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Science of the Total Environment 351–352 (2005) 4–56

Science of the  
Total Environment

An International Journal for Scientific Research  
into the Environment and its Relationship with Humankind

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## Review

# Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: An overview of spatial and temporal trends

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Accepted 20 October 2004

Available online 16 August 2005

## Abstract

This review summarizes and synthesizes the significant amount of data which was generated on mercury (Hg) and persistent organic pollutants (POPs) in Canadian Arctic marine biota since the first Canadian Arctic Contaminants Assessment Report (CACAR) was published in 1997. This recent body of work has led to a better understanding of the current levels and spatial and temporal trends of contaminants in biota, including the marine food species that northern peoples traditionally consume. Compared to other circumpolar countries, concentrations of many organochlorines (OCs) in Canadian Arctic marine biota are generally lower than in the European Arctic and eastern Greenland but are higher than in Alaska, whereas Hg concentrations are substantially higher in Canada than elsewhere. Spatial coverage of OCs in ringed seals, beluga and seabirds remains a strength of the Arctic contaminant data set for Canada. Concentrations of OCs in marine mammals and seabirds remain fairly consistent

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across the Canadian Arctic although subtle differences from west to east and south to north are found in the proportions of various chemicals.

The most significant development since 1997 is improvement in the temporal trend data sets, thanks to the use of archived tissue samples from the 1970s and 1980s, long-term studies using archeological material, as well as the continuation of sampling. These data cover a range of species and chemicals and also include retrospective studies on new chemicals such as polybrominated diphenyl ethers. There is solid evidence in a few species (beluga, polar bear, blue mussels) that Hg at some locations has significantly increased from pre-industrial times to the present; however, the temporal trends of Hg over the past 20–30 years are inconsistent. Some animal populations exhibited significant increases in Hg whereas others did not. Therefore, it is currently not possible to determine if anthropogenic Hg is generally increasing in Canadian Arctic biota. It is also not yet possible to evaluate whether the recent Hg increases observed in some biota may be due solely to increased anthropogenic inputs or are in part the product of environmental change, e.g., climate warming. Concentrations of most “legacy” OCs (PCBs, DDT, etc.) significantly declined in Canadian Arctic biota from the 1970s to the late 1990s, and today are generally less than half the levels of the 1970s, particularly in seabirds and ringed seals. Chlorobenzenes and endosulfan were among the few OCs to show increases during this period while  $\sum$ HCH remained relatively constant in most species.

A suite of new-use chemicals previously unreported in Arctic biota (e.g., polybrominated diphenyl ethers (PBDEs), short chain chlorinated paraffins (SCCPs), polychlorinated naphthalenes (PCNs), perfluoro-octane sulfonic acid (PFOS) and perfluorocarboxylic acids (PFCAs)) has recently been found, but there is insufficient information to assess species differences, spatial patterns or food web dynamics for these compounds. Concentrations of these new chemicals are generally lower than legacy OCs, but there is concern because some are rapidly increasing in concentration (e.g., PBDEs), while others such as PFOS have unique toxicological properties, and some were not expected to be found in the Arctic because of their supposedly low potential for long-range transport. Continuing temporal monitoring of POPs and Hg in a variety of marine biota must be a priority.

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*Keywords:* Arctic; Canadian Arctic; Chemical contaminants; Marine ecosystem; Organochlorines; Mercury; PCBs; Seabirds; Beluga; Ringed seals; Temporal trends

## Contents

|        |   |    |
|--------|---|----|
| 1.     | Introduction . . . . .  | 6  |
| 2.     | Pathways and processes of delivery of contaminants to marine ecosystems . . . . . | 7  |
| 2.1.   | Transport of air-borne and sea-borne contaminants . . . . .                       | 7  |
| 2.2.   | Bioaccumulation and biomagnification processes . . . . .                          | 7  |
| 3.     | Spatial patterns of contaminants in marine biota . . . . .                        | 8  |
| 3.1.   | Contaminants in invertebrates and marine fish . . . . .                           | 8  |
| 3.1.1. | Mercury in invertebrates . . . . .  | 8  |
| 3.1.2. | Mercury in marine and anadromous fish . . . . .                                   | 9  |
| 3.1.3. | Organochlorines in invertebrates . . . . .  | 10 |
| 3.1.4. | Organochlorines in marine fish . . . . .  | 12 |
| 3.2.   | Levels and spatial trends of contaminants in seabirds . . . . .                   | 13 |
| 3.2.1. | Mercury in seabirds . . . . .   | 14 |
| 3.2.2. | Organochlorines in seabirds . . . . .   | 15 |
| 3.2.3. | New chemicals in seabirds . . . . .   | 16 |
| 3.2.4. | Chiral contaminants in seabirds . . . . .   | 17 |
| 3.3.   | Levels and spatial patterns of contaminants in marine mammals . . . . .           | 18 |
| 3.3.1. | Mercury in marine mammals . . . . .   | 18 |
| 3.3.2. | POPs in marine mammals . . . . .  | 20 |
| 3.3.3. | New compounds in marine mammals . . . . .   | 25 |
| 4.     | Arctic marine food web studies . . . . .  | 30 |
| 4.1.   | Trophic transfer of mercury in marine food webs . . . . .                         | 30 |

|        |  |    |
|--------|--|----|
| 4.1.1. | Mercury in the Resolute Bay food web . . . . .                               | 30 |
| 4.1.2. | Mercury in the food web of northern Baffin Bay . . . . .                     | 30 |
| 4.2.   | Trophic transfer of organochlorines in marine food webs . . . . .            | 31 |
| 4.3.   | Trophic transfer of “new” and chiral chemicals in marine food webs . . . . . | 32 |
| 4.4.   | Trophic transfer of organochlorine metabolites in marine food webs . . . . . | 33 |
| 5.     | Temporal trends of contaminants in biota . . . . .                           | 34 |
| 5.1.   | Temporal trends of mercury in invertebrates . . . . .                        | 35 |
| 5.2.   | Temporal trends of contaminants in seabirds . . . . .                        | 35 |
| 5.2.1. | Temporal trends of organochlorines in seabird eggs. . . . .                  | 35 |
| 5.2.2. | Temporal trends of new chemicals in seabird eggs . . . . .                   | 36 |
| 5.2.3. | Temporal trends of mercury in seabird eggs. . . . .                          | 36 |
| 5.3.   | Temporal trends of contaminants in marine mammals . . . . .                  | 36 |
| 5.3.1. | Temporal trends of organochlorines . . . . .                                 | 36 |
| 5.3.2. | Temporal trends of new chemicals . . . . .                                   | 43 |
| 5.3.3. | Temporal trends of mercury . . . . .   | 45 |
| 6.     | Summary and conclusions. . . . .   | 48 |
| 6.1.   | Spatial trends of legacy organochlorines and mercury . . . . .               | 48 |
| 6.2.   | Temporal trends of legacy organochlorines and mercury. . . . .               | 49 |
| 6.3.   | New contaminants in the Arctic . . . . .                                     | 50 |
|        | Acknowledgements . . . . .   | 50 |
|        | References . . . . .   | 51 |

## 1. Introduction

The environmental and human health implications of persistent organic pollutants (POPs) and mercury (Hg; together referred to here as “contaminants”) in the Canadian Arctic have been of continuing public and scientific concern since the mid-1980s. The status of contaminants in Canadian Arctic marine ecosystems was previously reviewed by Muir et al. (1992b, 1999b) using information available up until 1996. The present paper updates and expands upon these earlier reviews, specifically with respect to spatial and temporal trends in marine mammals, seabirds, fish and invertebrates, by drawing largely on recent outputs of the Biotic Monitoring subprogram of Phase II of Canada’s Northern Contaminants Program (1997–2001). Published and unpublished results have also been included from other contaminant studies conducted in the Canadian Arctic during the period 1997–2002. The Biotic Monitoring subprogram was designed with a focus on “priority contaminants” identified under the Persistent Organic Pollutant and Heavy Metal Protocols of the United Nations Economic Commission for Europe Convention on Long-Range Transboundary Air Pollution that were under

negotiation in 1997. The program was also designed to conduct preliminary studies on other contaminants not currently identified in the protocols but having physical/chemical properties which gave reason for concern that they may pose a risk to Northerners (e.g., persistent, toxic, bioaccumulative, and prone to long-range transport and deposition).

The first Canadian Arctic Contaminants Assessment Report (CACAR) by Muir et al. (1997) found that the geographic coverage of contaminant data on marine mammal and seabird populations was very good. All major beluga, ringed seal and polar bear stocks along with several major seabird colonies had been sampled for organochlorine and heavy metal contaminants during the period 1990–1995. However, a number of knowledge gaps for the marine biological environment were identified, and the Biotic Monitoring subprogram was designed to address them. These knowledge gaps included:

1. *Temporal trends in biota*: The lack of temporal trend information for most contaminants was identified as the most significant knowledge gap. Temporal trend data were very limited for most organochlorines and metals because they were

based on 2 or at most 3 sampling times and some comparisons were limited because of changes in methodology. Temporal trend data for mercury was judged as being particularly important given the evidence of increasing concentrations in the tissues of marine mammals.

2. *Marine fish and food webs*: There was sparse information on contaminant levels and very limited geographic coverage for marine fish and invertebrates. This gap was significant because of the importance of understanding the pathways of bioaccumulation of contaminants.
3. *Lack of modelling of chemical fate and bioaccumulation*: The lack of integration of chemical measurement data with well established models of chemical fate and food chain bioaccumulation was identified as a major knowledge gap in the program.

In this paper, we update data on spatial patterns and temporal trends of POPs, including “legacy” organochlorines (OCs) as well as some of the “newer” non-chlorinated compounds, and Hg in Arctic marine invertebrates, fish, mammals and seabirds in Canada. We also present recent information on the transfer of POPs and Hg in marine food webs. New data for large scale (Alaska to Svalbard) trends of metals in seals (Riget et al., 2005) and POPs in polar bears (Verreault et al., 2005), as well as progress on the modelling of chemical fate and bioaccumulation in marine mammals (Hickie et al., 2005), are discussed in other papers in this issue.

## 2. Pathways and processes of delivery of contaminants to marine ecosystems

### 2.1. Transport of air-borne and sea-borne contaminants

Pathways of transport of the OCs, heavy metals, radionuclides, and hydrocarbon contaminants to the Canadian Arctic marine environment include transport in the troposphere in gas phase and on particles, as well as via ocean currents (Macdonald et al., 2000, 2003). Air-borne contaminants are removed from the atmosphere by gas absorption, precipitation and dry deposition. Many chlorinated organics are present as

gases even at low temperatures and are absorbed from the gas phase by water, snow, soil and plant surfaces. Precipitation scavenging of gas and particles from the air also deposits particle-associated OCs and metals in snow and rain. Dry deposition is a third pathway of input of aerosol-bound contaminants to marine ecosystems. For the more water-soluble, less volatile contaminants, however, transport via ocean currents may be more important than the air-borne route (Li et al., 2002, 2004). Waterborne contaminants also enter Canadian Arctic marine ecosystems from northward flowing rivers such as the Athabasca/Peace/Slave River system which feeds into Great Slave Lake which, in turn, feeds into the Mackenzie River, the Nelson River/Lake Winnipeg drainage, and other major rivers flowing into Hudson Bay.

