus scorpius*	Pangnirtung, NU	2007		mean ± SD		24 ± 7	muscle	wet	0.056 ± 0.05	9	3
Snailfish	*Liparis* sp.	Coats Island, NU	2009		one measured value			whole body	dry	0.156	1	2
Starry flounder	*Platichthys stellatus*	Beaufort Sea, epibenthic, NT	2002–2006		mean ± SE	9.0 ± 1.2	24.7 ± 1.4	muscle	dry	0.277 ± 0.073	11	1
Seabirds												
Arctic tern	*Sterna paradisaea*	Tern Island, N of Cornwallis Island, NU	2008		mean ± SE			egg*	dry	2.11 ± 0.13	17	4
Black guillemot	*Cepphus grylle*	Prince Leopold Island, NU	2004		mean ± SD			egg	dry	1.92 ± 0.37	15 (5)	5
Black guillemot	*Cepphus grylle*	Prince Leopold Island, NU	2008		mean ± SD			egg	dry	1.79 ± 0.14	9 (3)	6
Black guillemot	*Cepphus grylle*	St. Helena Island, NW of Devon Island, NU	2004		mean ± SD			egg	dry	2.65 ± 0.40	9 (3)	5
Black guillemot	*Cepphus grylle*	SE Southampton Island, NU	2004		mean ± SD			egg	dry	1.71 ± 0.65	9 (3)	5
Black guillemot	*Cepphus grylle*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			liver	dry	1.9, 1.4-3.6	8	7
Black guillemot	*Cepphus grylle*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			muscle	dry	0.9, 0.6-1.4	8	7
Black-legged kittiwake	*Rissa tridactyla*	Prince Leopold Island, NU	2003		mean ± SD			egg	dry	0.82 ± 0.09	12 (4)	8
Black-legged kittiwake	*Rissa tridactyla*	Prince Leopold Island, NU	2008		mean ± SD			egg	dry	0.80 ± 0.05	15 (5)	6
Common eider	*Somateria mollissima*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			liver	dry	1.2, 0.7-2.2	8	7

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (μg g^{-1})	Sample Size	Reference
Common eider	*Somateria mollissima*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			muscle	dry	0.4, 0.2-0.7	8	7
Common eider	*Somateria mollissima*	Tern Island, N of Cornwallis Island, NU	2008		mean ± SE			egg*	dry	0.86 ± 0.05	15	4
Common eider	*Somateria mollissima*	SE Southampton Island, NU	2008		mean ± SE			egg*	dry	0.50 ± 0.02	9	4
Common eider	*Somateria mollissima*	Queen Maud Gulf, NU	2001	F	mean, 95% CI	adult		blood	wet	0.13, 0.12-0.14	63	9
Common eider	*Somateria mollissima*	Queen Maud Gulf, NU	2002	F	mean, 95% CI	adult		blood	wet	0.18, 0.17-0.19	69	9
Common eider	*Somateria mollissima*	Queen Maud Gulf, NU	2003	F	mean, 95% CI	adult		blood	wet	0.17, 0.16-0.18	74	9
Glaucous gull	*Larus hyperboreus*	Prince Leopold Island, NU	2003		mean ± SD			egg	dry	2.57 ± 0.49	15 (5)	5
Glaucous gull	*Larus hyperboreus*	Prince Leopold Island, NU	2008		mean ± SD			egg	dry	2.31 ± 0.41	9 (3)	6
Glaucous gull	*Larus hyperboreus*	Resolute, Cornwallis Island, NU	2008		mean ± SD			egg	dry	0.89 ± 0.52	5	10
Glaucous gull	*Larus hyperboreus*	Karrak Lake, Queen Maud Gulf, NU	2004		mean ± SE	chick		liver	dry	0.26 ± 0.02	29	11
Glaucous gull	*Larus hyperboreus*	Devil Island, NW of Devon Island, NU	2005		mean ± SE	chick		liver	dry	2.80 ± 0.16	18	11
Glaucous gull	*Larus hyperboreus*	Devil Island, NW of Devon Island, NU	2006		mean ± SE	chick		liver	dry	1.50 ± 0.09	15	11
Glaucous gull	*Larus hyperboreus*	Qikiqtarjuaq area, E Baffin Island, NU	2001		one measured value			liver	dry	3.1	1	7
Glaucous gull	*Larus hyperboreus*	Qikiqtarjuaq area, E Baffin Island, NU	2001		one measured value			muscle	dry	1.1	1	7
Ivory gull	*Pagophila eburnea*	Seymour Island, NU	2004		mean ± SD			egg	dry	6.37 ± 5.17	6	5
Long-tailed duck	*Clangula hyemalis*	Tern Island, N of Cornwallis Island, NU	2008		mean ± SE			egg*	dry	0.85 ± 0.25	4	4
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2003		mean ± SD			egg	dry	1.41 ± 0.11	15 (5)	8