### 2.2. Bioaccumulation and biomagnification processes

Hydrophobic organics and heavy metals such as Hg are readily adsorbed by living and dead organic matter such as particulate organic carbon, waxy plant surfaces, animal membranes and lipids. Once adsorbed, the bioavailability of these chemicals to marine animals will depend on the properties of the chemical and on the physical, chemical and biological environments into which it is released. Persistent OCs accumulate in organisms due to a high affinity for lipids and, most importantly, high biological inertness of the parent chemical or metabolites. For metals, differences in uptake due to speciation of the element (which may be influenced by water hardness, salinity, redox conditions in sediment, pH and temperature), as well as metabolic rate of the organism, affect transfer across biological membranes (Heath, 1987).

Dietary accumulation is often the dominant route of exposure for organic contaminants and methylmercury (MeHg) for aquatic organisms (Borgå et al., 2004). This varies somewhat with the hydrophobicity of the chemical, with dietary accumulation making up a greater percentage of the exposure with increasing hydrophobicity. The increased dietary accumulation is due to relatively low concentrations in water (typically in the low pg/L range in the Arctic), high bioconcentration at the first trophic level, and low rates of elimination of these chemicals by organisms. Therefore, concentrations in food tend to be higher than

what is found in water, with the difference becoming greater with each step up the food web. Transfer within the food web through food ingestion is the dominant pathway for uptake of POPs and Hg in larger marine organisms. This pathway, coupled with the slow rate of excretion and metabolism, leads to biomagnification, or the increase in concentration of a chemical from prey to predator. The food requirements of an organism are controlled by metabolic rate and production (growth, lipid deposition, reproduction). Thus metabolic rate is linked to the rate of uptake of contaminants. Homeotherms (e.g., mammals, birds) have greater energy requirements per unit body weight than poikilotherms (e.g., fish) because of the requirement to maintain body temperature. Therefore, the metabolic rate and caloric requirements of homeotherms are higher. Consequently, the largest biomagnification factors (the concentration of contaminants in predator divided by concentration in prey) are usually observed between fish and marine mammals or sea birds for higher chlorinated PCBs, OC pesticides and Hg, in Arctic food webs (Muir et al., 1992b; Fisk et al., 2001a).

### 3. Spatial patterns of contaminants in marine biota

The presence of OCs has historically received more attention than metals in the marine environment of the Canadian Arctic. In fact, the marine environment was the first Arctic system to be examined for the presence of OC contaminants (Holden, 1970). Since the previous review (Muir et al., 1999b) which covered results to 1996, a large amount of OC data has been produced for the marine environment. A number of studies have been initiated on levels and dynamics of OCs and Hg in seals and whales, and a number of comprehensive studies on the dynamics and mechanisms of OCs and Hg in the marine food web have been completed.

Studies on Hg in the marine environment have focused predominantly on marine mammals. Mercury is a naturally occurring element whose level in the global environment has been augmented by human activities. This element is of particular concern because there are indications that levels have been increasing in the Arctic marine environment over the past few decades (see Section 5).

#### 3.1. Contaminants in invertebrates and marine fish

Marine invertebrates provide a link between phytoplankton and fish, seabirds and mammals in Arctic marine food webs. These invertebrates are, therefore, important in the transfer of carbon and nutrients as well as OCs and mercury to upper trophic level organisms. Marine and anadromous fish occupy a range of trophic positions in Arctic marine ecosystems and hence concentrations of contaminants in their tissues can be quite variable. The Arctic cod (*Boreogadus saida*) is a key link in marine food webs, connecting invertebrates with seabirds and marine mammals, and as a major dietary item of many important Arctic marine mammals, including the ringed seal (*Phoca hispida*) and beluga (*Delphinapterus leucas*). A knowledge of the trends and dynamics of OCs and metals in marine invertebrates and fish is important for the understanding of trends and patterns of contaminants overall in Arctic marine ecosystems. A number of marine fish species, e.g., Arctic char (*Salvelinus alpinus*), are important components of the traditional human diet, and some have become important commercial species, e.g., Greenland halibut or turbot (*Reinhardtius hippoglossoides*), but few Arctic invertebrates are consumed by humans. Historically there have been few studies of OCs and metals in marine invertebrates and fish, at least compared with marine mammals and seabirds. Recently, however, there have been a number of studies which examined contaminants in these organisms.

##### 3.1.1. Mercury in invertebrates

Low concentrations of Hg were found in muscle (meat) samples of scallops (*Placopecten magellanicus*) from Labrador (Muir et al., 1999c, 2000b). All of the Hg was in organic form, probably MeHg. Mercury levels in blue mussels (*Mytilus edulis*) sampled from seven communities along the Labrador coast, and Ungava Bay and Hudson Strait area of Nunavik, were low ranging from 0.01 to 0.03 µg/g wet weight (ww) and did not vary among locations (Muir et al., 1999c, 2000b). Organic Hg averaged 54% of total Hg. This organic Hg was probably all in the MeHg form (Wagemann et al., 1997). Doidge et al. (1993) found similar, low levels of Hg (0.03 µg/g ww) in blue mussels from six communities in the Hudson Bay, Hudson Strait and Ungava Bay area.



As part of a food web study (see Section 4), Atwell et al. (1998) measured total Hg in 12 invertebrates and particulate organic matter (POM) from the Lancaster Sound region. Total Hg was not detected in POM ( $<0.02 \mu\text{g/g}$  dry weight) and ranged from 0.03 to  $0.18 \mu\text{g/g}$  dw in the invertebrates. These levels are similar to those observed in blue mussels from Nunavik and Labrador (Muir et al., 1999c, 2000b). There appeared to be no relationship between Hg concentration and trophic level within the invertebrates as a single group. The authors reported no evidence of bioaccumulation of Hg with age in clams (*Mya truncata*), despite the age range covering 42 years.

Levels of total Hg and MeHg were recently determined in ice algae and four species of zooplankton collected between May and July 1998 in northern Baffin Bay (Campbell et al., this issue). Zooplankton samples included mixed samples (predominantly *Calanus* copepods), *Calanus hyperboreus*, *Themisto libellula* and *Mysis oculata*. Total Hg in ice algae was low ( $0.003 \mu\text{g/g}$  ww,  $n=1$ ) and MeHg was not detected. Total Hg concentrations ranged from  $0.006 \pm 0.002 \mu\text{g/g}$  ww (mean  $\pm$  S.D.) in mixed zooplankton to  $0.025 \pm 0.017 \mu\text{g/g}$  ww in *C. hyperbor-*

*eus*. Methylmercury varied widely among species, with the relative percentage of total Hg as MeHg being  $3.3 \pm 2.9$  in *C. hyperboreus*,  $48.6 \pm 27.3$  in mixed zooplankton, 66.9 to 87.6 in *T. libellula*, and  $88.0 \pm 9.5$  in *M. oculata*. Mercury levels in the zooplankton were not related to trophic position as a group but in the larger continuum of the food web, which included ice algae, fish, seabirds and ringed seals, Hg was found to biomagnify (see Section 4).

### 3.1.2. Mercury in marine and anadromous fish

An overview of Hg in sea-run Arctic char is provided in Fig. 1, which shows the ranges of concentrations for 55 locations. In general, concentrations of Hg ranged from 0.02 to  $0.08 \mu\text{g/g}$  ww in muscle at all these locations (Lockhart et al., 2005a). No distinctive geographical trends were apparent for Hg in those char. The site-to-site variation in Hg was probably due to age and trophic level of the char. Muir et al. (2000b) found that mean Hg levels in sea-run char from the eight communities in Labrador and the Ungava/Hudson Strait region of Nunavik ranged from 0.032 to  $0.040 \mu\text{g/g}$  ww (Fig. 1). Statistical analysis of the results showed that there were no

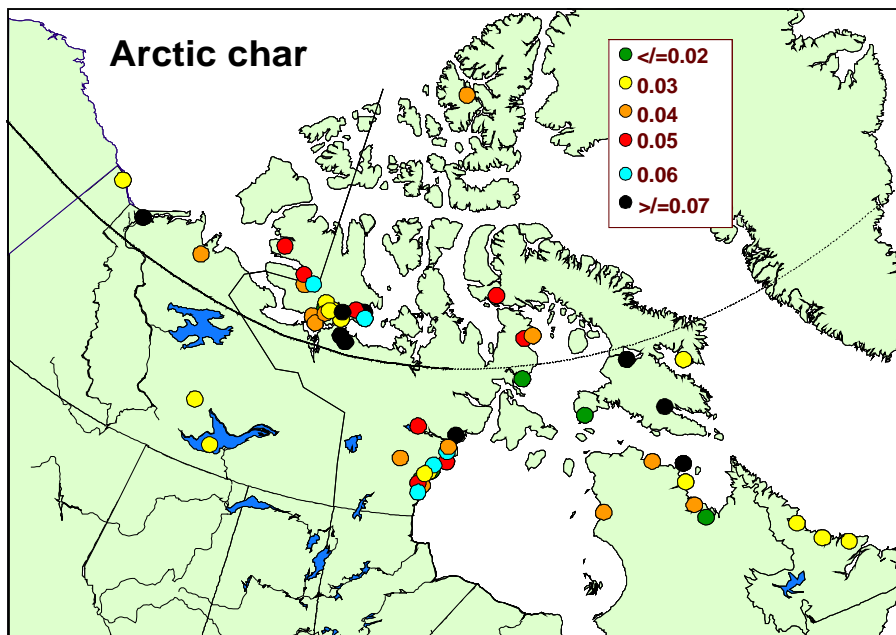


Fig. 1. Range of concentration of mercury in sea-run Arctic char from 55 locations in the Canadian Arctic (adapted from Lockhart et al., 2005a).

significant differences in Hg levels among the three Labrador sites; however, one location in Nunavik (Quaqtaq) had higher levels than all other sites (Fig. 1). The Hg levels were slightly lower than reported by Riget et al. (2000) in sea-run char from southwest Greenland.

As part of a food web study (Section 4), Atwell et al. (1998) measured total Hg in muscle tissue of two fish species, Arctic cod and twohorn sculpin (*Icelus bicornis*) from the Lancaster Sound region. Campbell et al. (this issue) also measured total Hg in Arctic cod from northern Baffin Bay. Concentrations of Hg in the Arctic cod ( $0.19 \pm 0.3 \mu\text{g/g dw}$ , mean  $\pm 1$  S.E.) and twohorn sculpin ( $0.19 \pm 0.3 \mu\text{g/g dw}$ , mean  $\pm 1$  S.E.) were similar to those observed in Arctic char if concentrations were converted to wet weight. Campbell et al. (this issue) reported total Hg concentrations in whole Arctic cod of  $0.04 \mu\text{g/g ww}$  and in liver of  $0.013\text{--}0.016 \mu\text{g/g ww}$ . The Hg concentrations in cod from northern Baffin Bay and Lancaster Sound were similar on a dry weight basis.

Recently, mercury data have been produced for livers of Greenland shark (*Somniosus microcephalus*), the only shark species that routinely inhabits Arctic waters (Fisk, University of Georgia, unpublished data). Little is known about these large fish but evidence suggests that they are long-lived, generalist feeders with a potential to feed at a high trophic level in the food web (Fisk et al., 2002b). Greenland shark samples ( $n=24$ ) were collected in Cumberland Sound in 1999 and 2000. Mercury concentrations in livers of the sharks ( $0.49 \pm 0.06 \mu\text{g/g, ww}$ ) varied little between sexes and were not related to length (linear regression,  $p>0.20$ ). Concentrations of total Hg in Greenland shark liver ( $0.49 \pm 0.06 \mu\text{g/g}$ ) were more than an order of magnitude greater than sea-run Arctic char muscle collected in the same region but an order of magnitude less than levels observed in marine mammal liver.

### 3.1.3. Organochlorines in invertebrates

A number of studies have been recently carried out to examine OC concentrations in Arctic marine invertebrates and the factors that influence OC concentrations in these lower trophic level organisms (Fisk et al., 2001b, 2003b; Hoekstra et al., 2002; Kuzyk et al., 2005). The most common OC compounds in invertebrates are the more water-soluble compounds, such as

HCH isomers and lower chlorinated PCB congeners (Fisk et al., 2001b, 2003b; Hoekstra et al., 2002). This reflects the smaller size, lower trophic level, and lack of biotransformation capacity generally found in invertebrates as compared to fish, mammals and birds. Water is an important exposure route to OCs for zooplankton but recent evidence suggests that diet may also play a role (Hoekstra et al., 2002; Borgå et al., 2004). Scavenging invertebrates, for example the benthic amphipod, *Anonyx nugax*, and invertebrates that are found near contaminated sites have a different pattern of OCs (Fisk et al., 2003b; Kuzyk et al., 2005).

Calanoid copepod samples were collected from Holman, N.W.T., and Barrow and Kaktovik, Alaska, in 1999–2000 to examine spatial trends of OCs in the Beaufort Sea region (Hoekstra et al., 2002). Calanoid copepods are dominant components, in both number and biomass, of high-latitude marine zooplankton communities and play an important role in polar food webs, as their high lipid reservoirs and biomass provide higher trophic level consumers with a high-energy diet (Springer et al., 1996). The ranking of OC group concentrations from highest to lowest were toxaphene  $\geq \sum\text{PCB} > \sum\text{HCH} > \sum\text{DDT} > \sum\text{CHL} > \sum\text{CBz}$ . The higher abundance of toxaphene congeners relative to other OCs in Beaufort Sea zooplankton was consistent with previous data from the Arctic Ocean near Axel Heiberg Island (Bidleman et al., 1989). The ranking by OC concentration was the same as that observed in samples of *C. hyperboreus*, a major component of the Beaufort Sea copepods, collected ( $n=20$ ) between April and July 1998 in northern Baffin Bay (Fisk et al., 2001b). The  $\alpha$ - and  $\gamma$ -HCH isomers and lower chlorinated PCB congeners were the most common OCs found in the calanoid copepods. The relative abundance of the OC groups and individual chemicals varied throughout the 4-month season in northern Baffin Bay *C. hyperboreus*, as did lipid content (Fisk et al., 2001b). Concentrations of  $\sum\text{HCH}$ ,  $\sum\text{CHL}$  and  $\sum\text{CBz}$  increased over the sampling period, but no change in  $\sum\text{PCB}$  or  $\sum\text{DDT}$  was observed.

No significant differences were found for any  $\sum\text{OC}$  groups in calanoid copepods among the western Arctic sites (Holman, Kaktovik, and Barrow) when the effects of lipid and water content covariates were removed ( $p>0.05$  for all comparisons) (Hoekstra et al., 2002). Significant differences were observed, however, between the Alaskan sites and

northern Baffin Bay/Rankin Inlet sites for toxaphene,  $\sum$ PCB,  $\sum$ DDT,  $\sum$ HCH and  $\sum$ CHL (Fig. 2). Hexachlorobenzene (HCB) and HCH isomers in zooplankton and water samples from the western Canadian Arctic were relatively higher than in more easterly locations. The relative abundance of HCB and HCH in the western Arctic probably reflects long-range atmospheric transport of the chemicals and geographic proximity to areas of recent application in Asia (Li, 1999; Bailey, 2001). In contrast, total toxaphene in zooplankton was lower at Alaskan sites compared to the eastern Canadian Arctic. The concentrations of OC compounds in zooplankton, including *C. hyperboreus* collected in the late 1980s, were in general agreement with this study (Bidleman et al., 1989), suggesting that OC concentrations in western Arctic water and zooplankton have remained constant over the past decade.

Organochlorine data for a number of other pelagic and benthic zooplankton have recently been determined in northern Baffin Bay (Fig. 3). Lipid-corrected

OC concentrations varied among species and appear to be related, in part, to a combination of trophic position (as determined by  $\delta^{15}\text{N}$ ) and body size. The relative ranking of OC groups varied with the species but, in general,  $\sum$ PCB had the highest concentrations and  $\sum$ CBz had the lowest concentrations. Benthic invertebrates have a larger range of sizes, feeding ecology and ecological niches than pelagic invertebrates and, therefore, had a greater range of OC concentrations (Fisk et al., 2003b). Relatively high levels of PCBs and high proportions of highly chlorinated PCB congeners were found in a small number of the pelecypod samples (*M. edulis* and *M. truncata*) (Fig. 3), suggesting that local harbors and communities might be point sources of PCBs. Scavenging benthic invertebrates may also have relatively high OC concentrations (Hargrave et al., 1992, 2000; Fisk et al., 2003b). The highest concentrations of OCs in Arctic invertebrates were found in the scavenging amphipod, *A. nugax*, and were similar to those in Arctic cod and the dovekie (*Alle alle*) (Fisk et al., 2001a). Organo-

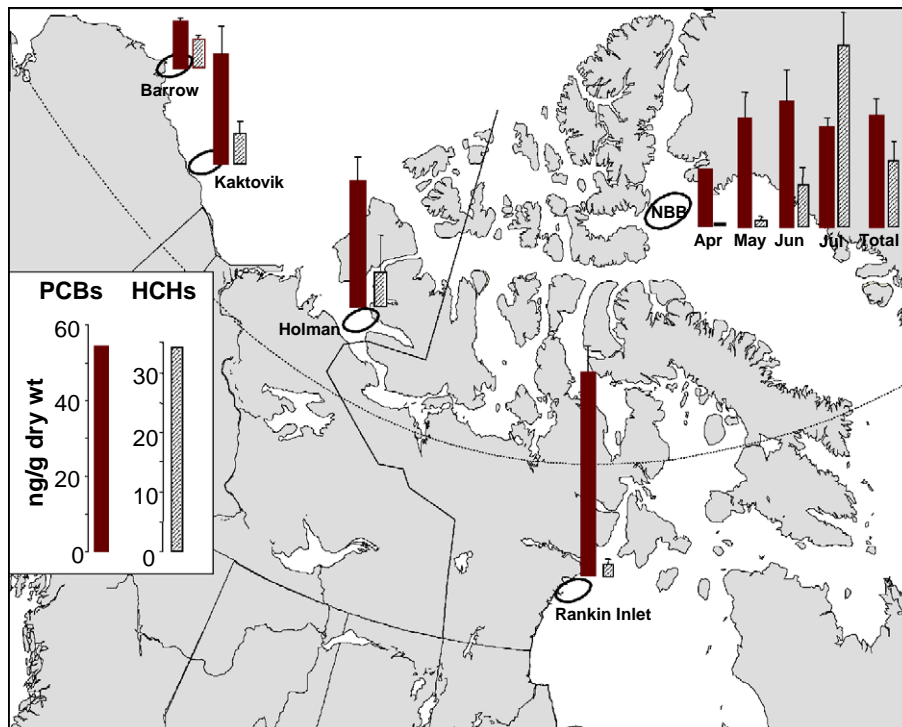


Fig. 2. Mean  $\sum$ PCB and  $\sum$ HCH ( $\pm 1$  S.E.) in zooplankton (ng/g dw) in Barrow and Kaktovik, Alaska; Holman, NWT; Rankin Inlet, Nunavut; and North Baffin Bay (NBB). Data from Hoekstra et al. (2002), Fisk et al. (2001b) and Muir and Lockhart (1997).



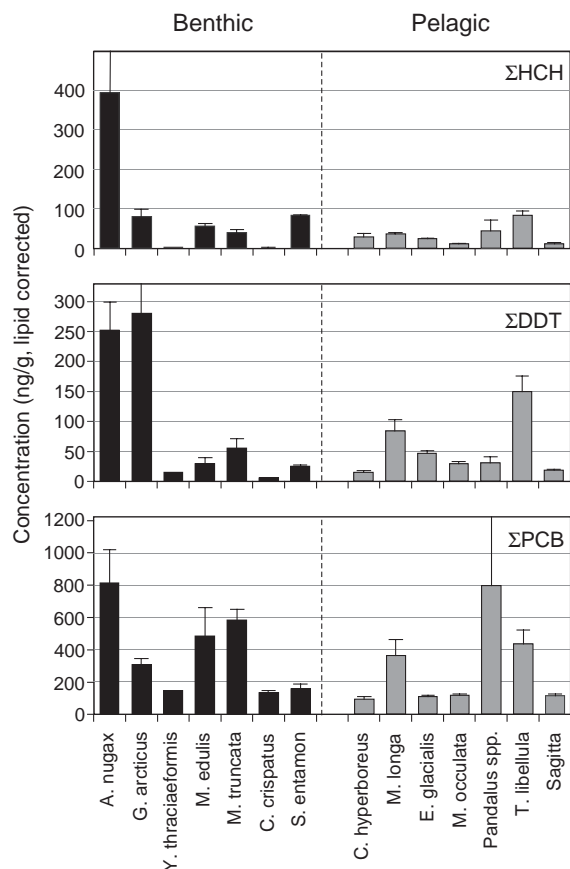


Fig. 3. Concentrations (ng/g, lipid normalized) of  $\Sigma$ HCH,  $\Sigma$ DDT and  $\Sigma$ PCB in Arctic benthic and pelagic invertebrates. All samples were collected in 1998 in northern Baffin Bay except *M. truncata* (1993 in Iqaluit harbor), *M. edulis* (1998 at seven locations in northern Québec) and *S. entamon* (1998 in northern Alaska near Barrow). Results of *A. nugax* and *G. arcticus* include samples from Cumberland Sound. All data from Fisk et al. (2003b).

chlorine concentrations in filter- and detritus-feeding invertebrates were among the lowest found in any biota in the world.

Concentrations of PCBs and other OCs were recently measured in a number of benthic invertebrates from Saglek Bay, Labrador (Kuzyk et al., 2005). Saglek Bay is the site of a former military radar operation and is a known point source of PCBs. Concentrations of PCBs were very high in sea urchins, clams and mussels within 1.5 km of the point sources and decreased exponentially with distance away from the site. Levels of other OCs, DDT

and chlordanes were in the range reported for other Arctic benthic invertebrates (Fisk et al., 2003b). These results further demonstrate the influence of local contamination on invertebrate OC levels but suggest that the radius of impact is limited to small distances (<10 km).