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2005		mean ± SD			egg	dry	0.96 ± 0.15	15 (5)	12
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2006		mean ± SD			egg	dry	1.17 ± 0.16	15 (5)	13
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2007		mean ± SD			egg	dry	1.13 ± 0.11	15 (5)	14
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2008		mean ± SD			egg	dry	1.15 ± 0.22	15 (5)	6
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2009		mean ± SD			egg	dry	1.17 ± 0.08	15 (5)	15
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2003	5F,10M	mean ± SD	adult		liver	dry	8.97 ± 3.13	15	16
Northern fulmar	*Fulmarus glacialis*	Prince Leopold Island, NU	2008	5F,5M	mean ± SD	adult		liver	dry	6.99 ± 3.59	10	17

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Northern fulmar	*Fulmarus glacialis*	NW Devon Island, NU	2003		mean ± SD			egg	dry	1.57 ± 0.27	15	5
Northern fulmar	*Fulmarus glacialis*	NW Devon Island, NU	2003	7F,8M	mean ± SD	adult		liver	dry	10.63 ± 2.64	15	16
Northern fulmar	*Fulmarus glacialis*	Minarets/Cape Searle, E Baffin Island, NU	2008	5F,5M	mean ± SD	adult		liver	dry	3.42 ± 1.68	10	17
Northern fulmar	*Fulmarus glacialis*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			liver	dry	0.8, 0.8-0.9	2	7
Northern fulmar	*Fulmarus glacialis*	Qikiqtarjuaq area, E Baffin Island, NU	2001		geometric mean, range			muscle	dry	0.1, 0.1-0.1	2	7
Ross' gull	*Rhodostethia rosea*	Tern Island, N of Cornwallis Island, NU	2008		one measured value			egg	dry	0.40	1	18
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2003		mean ± SD			egg	dry	1.33 ± 0.28	15 (5)	8
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2005		mean ± SD			egg	dry	1.06 ± 0.15	15 (5)	12
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2006		mean ± SD			egg	dry	1.33 ± 0.16	15 (5)	13
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2007		mean ± SD			egg	dry	1.30 ± 0.15	15 (5)	14
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2008		mean ± SD			egg	dry	1.43 ± 0.24	15 (5)	6
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2009		mean ± SD			egg	dry	1.49 ± 0.39	15 (5)	15
Thick-billed murre	*Uria lomvia*	Prince Leopold Island, NU	2008	5F,5M	mean ± SD	adult		liver	dry	4.13 ± 0.60	10	17
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2003		mean ± SD			egg	dry	0.56 ± 0.07	15 (5)	5
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2005		mean ± SD			egg	dry	0.68 ± 0.10	15 (5)	12
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2006		mean ± SD			egg	dry	0.76 ± 0.11	15 (5)	13
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2007		mean ± SD			egg	dry	0.76 ± 0.03	15 (5)	14
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2008		mean ± SD			egg	dry	0.86 ± 0.07	15 (5)	19
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2009		mean ± SD			egg	dry	0.60 ± 0.07	15 (5)	10
Thick-billed murre	*Uria lomvia*	Coats Island, NU	2007	5F	mean ± SD	adult		liver	dry	1.99 ± 0.48	5	17
Thick-billed murre	*Uria lomvia*	Minarets/Cape Searle, E Baffin Island, NU	2007	5F,5M	mean ± SD	adult		liver	dry	4.41 ± 1.03	10	17
Thick-billed murre	*Uria lomvia*	Akpatok Island, NU	2008	5F,5M	mean ± SD	adult		liver	dry	2.15 ± 0.54	10	17
Thick-billed murre	*Uria lomvia*	Digges Island, NU	2008	5F,5M	mean ± SD	adult		liver	dry	1.62 ± 0.38	10	17
Marine mammals												
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		muscle	wet	0.22 ± 0.02	4	20
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		muscle	wet	0.21 ± 0.09	2	20