#### 3.1.4. Organochlorines in marine fish

Low (ng/g ww) levels of OCs were detected in Arctic char muscle (with skin) from two locations in Labrador and three locations in Nunavik (Table 1). PCBs were the most prominent OCs, with averages ranging from 10 ng/g ww at Kangiqsujuaq to 31 ng/g ww at Nain. No significant differences in  $\Sigma$ PCB among locations were found. Lipid content, length and age were not significant co-variates. However,  $\Sigma$ PCB levels were influenced by the sex of the fish, with males having significantly higher levels than females. This may have been due to the timing of sampling which was near spawning time for the char. Lower concentrations may have been observed in the muscle of females due to contaminants being mobilized along with fat for deposition in the eggs.

The next most prominent groups of OCs in char muscle were the HCH and DDT groups which were both present at low ng/g levels (Table 1).  $\Sigma$ DDT levels were higher at Nain than other locations but this difference was marginally significant only between Nain and Kangiqsujuaq. The char from Kangiqsujuaq were landlocked. In general, these levels of persistent OCs are similar or lower than reported in char muscle from other locations in the Canadian Arctic (Muir et al., 1997).

Arctic or polar cod are a key link in Arctic marine foodwebs, providing a link between invertebrates and marine mammals and seabirds.  $\Sigma$ PCB was the dominant OC group followed by the chlordanes ( $\Sigma$ CHL) in whole Arctic cod collected in northern Baffin Bay in 1998 (Table 1). Levels were similar to those observed in sea-run Arctic char. Hoekstra et al. (National Water Research Institute, unpublished data) reported similar concentrations of OCs in Greenland cod (*Gadus ogac*) collected in 1999 near Holman Island, N.W.T.

Concentrations of PCBs and other OCs were recently measured in livers and whole bodies of short-horn sculpin from Saglek Bay, Labrador, and a reference site in the region (Kuzyk et al., 2005). As with the invertebrates, PCBs were very high in sculpin

Table 1  
Mean concentrations of organochlorines (ng/g ww) in marine and anadromous fish from the Canadian Arctic

| Species   | Region           | Location     | Tissue            | Year | % Lipid | <i>n</i> | ∑CBz | ∑HCH | ∑CHL | ∑DDT | ∑PCB | Ref <sup>a</sup> |
|---|------------------|--------------|-------------------|------|---------|----------|------|------|------|------|------|------------------|
| <i>Marine fish</i>  |                  |              |                   |      |         |          |      |      |      |      |      |                  |
| Greenland cod<br>( <i>Gadus ogac</i> )                              | Beaufort Sea     | Holman, NT   | whole body        | 1999 | 2.6     | 10       | 1.11 | 2.40 | 2.44 | 0.71 | 0.89 | 1                |
| Arctic cod<br>( <i>Boreogadus saida</i> )                           | Baffin Bay       |              | whole body        | 1998 | 1.2     | 8        | 1.16 | 0.93 | 3.13 | 2.61 | 3.73 | 2                |
| Turbot/Greenland halibut<br>( <i>Reinhardtius hippoglossoides</i> ) | Cumberland Sound | Pangnirtung  | muscle, with skin | 1999 | 18.6    | 4        | 10.2 | 14.8 | 31.1 | 15.0 | 11.1 | 3                |
| Greenland shark<br>( <i>Somniosus microcephalus</i> )               | Cumberland Sound | Pangnirtung  | liver             | 1999 | 39.7    | 15       | 192  | 29.1 | 1050 | 4158 | 2000 | 3                |
|   | Davis Strait     |              | liver             | 1997 | 55.9    | 2        | 53.5 | 27.1 | 262  | 817  | 590  | 3                |
| <i>Anadromous fish</i>  |                  |              |                   |      |         |          |      |      |      |      |      |                  |
| Arctic char<br>( <i>Salvelinus alpinus</i> )                        | Eastern Canada   | Kangirsuk    | muscle, with skin | 1998 | 8.5     | 10       | 0.90 | 1.14 | 1.19 | 1.66 | 17.6 | 4                |
|   |                  | Makkovik     | muscle, with skin | 1998 | 5.1     | 9        | 0.94 | 2.38 | 1.40 | 1.75 | 10.6 | 4                |
|   |                  | Nain         | muscle, with skin | 1998 | 6.8     | 11       | 1.37 | 3.14 | 1.84 | 2.50 | 30.8 | 4                |
|   |                  | Quaqtaq      | muscle, with skin | 1998 | 6.4     | 7        | 1.14 | 1.29 | 1.30 | 1.44 | 18.2 | 4                |
|   |                  | Kangiqsujuaq | muscle, with skin | 1998 | 3.9     | 10       | 0.84 | 0.48 | 1.26 | 1.18 | 10.3 | 4                |

<sup>a</sup> References: 1. Hoekstra et al. (National Water Research Institute, unpublished data); 2. Fisk et al. (2002a); 3. Fisk et al. (2002b); 4. Muir et al. (2000b).

collected within 1.5 km of the point source but declined rapidly away from the source. Levels of PCBs in whole sculpin from the reference site (Okak) and at sites more than 7.5 km away from the point sources were similar to those in cod from northern Baffin Bay (Fisk et al., 2003b) and Holman Island (Hoekstra et al., National Water Research Institute, unpublished data).

Organochlorines were measured in 17 Greenland sharks and four turbot collected in the Davis Strait and Cumberland Sound region in 1997 and 1999 (Table 1) (Fisk et al., 2002b). Concentrations in the Greenland shark were in the range of other top Arctic marine predators, such as polar bears (*Ursus maritimus*) or glaucous gulls (*Larus hyperboreus*). Concentrations (lipid basis) of OCs in the Greenland sharks were not related to fork length, sex,  $\delta^{13}\text{C}$  or  $\delta^{15}\text{N}$  values, but were 3–10 times higher than those in ringed seals, suggesting a higher trophic level for

the sharks than implied by  $\delta^{15}\text{N}$  values. Concentrations of  $\sum\text{DDT}$  in the sharks were among the highest in Canadian Arctic marine biota, which may be related to the low metabolism, high trophic level and long life span of these sharks. Concentrations of OCs were 10–100 times lower than the Greenland shark but slightly higher than levels observed in anadromous char. Reported concentrations of  $\sum\text{PCB}$  and  $\sum\text{DDT}$  in turbot collected in the Davis Strait in 1992 were 5 and 10 times higher, respectively, but concentrations of  $\sum\text{CHL}$  and  $\sum\text{HCH}$  were very similar to those reported for turbot in this study (see Berg et al., 1997).

### 3.2. Levels and spatial trends of contaminants in seabirds

There are approximately 50 species of Arctic seabirds (de March et al., 1998) that have a variety of

feeding and migration strategies, and consequently OC and Hg concentrations can vary considerably among species (Fisk et al., 2001c; Braune et al., 2002; Braune and Simon, 2003). Species, such as the glaucous gull, that migrate to more contaminated regions and/or that scavenge, particularly on dead marine mammals, have the highest OC and Hg concentrations (Braune et al., 2002; Buckman et al., 2004).

There has been a considerable amount of work on OCs and Hg in Arctic seabirds since the first CACAR assessment. Continued monitoring of OCs in Canadian Arctic seabird eggs, from 1975 to 1998 and beyond, has proved a strong data set for examining temporal trends among different species (see Section 5). Recently, OC and Hg data have also been generated for a range of species from northern Baffin Bay, and new data on polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), toxaphene and new OCs have also been generated for Canadian seabirds (Braune, 2003; Braune and Simon, 2003, 2004).

### 3.2.1. Mercury in seabirds

**3.2.1.1. Seabirds of northern Baffin Bay.** A recent study measured total Hg and MeHg in the muscle and total Hg in the liver of eight species of Arctic seabirds collected in the High Arctic between Ellesmere Island and Greenland in 1998 and 1999 to examine inter-species differences in seabirds (Campbell et al., this issue; Fisk, University of Georgia, unpublished data). Liver concentrations of Hg were greater than in muscle across all species. Concentrations of total Hg varied between gender when liver and muscle samples were combined but was marginally different when only muscle data was compared. Methylmercury concentrations did not vary between sexes for any of the seabird species and were greater than 75% of the total Hg for all seabird species examined.

An initial examination of Hg concentrations in the seabird species indicated that age, migration and trophic level are important parameters to consider when interpreting seabird Hg concentrations. Dovekies had significantly lower Hg concentrations than all other seabirds (Fig. 4), probably because of their comparatively low trophic position. Mercury concentrations were highest in glaucous gull, and

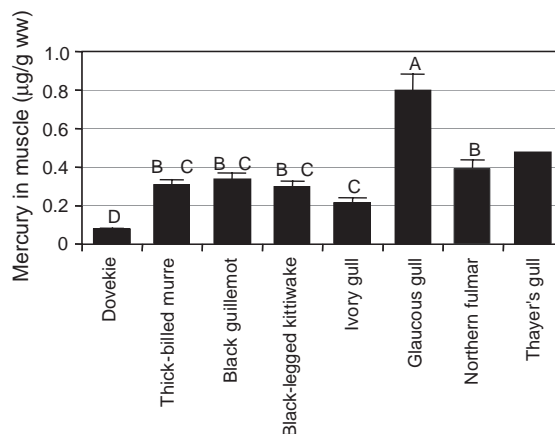


Fig. 4. Total mercury concentrations (bars are mean  $\pm$  S.E., outliers removed) in the muscle of seabirds collected in northern Baffin Bay in May or June 1998. Concentrations that were not statistically different ( $p > 0.05$ ) have the same letter and were tested using a general linear model (GLM) and log-transformed data. Statistics did not include Thayer's gull because  $n = 1$  for this seabird.

were highly correlated across all species to trophic position (see Section 4.1.2).

**3.2.1.2. Canadian Arctic seabird eggs.** As part of a continuing temporal study of contaminants in seabirds, eggs of northern fulmars (*Fulmarus glacialis*), glaucous gulls, black-legged kittiwakes (*Rissa tridactyla*), thick-billed murres (*Uria lomvia*) and black guillemots (*Cepphus grylle*) were collected from nests on Prince Leopold Island in Lancaster Sound, Nunavut, in 1998 and analyzed for Hg (Braune et al., 2001; Braune, Canadian Wildlife Service, unpublished data). Concentrations of total Hg ( $3.4 \pm 0.3$   $\mu\text{g/g dw}$ ) in glaucous gull eggs were significantly higher than concentrations measured in all other species which may relate to the gull's higher trophic level. Kittiwake eggs contained significantly lower total Hg concentrations ( $0.6 \pm 0.1$   $\mu\text{g/g dw}$ ) compared to all other species. Total Hg was not statistically distinct between thick-billed murres and northern fulmars. Mercury has a tendency to accumulate at higher latitudes (Barrie et al., 1997); therefore, those species spending more time at higher latitudes may be expected to exhibit higher concentrations. This was reflected by data on black guillemots, which are most likely to remain resident in the Arctic, and had higher Hg concentrations. Black-

legged kittiwakes migrate south along the eastern seaboard as far south as Florida and thus exhibited the lowest Hg concentrations.