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		muscle	wet	0.12 ± 0.01	6	20
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		liver	wet	8.36 ± 2.26	4	20
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		liver	wet	34.40 ± 25.1	2	20
Bearded seal	*Erignathus barbatus*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		liver	wet	31.10 ± 6.93	6	20
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2003	F	mean ± SE	19.7 ± 2.4	324 ± 7	muscle	wet	1.1 ± 0.1	13	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2003	M	mean ± SE	18.3 ± 3.1	334 ± 13	muscle	wet	0.68 ± 0.05	16	21

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2005	F	mean ± SE	17.0 ± 13.0		muscle	wet	0.75 ± 0.28	3	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2005	M	mean ± SE	21.3 ± 3.9		muscle	wet	1.3 ± 0.36	13	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2007	F	mean ± SE	31.0 ± 11.6	230 ± 14	muscle	wet	0.86 ± 0.2	3	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2007	M	mean ± SE	7.9 ± 3.3	329 ± 20	muscle	wet	0.78 ± 0.15	9	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2008	F	mean ± SE	23.7 ± 10.1	363 ± 16	muscle	wet	0.75 ± 0.08	6	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2008	M	mean ± SE	17.3 ± 6.5	363 ± 15	muscle	wet	0.98 ± 0.20	6	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2003	F	mean ± SE	19.7 ± 2.4	324 ± 7	liver	wet	17.7 ± 4.5	13	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2003	M	mean ± SE	19.3 ± 2.6	343 ± 12	liver	wet	5.8 ± 1.4	19	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2005	M	mean ± SE	21.3 ± 3.9		liver	wet	13.6 ± 4.7	12	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2007	F	mean ± SE	31.0 ± 11.6	230 ± 14	liver	wet	14.9 ± 5.0	3	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2007	M	mean ± SE	7.9 ± 3.3	329 ± 20	liver	wet	4.7 ± 1.5	9	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2008	F	mean ± SE	23.7 ± 10.1	363 ± 16	liver	wet	11.6 ± 4.4	6	21
Beluga whale	*Delphinapterus leucas*	Arviat, NU	2008	M	mean ± SE	17.3 ± 6.5	363 ± 15	liver	wet	8.9 ± 2.9	6	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2002	M	mean ± SE	22.5 ± 3.9	372 ± 32	muscle	wet	0.89 ± 0.28	6	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2003	F	mean ± SE	17.8 ± 8.0	273 ± 16	muscle	wet	0.58 ± 0.12	4	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2003	M	mean ± SE	17.9 ± 3.3	353 ± 33	muscle	wet	0.70 ± 0.14	7	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2004	F	mean ± SE	40.9 ± 7.2	375 ± 15	muscle	wet	1.2 ± 0.17	8	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2005	F	mean ± SE	32 ± 11.7		muscle	wet	1.2 ± 0.38	4	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2005	M	mean ± SE	22.2 ± 1.6		muscle	wet	0.92 ± 0.13	6	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2007	F	mean ± SE	16.3 ± 5.1	304 ± 20	muscle	wet	0.49 ± 0.06	3	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2007	M	mean ± SE	18.3 ± 5.2	355 ± 16	muscle	wet	0.95 ± 0.16	6	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2008	F	mean ± SE	18.5 ± 3.7	328 ± 2.6	muscle	wet	0.63 ± 0.02	5	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2008	M	mean ± SE	16.0 ± 2.7	362 ± 10	muscle	wet	0.73 ± 0.11	7	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2009		mean ± SD		341.7	muscle	wet	0.83 ± 0.46	18	22
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2002	M	mean ± SE	22.5 ± 3.9	372 ± 32	liver	wet	10.2 ± 4.3	6	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2003	F	mean ± SE	17.8 ± 8.0	273 ± 16	liver	wet	7.6 ± 3.6	4	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2003	M	mean ± SE	17.9 ± 3.3	353 ± 33	liver	wet	5.8 ± 1.2	7	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2004	F	mean ± SE	40.9 ± 7.2	375 ± 15	liver	wet	13.3 ± 1.5	8	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2005	F	mean ± SE	27.8 ± 11.6		liver	wet	18.2 ± 11.0	4	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2005	M	mean ± SE	22.5 ± 2.8		liver	wet	49.4 ± 3.6	8	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2007	F	mean ± SE	16.3 ± 5.1	305 ± 14	liver	wet	8.4 ± 4.1	4	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2007	M	mean ± SE	18.3 ± 5.2	366 ± 12	liver	wet	15.9 ± 4.1	9	21

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2008	F	mean ± SE	18.5 ± 3.7	328 ± 2.6	liver	wet	10.2 ± 2.9	5	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2008	M	mean ± SE	16.0 ± 2.7	362 ± 10	liver	wet	10.2 ± 4.0	7	21
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2009		mean ± SD		341.7	liver	wet	13.7 ± 11.3	18	22
Beluga whale	*Delphinapterus leucas*	Sanikiluaq, NU	2009		mean ± SD		341.7	kidney	wet	2.62 ± 1.40	18	22
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2009		mean ± SD		390.5	liver	wet	10.6 ± 15.4	5	22
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2009		mean ± SD		390.5	kidney	wet	3.83 ± 2.70	5	22
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2009		mean ± SD		390.5	muscle	wet	0.91 ± 0.36	4	22
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2008		mean ± SD	28.5		liver	wet	9.79 ± 5.71	4	23
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2008		mean ± SD	28.5		kidney	wet	5.12 ± 3.65	4	23
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2006		mean ± SD		98.2	liver	wet	16.26 ± 13.40	4	24
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2006		mean ± SD		98.2	kidney	wet	7.89 ± 4.69	4	24
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2007		mean ± SD		407.7	liver	wet	13.69 ± 6.61	4	24
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2007		mean ± SD		407.7	kidney	wet	6.79 ± 2.94	4	24
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2002		mean ± SD		400.3	liver	wet	11.74 ± 7.74	14	25
Beluga whale	*Delphinapterus leucas*	Pangnirtung, NU	2005		mean ± SD		380.7	liver	wet	8.88 ± 4.69	9	25
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2009		mean ± SD		391.9	liver	wet	22.1 ± 21.7	27	22
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2009		mean ± SD		391.9	kidney	wet	3.20 ± 2.28	27	22
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2009		mean ± SD		391.9	muscle	wet	0.94 ± 0.61	27	22
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2008		mean ± SD		402	liver	wet	22.7 ± 17.0	28	23
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2008		mean ± SD		400.9	kidney	wet	5.32 ± 2.80	27	23
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2008		mean ± SD		402	muscle	wet	1.14 ± 0.52	28	23
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2008		mean ± SD		402	muktuk	wet	0.37 ± 0.19	28	23
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2007		mean ± SD		416.6	liver	wet	27.51 ± 26.9	18	24
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2007		mean ± SD		416.6	kidney	wet	5.28 ± 3.65	18	24
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2007		mean ± SD		416.6	muscle	wet	1.16 ± 0.78	18	24
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2007		mean ± SD		416.6	muktuk	wet	0.57 ± 0.40	18	24
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2006		mean	25.5		liver	wet	24.0	35	26