### 3.2.2. Organochlorines in seabirds

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

**3.2.2.1. Seabirds of northern Baffin Bay.** Organochlorine concentrations and dynamics were measured in the liver and fat of seven Arctic seabird species collected in northern Baffin Bay in 1998 (Fisk et al., 2001c; Buckman et al., 2004).  $\Sigma$ PCB was the most predominant OC group observed in all seabird species and, along with most of the other OC groups, attained the highest concentrations in the gulls (glaucous and ivory gulls, black-legged kittiwakes) and northern fulmars (Fig. 5). Glaucous gulls, ivory gulls (*Pagophila eburnea*) and northern fulmars scavenge, and glaucous gulls prey on other seabirds. These foraging habits account for the high OC concentrations. Migration to more contaminated southern locations may also affect OC levels in the glaucous gull and black-legged kittiwake. Black-legged kittiwakes, a non-scavenging species, had higher concentrations than the alcids (black guillemot, thick-billed murre, dovekie), which is likely due to their migration to southern habitats. The kittiwakes were found to be feeding at a lower trophic level than the black guillemot in northern Baffin Bay (Hobson et al., 2002), and since the black guillemot does not undergo a major migration, the difference between the species highlights the impact of exposure to OCs by migrating seabirds in non-Arctic habitats.  $\Sigma$ PCB had the highest concentrations among OCs, followed by  $\Sigma$ DDT and

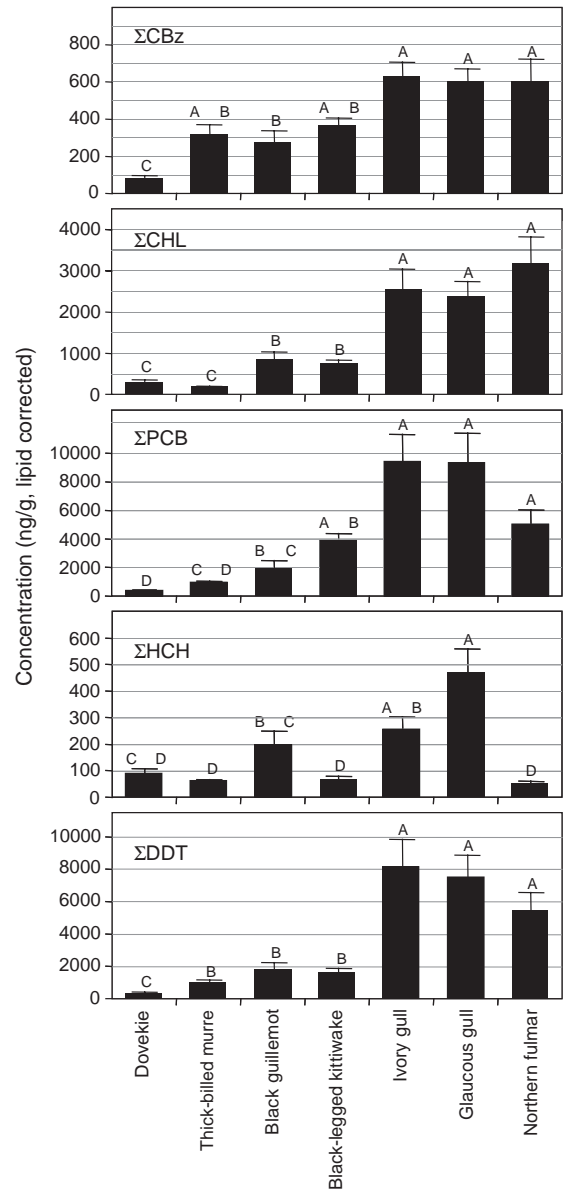


Fig. 5. Lipid-corrected concentrations (mean  $\pm$  S.E., ng/g) of  $\Sigma$ CBz,  $\Sigma$ HCH,  $\Sigma$ CHL,  $\Sigma$ DDT and  $\Sigma$ PCB in liver tissue of North Water (NOW) Polynya seabirds (Buckman et al., 2004). Males and females did not significantly differ ( $p < 0.05$ ), except for  $\Sigma$ DDT, and were combined. Bars with the same letter do not differ significantly ( $p > 0.05$ ).

$\Sigma$ CHL, which were similar, chlorobenzenes ( $\Sigma$ CBz) and, finally,  $\Sigma$ HCH. The exception to this ranking is thick-billed murres in which  $\Sigma$ CHL was found at much lower concentrations than in other seabirds.

Thick-billed murres exhibit an exceptional ability to biotransform chlordane-related compounds (Fisk et al., 2001c).

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

**3.2.2.2. Seabirds of Lancaster Sound.** As part of a continuing temporal study of contaminants in seabird eggs (see Section 5), thick-billed murre, northern fulmar and black-legged kittiwake eggs were collected on Prince Leopold Island in Lancaster Sound in 1998 and analyzed for OCs (Braune et al., 2001). Concentrations and the relative proportions of OC groups were in general agreement with those observed in tissue of adult birds of the same species collected in northern Baffin Bay (Fisk et al., 2001c; Buckman et al., 2004). Lipid content in the eggs of the three species was similar, allowing comparisons of wet weight concentration data.  $\sum$ PCB was the predominant OC in all three species. PCB concentrations were similar in black-legged kittiwakes and northern fulmars ( $0.28 \pm 0.02$  and  $0.27 \pm 0.02$   $\mu\text{g/g}$  ww, respectively), which were nearly double those in thick-billed murres ( $0.13 \pm 0.01$   $\mu\text{g/g}$  ww). This may reflect migration to more contaminated habitats by kittiwakes and fulmars.  $\sum$ DDT was highest in the northern fulmar ( $0.21 \pm 0.02$   $\mu\text{g/g}$  ww), followed by the thick-billed murre ( $0.10 \pm 0.01$   $\mu\text{g/g}$  ww) and the black-legged kittiwake ( $0.06 \pm 0.01$   $\mu\text{g/g}$  ww), and similar trends were observed for  $\sum$ CHL.

Recently, compounds such as PCDDs, PCDFs, non-ortho PCBs and toxaphene were analyzed in thick-billed murres, northern fulmars and black-legged kittiwakes collected on Prince Leopold Island

in Lancaster Sound (Braune and Simon, 2003, 2004). These compounds had never been measured previously in Canadian Arctic seabirds. Samples included livers from 1975 and 1993, and egg samples from 1993. PCDDs ( $\sum$ PCDD=0.03–8.52 ng/g lipid weight), PCDFs ( $\sum$ PCDF=0.05–27.4 ng/g lw) and non-ortho PCBs ( $\sum$ nPCB=5.01–32.1 ng/g lw) were found in all the Arctic seabird samples analyzed in this study. Concentrations of HxCDDs, PnCDFs and HxCDFs, in particular, found in Arctic seabird livers exceeded levels reported for marine mammals in the Canadian Arctic (de March et al., 1998) by several orders of magnitude. Ringed seals, polar bears and walrus (*Odobenus rosmarus*) in the Canadian Arctic tend to contain relatively high TCDD and low PnCDD and PnCDF levels (de March et al., 1998) whereas Arctic seabirds showed the opposite pattern. Concentrations of PCDD/F homologs in the Arctic seabirds were in the range reported for seabirds from temperate North America and Europe (van den Berg et al., 1987; Hebert et al., 1994). Toxaphene was detected in every seabird sample analyzed (up to 64 ng/g ww) except for the pool of kittiwake livers from 1975 (Braune and Simon, 2004) and was one to two orders of magnitude lower than levels reported for marine mammals from the Canadian Arctic (Muir et al., 1999b).

### 3.2.3. New chemicals in seabirds

A number of new chemicals were also analyzed as part of the study on dioxins, furans and toxaphene in Arctic seabirds from Lancaster Sound described in Section 3.2.2.2. Total polybrominated diphenyl ethers (PBDEs) were present at low ng/g levels in most of the samples analyzed (Braune and Simon, 2004). The highest estimated PBDE levels were detected in the 1993 egg (3 ng/g ww) and liver samples of kittiwakes, and the 1993 murre and fulmar samples also contained ng/g levels. PBDE-47 was the major PBDE in all the samples. Although the data suggest that exposure to PBDEs has increased from 1975 to 1993, the highest level reported is an order of magnitude lower than levels reported for Swedish black guillemots (Sellström et al., 1993).

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



of the 1993 samples but only one of the liver samples from 1975 (Braune and Simon, 2004). The major contaminant found was 1,1'-dimethyl-3,3',4,4'-tetrabromo-5,5'-dichloro-2,2'-bipyrrole (DBP-Br<sub>4</sub>Cl<sub>2</sub>). The highest estimated level (a total of 5 ng/g ww) was found in the kittiwake egg sample from 1993. Trace amounts (<2 ng/g ww) of chlorinated terphenyls, a hexabromo biphenyl and TCP-methane were also detected in most of the Arctic seabird samples. No chlorinated diphenyl ethers, TCP-methanol or chlorinated naphthalenes were detected (detection limit ~2 ng/g ww) in any of these samples.

In a separate study, Tittlemier et al. (2002) examined levels of halogenated dimethyl bipyrroles in four seabird species (dovekie, black guillemot, black-legged kittiwake and glaucous gull) collected in northern Baffin Bay. HDBPs were detected in all seabird samples studied from the North Water Polynya, and this represents the first record of this group of compounds found in an Arctic species. Levels of DBP-Br<sub>4</sub>Cl<sub>2</sub> in the seabirds were generally similar to those recorded in seabird samples from Atlantic Canada (Tittlemier et al., 1999). DBP-Br<sub>4</sub>Cl<sub>2</sub> concentrations (1.76 ng/g) in dovekies were approximately 10 times lower than those of the Atlantic puffin (*Fratercula arctica*, 20 ng/g), a species that has a similar winter habitat but a more piscivorous diet. The difference in concentrations is likely driven by the fact that the dovekies were feeding at a lower trophic position just prior to their collection in early summer (Fisk et al., 2001c). In most of the bird species, DBP-Br<sub>4</sub>Cl<sub>2</sub> was the predominant congener. The exception occurred with black guillemots, where DBP-Br<sub>6</sub> was the most abundant congener. This may be due to black guillemots feeding on benthic organisms during part of the year (Gaston and Jones, 1998).

Perfluorooctane sulphonate (PFOS) is a member of the perfluorinated acid group of chemicals which are used in stain-repellents and in fire-fighting foams. In a recent survey of Arctic biota, detectable concentrations (1.3 ng/g ww) of PFOS were found in livers of northern fulmars from Lancaster Sound (Martin et al., 2004) whereas this compound was not detected in the black guillemots. The guillemots stay close to their breeding grounds all year, migrating only as far as the open water ice edges in winter and feeding primarily on marine fish and amphipods. In contrast, the fulmars, which also feed on marine amphipods and fish,

will also occasionally scavenge marine mammal carcasses and they migrate greater distances south toward the North Atlantic in winter.

#### 3.2.4. Chiral contaminants in seabirds

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

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

Hoekstra et al. (2003b) examined EFs of  $\alpha$ -HCH and chlordane compounds in the Arctic marine food web of the southern Beaufort-Chukchi Seas. The enantiomer fractions (EFs) of all chiral OCs were near racemic (EF=0.50) in seawater, zooplankton,

and all fish analyzed indicating lack of transformation. In contrast, the EFs for most OCs analyzed were non-racemic in marine mammal blubber (range: 0.09–0.79) because of enantiomer-specific biotransformation and (or) accumulation. However, as observed by Fisk et al. (2001c), EF values were not significantly correlated with trophic level.