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2002		mean ± SD		409.2	liver	wet	26.48 ± 27.62	24	25
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2003		mean ± SD			liver	wet	40.10 ± 34.71	19	25
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2004		mean ± SD		403.2	liver	wet	29.94 ± 28.07	26	25
Beluga whale	*Delphinapterus leucas*	Hendrickson Island, NT	2005		mean ± SD		406.7	liver	wet	21.74 ± 17.22	22	25

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Beluga whale	*Delphinapterus leucas*	Iqaluit, NU	2002		mean	6		liver	wet	5.8	1	26
Beluga whale	*Delphinapterus leucas*	Iqaluit, NU	2003		mean	4		liver	wet	2.5	1	26
Beluga whale	*Delphinapterus leucas*	Iqaluit, NU	2004		mean	22		liver	wet	22.8	6	26
Beluga whale	*Delphinapterus leucas*	Resolute Bay, NU	2003		mean ± SD		377.9	liver	wet	5.5 ± 4.28	5	25
Beluga whale	*Delphinapterus leucas*	Igloolik, NU	2005		mean ± SD		375.9	liver	wet	11.32 ± 11.16	5	25
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		muscle	wet	0.66 ± 0.13	10	20
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		muscle	wet	1.78 ± 0.38	4	20
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		muscle	wet	2.23 ± 1.06	2	20
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		liver	wet	3.42 ± 0.77	10	20
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		liver	wet	28.25 ± 13.14	4	20
Harbour seal	*Phoca vitulina*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		liver	wet	243.21 ± 105.85	2	20
Harp seal	*Phoca groenlandica*	Pangnirtung, NU	2007		mean ± SD		171 ± 26	muscle	wet	0.386 ± 0.301	9	3
Harp seal	*Phoca groenlandica*	Pangnirtung, NU	1999		geometric mean			liver	dry	38	18	27
Harp seal	*Phoca groenlandica*	Pangnirtung, NU	1999		geometric mean			muscle	dry	1.8	18	27
Narwhal	*Monodon monoceros*	Pangnirtung, NU	2007		mean ± SD		389.5	liver	wet	11.39 ± 7.56	8	24
Narwhal	*Monodon monoceros*	Pangnirtung, NU	2007		mean ± SD		389.5	kidney	wet	2.63 ± 2.15	6	24
Narwhal	*Monodon monoceros*	Pangnirtung, NU	2002		mean ± SD			liver	wet	10.35 ± 9.64	2	25
Narwhal	*Monodon monoceros*	Grise Fiord, NU	2007		mean ± SD		439	liver	wet	9.08 ± 5.08	12	24
Narwhal	*Monodon monoceros*	Grise Fiord, NU	2007		mean ± SD		439	kidney	wet	2.10 ± 1.00	13	24
Narwhal	*Monodon monoceros*	Grise Fiord, NU	2003		mean ± SD		463	liver	wet	9.20 ± 2.46	8	25
Narwhal	*Monodon monoceros*	Arctic Bay, NU	2004		mean ± SD		422.9	liver	wet	9.05 ± 6.70	14	25
Narwhal	*Monodon monoceros*	Broughton Island, NU	2004		mean ± SD		387.1	liver	wet	9.18 ± 6.13	10	25
Narwhal	*Monodon monoceros*	Clyde River, NU	2004		mean ± SD		376.8	liver	wet	5.95 ± 4.22	9	25
Narwhal	*Monodon monoceros*	Pond Inlet, NU	2004		mean ± SD		398.1	liver	wet	11.54 ± 6.46	12	25
Polar bear	*Ursus maritimus*	Baffin Bay, NU	2007–2008		mean ± SD	5.7 ± 2.2		liver	wet	25.52 ± 15.30	14	28
Polar bear	*Ursus maritimus*	Davis Strait, NU	2008		mean ± SD	5.3 ± 1.8		liver	wet	17.10 ± 14.25	8	28
Polar bear	*Ursus maritimus*	Gulf of Boothia, NU	2007		mean ± SD	10.0 ± 7.7		liver	wet	28.64 ± 7.87	6	28
Polar bear	*Ursus maritimus*	Lancaster/Jones Sound,NU	2007–2008		mean ± SD	6.5 ± 2.5		liver	wet	32.88 ± 15.67	13	28
Polar bear	*Ursus maritimus*	Northern Beaufort Sea, NT	2006–2007		mean ± SD	8.2 ± 5.3		liver	wet	75.97 ± 77.30	27	28
Polar bear	*Ursus maritimus*	Southern Beaufort Sea, NT	2006–2007		mean ± SD	10.5 ± 6.0		liver	wet	53.11 ± 35.33	11	28
Polar bear	*Ursus maritimus*	Southern Hudson Bay, NU	2007–2008		mean ± SD	9.4 ± 4.8		liver	wet	6.96 ± 4.31	14	28

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Polar bear	*Ursus maritimus*	Western Hudson Bay, NU	2007–2008		mean ± SD	9.5 ± 7.6		liver	wet	7.79 ± 5.14	11	28
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2002	F	mean ± SE	19.3 (11–26)		muscle	wet	0.44 ± 0.10	4	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2002	M	mean ± SE	15.2 (7–26)		muscle	wet	0.52 ± 0.04	13	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2003	F	mean ± SE	16.4 (11–26)		muscle	wet	0.44 ± 0.07	7	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2003	M	mean ± SE	19.2 (11–25)		muscle	wet	0.56 ± 0.07	11	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2004	F	mean ± SE	9.5 (7–12)		muscle	wet	0.29 ± 0.03	2	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2004	M	mean ± SE	16.5 (7–26)		muscle	wet	0.50 ± 0.05	15	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2005	F	mean ± SE	14.7 (7–21)		muscle	wet	0.41 ± 0.12	3	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2005	M	mean ± SE	15.6 (8–21)		muscle	wet	0.62 ± 0.07	14	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2007	F	mean ± SE	17.0 (8–26)		muscle	wet	0.46 ± 0.07	6	29
Ringed seal	*Phoca hispida*	Ulukhaktok, NT	2007	M	mean ± SE	15.8 (8–26)		muscle	wet	0.61 ± 0.07	11	29
Ringed seal	*Phoca hispida*	Pangnirtung, NU	2007		mean ± SD			muscle	wet	0.25 ± 0.20	3	3
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		muscle	wet	0.11 ± 0.01	9	20
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		muscle	wet	0.14 ± 0.02	4	20
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		muscle	wet	0.22 ± 0.07	3	20
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	pups		liver	wet	1.32 ± 0.19	9	20
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	juveniles		liver	wet	4.21 ± 1.61	4	20
Ringed seal	*Phoca hispida*	western Hudson Bay, NU	1999–2006		mean ± SE	adults		liver	wet	12.69 ± 2.95	3	20
Ringed seal	*Phoca hispida*	Nachvak Fiord, NFL	2008–2009		mean ± SD			muscle	wet	0.171 ± 0.107	25	30
Ringed seal	*Phoca hispida*	Saglek Fiord, NFL	2008–2009		mean ± SD			muscle	wet	0.170 ± 0.072	28	30
Ringed seal	*Phoca hispida*	Okak Fiord, NFL	2008–2009		mean ± SD			muscle	wet	0.230 ± 0.159	30	30
Ringed seal	*Phoca hispida*	Anaktalak Fiord, NFL	2008		mean ± SD			muscle	wet	0.128 ± 0.063	12	30
Ringed seal	*Phoca hispida*	Arctic Bay, NU	1999–2008		mean ± SD			liver	wet	6.71 ± 0.78	24	31
Ringed seal	*Phoca hispida*	Arviat, NU	1999–2008		mean ± SD			liver	wet	18.67 ± 0.98	91	31
Ringed seal	*Phoca hispida*	Gjoa Haven, NU	1999–2008		mean ± SD			liver	wet	9.57 ± 0.81	16	31
Ringed seal	*Phoca hispida*	Grise Fiord, NU	1999–2008		mean ± SD			liver	wet	28.73 ± 25.49	20	31
Ringed seal	*Phoca hispida*	Inukjuaq, QC	1999–2008		mean ± SD			liver	wet	2.74 ± 0.60	17	31
Ringed seal	*Phoca hispida*	Nain, NFL	1999–2008		mean ± SD			liver	wet	3.20 ± 0.75	25	31
Ringed seal	*Phoca hispida*	Pangnirtung, NU	1999–2008		mean ± SD			liver	wet	6.92 ± 0.81	59	31

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (μg g^{-1})	Sample Size	Reference
Ringed seal	*Phoca hispida*	Pond Inlet, NU	1999–2008		mean ± SD			liver	wet	7.44 ± 0.49	47	31
Ringed seal	*Phoca hispida*	Qikiqtarjuaq, NU	1999–2008		mean ± SD			liver	wet	9.11 ± 0.68	20	31
Ringed seal	*Phoca hispida*	Resolute Bay, NU	1999–2008		mean ± SD			liver	wet	10.91 ± 0.57	95	31
Ringed seal	*Phoca hispida*	Sachs Harbour, NU	1999–2008		mean ± SD			liver	wet	44.23 ± 0.97	91	31
Ringed seal	*Phoca hispida*	Ulukhaktok, NU	1999–2008		mean ± SD			liver	wet	35.34 ± 25.96	25	31