### 3.3. Levels and spatial patterns of contaminants in marine mammals

More studies of OCs in Arctic organisms have focused on marine mammals than any other group of organisms, both historically and recently. The amount of data produced for Hg in marine mammals is much less but is still a significant contribution. The focus on marine mammals stems from their importance in the culture and diet of Inuit, but also because levels of persistent OCs and Hg, in general, are elevated in marine mammal tissues. It must be emphasized that the spatial trends of persistent OCs discussed in the following sections are often qualitative because they are based on evaluation of means and ranges of concentrations from different studies which may use different analytically based definitions of various OC groups, or may originally present concentration data on a different basis such as wet weight or lipid weight. In the case of the sums of groups of compounds, such as PCBs and chlordanes (CHL), some laboratories have included more congeners or compounds than others in  $\sum$ PCB and  $\sum$ CHL results. Rigorous comparisons of OCs between locations also require information on age, sex, blubber thickness, nutritional status, collection season, and reproductive status of the animals, all of which can have an important influence on contaminant concentrations (Muir et al., 2000c), while Hg concentrations may be affected by age, sex and dietary habit. This information has been collected for many locations, but has only been used qualitatively in this assessment of spatial trends.

#### 3.3.1. Mercury in marine mammals

**3.3.1.1. Ringed seals.** As part of a circumpolar study of hepatic and renal Hg in ringed seals, Riget et al. (2005) found that Hg concentrations overall were highest at three Canadian locations (Grise Fiord, Nunavut, and Sachs Harbour and Holman, NWT).

These three sites were part of a more extensive study of spatial trends within Canada, including ringed seals from Labrador, Northern Quebec (Nunavik), and Nunavut. Samples of liver, kidney and muscle were collected over the period 1998 to 2000 as part of the spring/summer hunt to determine total Hg concentrations (Muir et al., 2001; Muir and Kwan, 2003). For the data set as a whole, Hg concentrations were significantly correlated with age. Seals from Arviat and Grise Fiord were generally older than those from all other sites, making comparison among sites difficult. To examine spatial trends, a subset of similar aged animals was selected. Mercury concentrations were adjusted for age using analysis of covariance for samples ranging from 2 to 15 years of age, which represented about 70% of the total samples. After adjustment for age (to a mean age of 7.7 years), there were few significant differences among locations; however, concentrations of hepatic Hg were significantly lower in seals from Resolute than from Ungava Bay and Hudson Strait (Fig. 6). For kidney, Hg levels were significantly lower in Hudson Strait and Labrador compared with Pond Inlet or Resolute.

Organic Hg levels were determined in muscle, liver and kidney of 20 ringed seals from sites in Nunavik for which total Hg was determined (Muir et al., 1999c). The results show that organic Hg levels in muscle increased with age from about 80% of total Hg in animals aged 0–2 years to about 100% in adult animals (Muir et al., 2000b). Conversely, organic Hg in seal liver and kidney declined as a percentage of

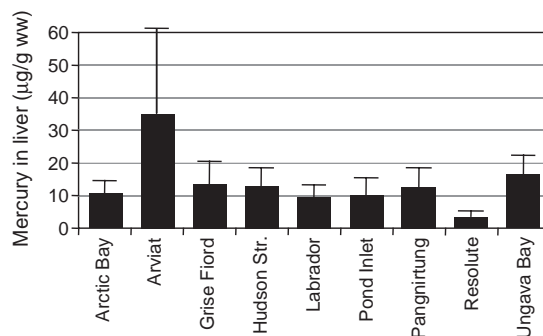


Fig. 6. Age-adjusted geometric mean concentrations (+95% confidence intervals) of mercury in ringed seal liver from nine locations in the eastern/central Canadian Arctic. Results are based on seals aged 2–15 years. Source: Muir et al. (1999c, 2001).

total Hg with increasing age, representing <5% in the oldest animals. Based on the results of Wagemann et al. (1997), organic Hg is likely to be entirely in the form of MeHg.

**3.3.1.2. Walrus.** All walrus in Canadian waters are from the Atlantic sub-species (*O. rosmarus rosmarus*), which plays an important role in traditional Inuit diets. Walrus feed predominantly on benthic invertebrates; however, some individual male walrus are believed to prey on ringed seals (Muir et al., 1995) and therefore feed at a higher trophic level than other walrus. These individuals have much higher concentrations of POPs than other walrus in the same area (de March et al., 1998) and also may have higher Hg concentrations, although this has not been determined.

Mercury levels in walrus muscle were relatively low compared with liver and kidney (Muir et al., 2000b) and are generally lower than in other species of marine mammals (Wagemann et al., 1996), which likely reflects the low trophic level of most walrus. Mercury concentrations in walrus landed at Inukjuak

(Muir et al., 2000b) were comparable to those reported previously by Wagemann et al. (1996) for the same population. About 75% of Hg in muscle, 5% in liver and 20% in kidney was in the form of organic Hg, probably MeHg (Wagemann et al., 1997).

**3.3.1.3. Beluga.** Beluga liver samples from 10 community or regional hunts (seven in Nunavut; two in Inuvialuit Region (Mackenzie Bay and Paulatuk); one in Hudson Strait in Nunavik) conducted between 1993 and 2000 were analyzed for Hg (Sang et al., 2000; Lockhart et al., 2001). The geographic pattern for Hg in adult beluga (range of mean ages: 8–15 years) is shown in Fig. 7. There were large differences between the beluga caught in Mackenzie Bay in 1996 and at Paulatuk in 1993, despite all being regarded as part of the same southern Beaufort Sea stock and the same mean age (~14 years). Within-stock variation was also apparent in beluga landed by communities in Hudson Bay and Baffin Island (Fig. 7). These intra-stock differences were apparent in previous assessments which compared animals collected the same year (Wagemann et al.,

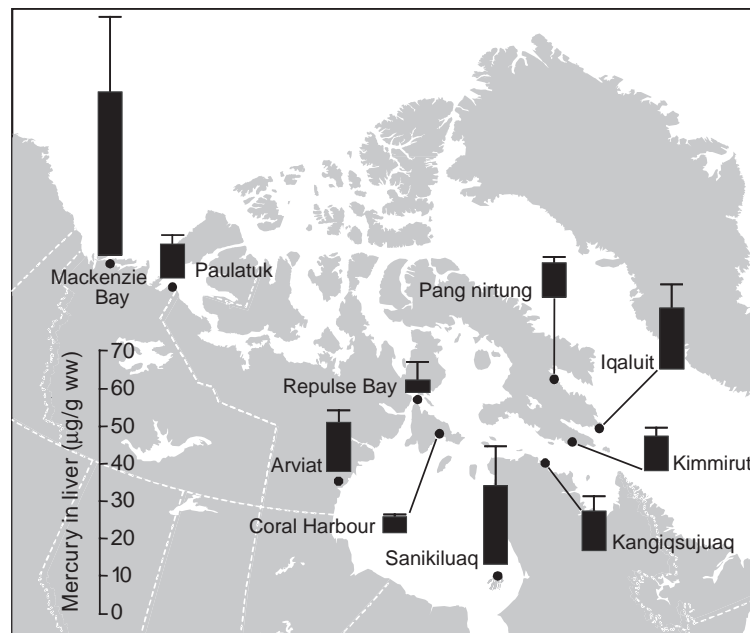


Fig. 7. Arithmetic mean concentrations ( $\pm 95\%$  confidence limits) of mercury in liver of adult belugas from 10 locations in the Canadian Arctic. Sample collections from 1993 to 2000. Average ages range from 8 to 15 years.

1996; Dietz et al., 1998), suggesting that the differences are long-standing. They may possibly be caused by subgroups of animals within stocks having a long-term affinity for specific feeding grounds which exhibit differences of Hg in prey species.

### 3.3.2. POPs in marine mammals

The bulk of the data reported for OCs and other POPs in marine mammals was concerned with concentrations in fat or blubber. The following sections deal mainly with those data, with a final Section (3.3.2.7) summarizing the data on OCs in other types of marine mammal tissue.

**3.3.2.1. Ringed seals.** There have been a large number of new measurements of OCs in the blubber of Canadian Arctic ringed seals since the first CACAR report, mainly as a result of temporal trend studies. These also allow spatial patterns to be studied. Previous studies had shown that OC levels in ringed seals were higher in Canada than in Alaska but lower than in the European Arctic (Muir et al., 2000c). Some geographical variation of OCs in ringed seal blubber was observed within the Canadian Arctic but the magnitude was small and generally within a factor of 2 (Muir et al., 1999b). OC concentrations in this species reflect its trophic position, falling as it does between lower trophic level organisms such as Arctic cod and top trophic feeders such as polar bear.

For most OCs, the lowest levels were observed in the western and central Canadian Arctic archipelago as illustrated for  $\Sigma$ PCB in female ringed seals (Fig. 8). Females were selected because their concentrations of OCs are not strongly affected by age. Ringed seals from Holman, N.W.T. (Hoekstra et al., National Water Research Institute, unpublished data), had levels of PCBs and OC pesticides that were intermediate between those from the Beaufort/Chukchi Seas (Hoekstra et al., 2003a; Kucklick et al., 2002) and those in the eastern Canadian Arctic (Fisk et al., 2002a). Similar trends were seen for  $\Sigma$ DDT and chlordane-related compounds. The exception was HCH (Fig. 8), which was higher in seals from the western Canadian Arctic and Alaska due to prevailing high levels in seawater in the Chukchi/Beaufort Seas (Li et al., 2002).

Smaller differences in mean OC concentrations in ringed seals were seen among seven sites within Nunavik and Labrador in 1998 and 1999 (Muir et al., 2000b). For comparison among regions, three sites within Ungava Bay (Kuujuuaq, Kangirsuk and Kangiqsualujuaq) were combined, as were two sites in the Hudson Strait (Salluit and Quaqtaq). The two sites within Labrador (Nain and Makkovik) were treated separately. Average blubber concentrations of  $\Sigma$ PCB, the major OC contaminants in seals from these four locations, ranged from 572 to 1042 ng/g ww in males, and from 512 to 730 ng/g in

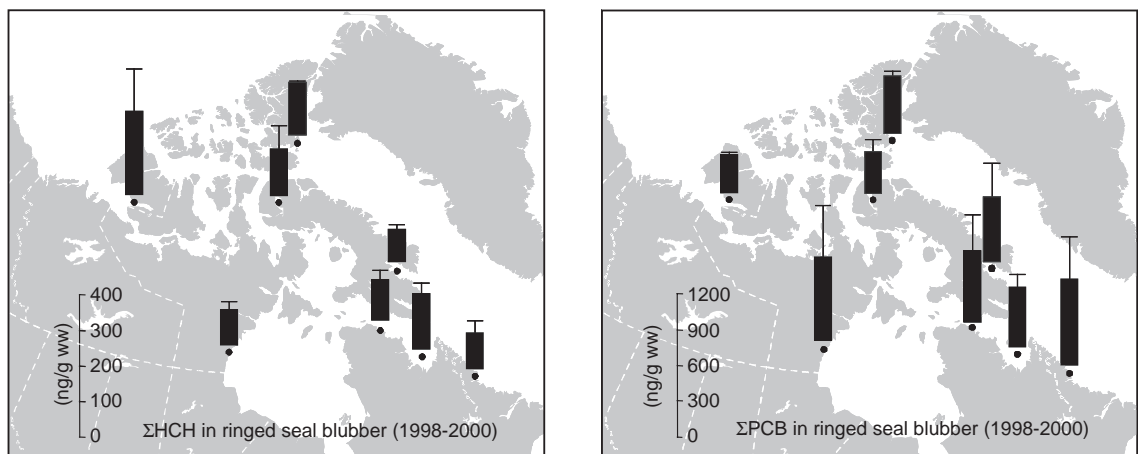


Fig. 8. Mean ( $\pm$  95% confidence interval) concentrations of  $\Sigma$ PCB and  $\Sigma$ HCH in female ringed seal blubber (ng/g ww) from samples collected between 1998–2000.

females (Muir et al., 2000b). The highest levels in males were found in the more westerly locations of Ungava Bay and Hudson Strait, while the highest average levels in females occurred at Nain. DDT metabolites were also prominent in ringed seal blubber, with average concentrations ranging from 198 to 884 ng/g in males. Analysis of covariance did not reveal any significant differences in concentrations of  $\sum$ PCB or  $\sum$ DDT levels among the four locations for females. For males,  $\sum$ PCB,  $\sum$ DDT and  $\sum$ HCH concentrations were significantly higher in samples from Ungava Bay, after adjusting for age. However, the results may have been affected by a spatially varying relationship of age with OC concentrations among males.