Ringed seal	*Phoca hispida*	Arctic Bay, NU	2000–2010		mean ± SD			muscle	wet	0.63 ± 0.78	24	31
Ringed seal	*Phoca hispida*	Arviat, NU	2000–2010		mean ± SD			muscle	wet	0.53 ± 0.97	94	31
Ringed seal	*Phoca hispida*	Gjoa Haven, NU	2000–2010		mean ± SD			muscle	wet	0.38 ± 0.81	16	31
Ringed seal	*Phoca hispida*	Grise Fiord, NU	2000–2010		mean ± SD			muscle	wet	0.34 ± 0.25	30	31
Ringed seal	*Phoca hispida*	Inukjuaq, QC	2000–2010		mean ± SD			muscle	wet	0.13 ± 0.59	18	31
Ringed seal	*Phoca hispida*	Kangiqsuluaq, QC	2000–2010		mean ± SD			muscle	wet	0.15 ± 0.71	6	31
Ringed seal	*Phoca hispida*	Nain, NFL	2000–2010		mean ± SD			muscle	wet	0.19 ± 0.75	31	31
Ringed seal	*Phoca hispida*	Pangnirtung, NU	2000–2010		mean ± SD			muscle	wet	0.27 ± 0.81	129	31
Ringed seal	*Phoca hispida*	Pond Inlet, NU	2000–2010		mean ± SD			muscle	wet	0.25 ± 0.49	25	31
Ringed seal	*Phoca hispida*	Qikiqtarjuaq, NU	2000–2010		mean ± SD			muscle	wet	0.49 ± 0.68	20	31
Ringed seal	*Phoca hispida*	Resolute Bay, NU	2000–2010		mean ± SD			muscle	wet	0.64 ± 0.58	79	31
Ringed seal	*Phoca hispida*	Sachs Harbour, NU	2000–2010		mean ± SD			muscle	wet	0.70 ± 0.97	82	31
Ringed seal	*Phoca hispida*	Ulukhaktok, NU	2000–2010		mean ± SD			muscle	wet	0.53 ± 0.13	20	31
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2004		mean ± SD			liver	wet	1.66 ± 0.92	16	26
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2007		mean ± SD			liver	wet	4.85 ± 4.02	7	24
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2007		mean ± SD			kidney	wet	0.35 ± 0.16	7	24
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2008		mean ± SD		305.5	liver	wet	3.08 ± 3.21	4	23
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2008		mean ± SD		305.5	kidney	wet	0.29 ± 0.09	4	23
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2009		mean ± SD		317.8	liver	wet	2.05 ± 1.32	21	22
Walrus	*Odobenus rosmarus*	Hall Beach, NU	2009		mean ± SD		317.8	kidney	wet	0.29 ± 0.12	21	22
Walrus	*Odobenus rosmarus*	Igloolik, NU	2008		mean ± SD		300.1	liver	wet	1.78 ± 2.95	7	23
Walrus	*Odobenus rosmarus*	Igloolik, NU	2008		mean ± SD		300.1	kidney	wet	0.30 ± 0.20	7	23
Walrus	*Odobenus rosmarus*	Igloolik, NU	2009		mean ± SD		300.6	liver	wet	1.62 ± 0.62	8	22
Walrus	*Odobenus rosmarus*	Igloolik, NU	2009		mean ± SD		300.6	kidney	wet	0.34 ± 0.60	9	22
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2009		mean ± SD		346.3	liver	wet	6.62 ± 4.26	3	22
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2009		mean ± SD		346.3	kidney	wet	0.51 ± 0.07	3	22
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2008		mean		365.8	liver	wet	6.87	1	23

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TABLE A2.5 Mercury concentrations in marine invertebrates, fish, seabirds, and mammals in the Canadian Arctic.

Common Name	Species	Location	Sampling Year	Sex	Statistic	Age (years)	Length (cm)	Tissue	Tissue Weight	THg (µg g^{-1})	Sample Size	Reference
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2008		mean		365.8	kidney	wet	1.04	1	23
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2007		mean		289.6	liver	wet	6.37	1	23
Walrus	*Odobenus rosmarus*	Grise Fiord, NU	2007		mean		289.6	kidney	wet	0.85	1	23

Notes: Sample size = number of individuals; number of sample pools is provided in brackets. NU = Nunavut, NT = Northwest Territories, NFL = Newfoundland and Labrador, F = female, M = male, SE = standard error, SD = standard deviation, CI = confidence interval

* Early egg

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