In general, the spatial trends in levels of  $\sum$ PCBs,  $\sum$ DDT and  $\sum$ HCH in ringed seal blubber collected in 1998–2000 were comparable to those found previously at other eastern Canadian Arctic locations (Muir and Addison, 1997; Weis and Muir, 1997; Letcher et al., 1998). Results for ringed seals in northern Baffin Bay were also in the range reported in northeast and central-west Greenland (Johansen et al., 2004). After removing the influence of age, sex and blubber thickness, OC concentrations did not differ between ringed seals from the east and west sides of northern Baffin Bay (Fisk et al., 2002a).

Samples from ringed seals collected in 1993 near Kimmirut in Hudson Strait were analyzed for coplanar PCBs (Helm et al., 2002).  $\sum$ CoPCB (sum of congeners 77, 81, 105, 126, 118, 114, 156, 169) levels ranged from 14.9 to 32.7 ng/g ww, with congeners 118, 105 and 156 being the most dominant. Mean ( $\pm$ S.D.) levels of  $\sum$ CoPCB were lower in females ( $15.4 \pm 0.8$  ng/g) than in males ( $26.7 \pm 8.0$  ng/g). Toxic equivalents (TEQs) for CoPCBs in males and females were  $624 \pm 144$  fg/g and  $508 \pm 88$  fg/g, respectively, most of which was accounted for by congener 126 (Helm et al., 2002). Earlier work by Ford et al. (1993) and Muir et al. (1995) found higher  $\sum$ CoPCBs in ringed seals collected in the late 1980s and early 1990s in Hudson Bay and Baffin Bay (58–111 ng/g lipid wt). The difference may be due to the fact that ringed seals from the latter locations generally had higher total PCBs than at Pangnirtung (Weis and Muir, 1997).

Toxaphene was measured in Hudson Strait and Ungava Bay ringed seals (Muir et al., 2000b). As is

typical of marine mammals, congeners 26 and 50 were predominant. Octa- and nonachlorobornanes were the major homolog groups (Muir et al., 1999d), and total toxaphene levels in blubber were similar to those reported by Cleemann et al. (2000) for seals in Greenland. Toxaphene congeners 39, 40 and 42 were also identified in the blubber of ringed seals from Arviat (Loewen et al., 1998), marking the first time that congener 42, the most toxic congener in technical toxaphene, has been found in any significant concentration in a marine mammal.

**3.3.2.2. Walrus.** Relatively little is known about levels of OCs in walrus compared to other mammals. Individual male walrus which prey on ringed seals (Muir et al., 1995) have higher contaminant concentrations than other benthic-feeding walrus in the same area (de March et al., 1998). Organochlorine levels recently determined in walrus from eastern Hudson Bay (northern Quebec) were much lower than those found in earlier studies from the same area (Muir et al., 2000b), likely because the earlier studies included walrus that consumed seals (Muir et al., 1995). There appears to be little geographic variation of OCs among walrus. Eastern Hudson Bay walrus (excluding the seal-eaters) had similar OC levels to those in Foxe Basin (Muir et al., 1995), while their  $\sum$ PCB,  $\sum$ HCH and HCB concentrations were similar to those in the Pacific walrus sub-species (*O. rosmarus pacificus*) from the Bering Sea and in Atlantic walrus in eastern and northwestern Greenland (Muir et al., 2000a). Their  $\sum$ DDT levels were also comparable to walrus from northwestern Greenland. The exception was  $\sum$ CHL, with levels in eastern Hudson Bay walrus higher than those in the Bering Sea and northwestern Greenland, but lower than in east Greenland (Muir et al., 2000a).

**3.3.2.3. Beluga.** The previous CACAR assessment included OC data (up to 1996) for beluga from Hudson Bay, Hudson Strait and Cumberland Sound in the eastern Arctic and Mackenzie Bay, N.W.T. Since then, additional blubber samples have been analyzed from the latter two locations as a result of temporal trend studies (Stern, 1999; Stern and Addison, 1999; Stern and Ikonou, 2001; Stern et al., 2005) and as part of studies in Nunavik (Sang et al., 2000).



Stern et al. (2005) summarized concentrations of PCBs and OC pesticides in beluga blubber from 13 locations in the Canadian Arctic (Nunavut and the N.W.T.) collected over the period 1992–2000. The major OC contaminants were PCBs and toxaphene. Stern et al. (2005) found that age-corrected concentrations (least square means) of *p,p'*-DDE, total PCBs, toxaphene and total chlordane in males were significantly higher at south Baffin Island sites (Kimmirut and Iqaluit) than in the western Canadian Arctic (Mackenzie Bay). Notable exceptions were HCB and  $\beta$ -HCH which were generally higher in the west.

The spatial trends of PCBs and toxaphene in beluga are shown in Fig. 9, which combines results from several recent studies. Levels of major OC groups in male beluga from Nunavut (Sang et al., 2000) were similar to previous data for males from eastern Hudson Bay in the mid-1980s (Muir et al., 1990) but were much lower than in the southeast Baffin beluga stock. Muir et al. (1990) concluded that beluga sampled from the Kangiqsujuaq area of Hudson Strait were from a population that inhabits eastern Hudson Bay and southern Hudson Strait, and not the southeast Baffin Island area. Innes et al. (2002)

also concluded that there were distinct differences in the OC signature of southeast Baffin and Hudson Bay beluga. Likewise, Stern et al. (2005) observed that beluga from Sanikiluaq in southeastern Hudson Bay could be distinguished from other Hudson Bay and south Baffin Island samples on the basis of a unique pattern of PCB congeners and OC pesticides using Principle Component analysis.

Levels of coplanar PCBs in six beluga from Kimmirut (southeastern Baffin Island) ranged from 14.4 to 294 ng/g, with congeners 118, 105 and 156 being the most dominant (Helm et al., 2002).  $\Sigma$ CoPCB was lower in females ( $60.2 \pm 40.3$  ng/g) than in males ( $228 \pm 113$  ng/g). Toxic equivalents (TEQs) for CoPCBs in males and females were  $1730 \pm 627$  fg/g and  $1320 \pm 909$  fg/g, respectively, most of which was accounted for by congener 126 (Helm et al., 2002). These TEQs in beluga were generally lower than reported in previous studies (Ford et al., 1993; de March et al., 1998). This may be due in part to differences in the toxic equivalency factors for dioxin-like PCBs used in the previous work.

Mean toxaphene levels in beluga blubber from various locations in the Canadian Arctic ranged

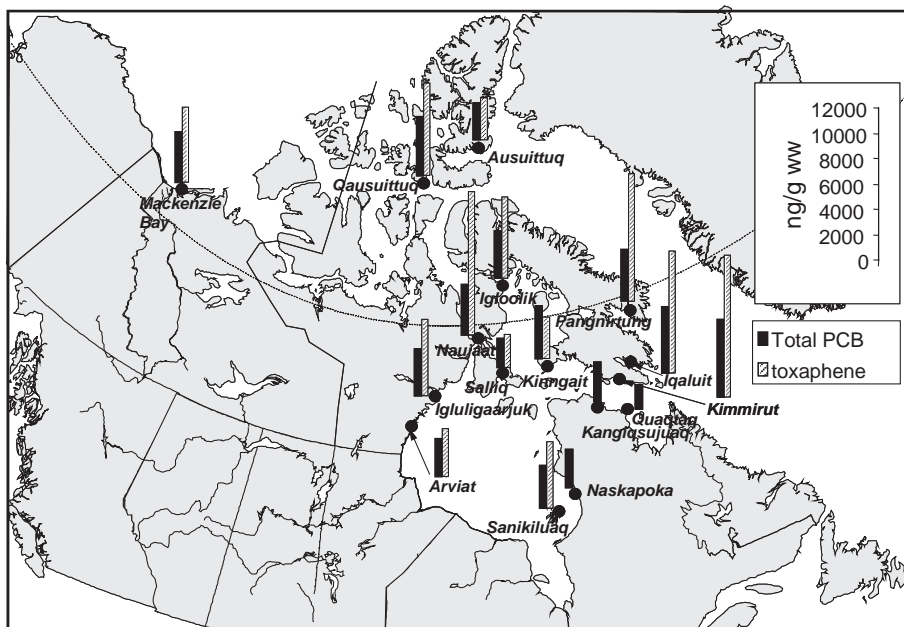


Fig. 9. Geometric mean concentrations of  $\Sigma$ PCB and toxaphene in beluga whale blubber (ng/g ww) from samples collected 1994–2000 (from Sang et al., 2000; Stern et al., this issue).

from means of 2654 to 11,300 ng/g (ww) (Stern et al., 2005), which is on the high end of the range reported in beluga from Alaska, Greenland and Svalbard (Wade et al., 1997; Johansen et al., 2004). The toxaphene results from Canada were based on quantification by gas chromatography-electron capture detection while other results were by mass spectrometry, which may explain, in part, the higher levels.

**3.3.2.4. Narwhal.** Narwhal (*Monodon monoceros*) is an important species in the traditional diet of Inuit. Despite this, they have received less attention in terms of contaminant studies than beluga. In the Canadian Arctic, levels of PCBs, DDTs and chlordanes (uncorrected for age) were generally higher in male narwhal from Pond Inlet (northeast Baffin Island) than those from Broughton Island (eastern Baffin Island), or Grise Fiord (southern Ellesmere Island).  $\sum$ PCB concentrations in male narwhal blubber ranged from means of 3440 ng/g ww at Grise Fiord to 5820 ng/g at Pond Inlet (Stern, Fisheries and Oceans Canada, unpublished data). Similar patterns occurred for toxaphene in narwhal from these sites with means ranging from 7400 ng/g ww at Broughton Island to 11,800 ng/g ww at Pond Inlet. Levels of PCBs and DDTs in these narwhal were quite similar to levels reported in western Greenland (Dietz et al., 2004), but they were only about half those measured in narwhal from Svalbard (Wolkers, Norwegian Polar Institute, unpublished data).

**3.3.2.5. Polar bears.** Polar bears are widely distributed throughout the Arctic and sub-Arctic region and range over large areas in search for food. They move south with the ice in the fall and winter and then north as the pack ice melts in the spring and summer. These seasonal movements of sea ice also influence the distribution and concentration of their primary prey, ringed and bearded seals (Stirling et al., 1982; Kingsley et al., 1985). Polar bears are the top Arctic predator, and often eat only the blubber from a seal (Stirling and McEwan, 1975), where the highest concentrations of OC are found. Therefore polar bears generally have among the highest concentrations of OCs of any Arctic animal. Polar bears also have superior OC biotransformation capacity and have the highest levels of OC metabolites, some of which have demonstrated endocrine disrupting activity (Letcher et

al., 2000a). Because of this, concerns about the effects of OC exposure in Arctic wildlife are greatest for this species. Although sometimes consumed by the Inuit, the polar bear makes up a small percentage of their diet.

Verreault et al. (2005) determined PCBs and OC pesticides in adipose tissue of adult and subadult polar bears, almost exclusively females, sampled between 1999 and 2002 from 6 populations in the Canadian Arctic as well as northwest Alaska, east Greenland, and Svalbard. The Svalbard and East Greenland populations of polar bear, which are in the eastern portion of the Western Hemisphere Arctic range of these animals, had significantly higher concentrations of PCBs and DDT than those from Canada or Alaska (see also Lie et al., 2003). Within the Canadian Arctic, geometric mean concentrations of  $\sum$ PCB ranged from 1130 ng/g lipid wt in the Foxe Basin/Gulf of Boothia population to 2800 ng/g in the southern Baffin Island group (Fig. 10).  $\sum$ HCH concentrations were distinctly higher highest in samples from Alaska and Amundsen Gulf (southeastern Beaufort Sea) than in the eastern Canadian Arctic or Greenland. The distribution of chlordanes-related compounds (CHLs) was relatively uniform throughout the North America Arctic (Fig. 10). The results from 1999 to 2002 were in general agreement with those of Norstrom et al. (1998) which were based on samples collected in 1989–1991.

**3.3.2.6. Effects of age and sex on organochlorine levels in marine mammals.** Age and sex are important factors that must be taken into account to assure accurate comparisons of contaminant levels in marine mammals, such as variations in geographical and temporal trends. Concentrations of  $\sum$ PCB,  $\sum$ DDT and  $\sum$ CHL compounds were found to increase with age for both male and female ringed seals in the Nunavut region (Fisk et al., 2002a). Sex was an important variable within the context of seal age, and in seals of similar ages OC concentrations were greater in males than in females (Weis and Muir, 1997; Fisk et al., 2002a). These relationships were not observed for  $\sum$ CBz and  $\sum$ HCH in seals from the Nunavut region, although there was an age-within-site significance for  $\sum$ HCH (Fisk et al., 2002a). These data are consistent with previous findings for ringed seals, in which positive relationships between OC

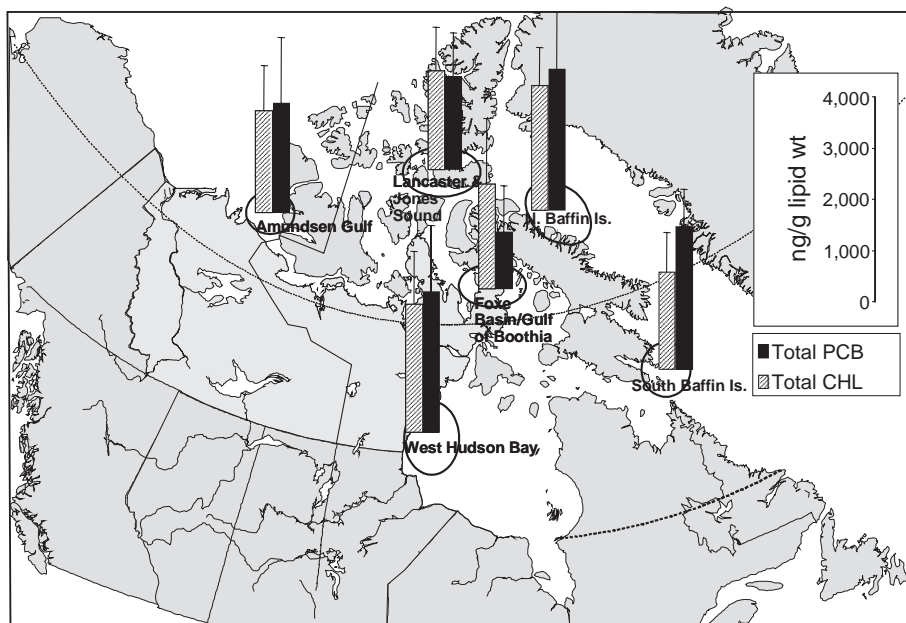


Fig. 10. Geometric mean concentrations (with 95% confidence limits) for sums ( $\Sigma$ ) of PCB congeners and chlordane-related compounds (CHL) (ng/g lipid weight) from 6 locations in the Canadian Arctic (from Verreault et al., this issue).

concentration and age were observed for males (Addison and Smith, 1998; Wolkers et al., 1998; Muir et al., 2000c) but less commonly so in females (Kostamo et al., 2000). Reproduction, including birth and lactation, provides a means for female seals to reduce body burdens of OCs, and counters the effect of accumulation of OCs with age (Fisk et al., 2002a).

The effects of age and sex are also important considerations with respect to PCBs and OC pesticides in polar bears. In 1995, Norstrom (1999) examined concentrations of OCs in male and female polar bears from Hudson Bay to test what the effect of gender was in a single year. Concentrations of OCs in males and females were similar; however, there was a (generally non-significant) tendency for most residues to be lower in males, except PCBs and DDTs which were slightly higher in males. The difference in PCBs between males and females was not as large as observed in more extensive data sets. For bears from the European and North American Arctic as a whole, Norstrom et al. (1998) showed that males had 40% higher PCBs than females on average. Younger male bears had PCB concentra-

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

**3.3.2.7. Organochlorines in other marine mammal tissues.** Fewer analyses of OCs in brain, liver, kidney, muscle and blood have been made as compared to blubber in pinnipeds and cetaceans. In all other tissues, however, OC concentrations are lower than in blubber because of lower lipid content. Low ng/g ww levels of PCBs and OC pesticides were found in brain, liver and muscle from beluga from Hendrickson Island, in the western Canadian Arctic

(Metcalf et al., 1999). Lipid content is an important factor in controlling the accumulation of lipophilic OCs, and in beluga the concentrations of OCs in these tissues, with the exception of brain, were generally more comparable when the values were calculated on a lipid weight rather than on a wet weight basis (Krahn et al., 2001). Beluga brain samples could be distinguished from other tissues by differences in PCB congener patterns and higher concentrations of  $\sum$ HCH (primarily  $\alpha$ -HCH) (Metcalf et al., 1999). These variations among tissues may be influenced by differences in contaminant metabolism, the content and composition of lipids, or the degree of blood perfusion in the various tissues (Metcalf et al., 1999).

Concentrations of OCs were determined in the blood plasma of Resolute Bay polar bears (Sandau et al., 2000). This work also included plasma samples from Svalbard bears.  $\sum$ CHL and  $\sum$ PCB were the dominant OC groups found in plasma in both regions. OC concentrations were two times higher in subadults than adults, except for DDTs which were similar in both age groups and in line with previous findings in bear adipose tissue. The exceptions were chlordanes, which were 30–60% lower in males, but concentrations were similar comparing the same sex in both areas. It has been shown that males metabolize chlordanes more readily than do females, which accounts for the differences between genders (Polischuk et al., 2002).  $\sum$ PCB concentrations were similar in males and females from both areas and 2–3 times higher in the Svalbard bears, which is in agreement with previous analyses of adipose tissue from these areas (Norstrom et al., 1998).

The need for caution in interpreting OC concentration data from different tissues is further demonstrated by findings reported by Lydersen et al. (2002), who examined how concentrations of OCs in blood and blubber varied with nutritional condition in captive and wild fasting ringed seals. The study demonstrated that extreme variability occurs in OC concentrations in blood in response to changes of body condition during fasting, and that the response of blubber OC concentrations is also very different than those in blood. The authors recommended that since the natural annual variation of condition is extreme for phocid seals, blood should not be used in studies of OCs where the aim of the study is to monitor OC levels for

comparative purposes or time-trend analysis. The same caution may well apply to studies involving other Arctic marine mammals.

### 3.3.3. New compounds in marine mammals

Of the data produced on new chemicals in the Arctic, the greatest amount concerns marine mammals and, in particular, seals and whales. These are, logically, the animals to check for new chemicals because seals and whales have among the highest levels of OCs in the Arctic.

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

**3.3.3.2. Perfluorooctane sulphonate (PFOS).** Perfluorooctane sulphonate (PFOS), a member of the perfluorinated acid group of chemicals which are used as stain resisting surfactants and in fire fighting foams, was detected in the blood of ringed seals from Pangnirtung with concentrations ranging from <3 to 12 ng/mL (Giesy and Kannan, 2001; Kannan et al., 2001). No obvious differences of PFOS were observed between ringed seals in the Canadian and European Arctic, although only a very small data set was available. More recently, Martin et al. (2004) found mean PFOS concentrations of 16 and 19 ng/g ww in livers of ringed seals from Holman Island and Grise Fiord, respectively. PFOS was also detected in the liver of polar bears (mean 350 ng/g ww) from northern Alaska (Giesy and Kannan, 2001; Kannan et al., 2001) and, more recently, concentrations 10-fold



higher than these were found in livers of polar bears from eastern Hudson Bay (Martin et al., 2004). This geographical pattern suggests that Hudson Bay may be closer to important regional sources of PFOS than Alaska. A similar latitudinal difference in PFOS concentrations was found in ringed seals from the Norwegian Arctic versus the Baltic Sea (Giesy and Kannan, 2001). The mean level of PFOS in the Hudson Bay bear livers was 3.1 µg/g ww, making PFOS the most abundant organohalogen contaminant detected in polar bears to date (Martin et al., 2004).

Tomy and Helm (2003) reported PFOS and its precursors, perfluorooctanesulfonamide (PFOSA) and *N*-ethyl-perfluorofluorosulfonamide (NEtFOSA) in narwhal and beluga whale liver samples from the eastern Canadian Arctic. PFOS averaged 10 and 12 ng/g ww and PFOSA was present at 1.5 and 2.8 ng/g (ww), respectively, in narwhal and beluga liver. The precursor NEtFOSA was undetectable in whale liver but was detected in redfish (*Sebastes marinus mentella*), indicating that cetaceans which feed on redfish may be capable of degrading NEtFOSA.

Martin et al. (2004) also identified, for the first time, a series of perfluorocarboxylic acids (PFCAs) in seals, seabird liver and fish. Concentrations of total PFCAs (C8–C15-PFCAs) were lower than total PFOS equivalents (PFOS) in all marine and freshwater samples.

**3.3.3.3. Polychlorinated naphthalenes (PCNs).** Polychlorinated naphthalene ( $\sum$ PCN) concentrations in blubber from six ringed seals sampled in 1993 near Kimmirut, Nunavut, ranged from 29 to 63 pg/g ww (Helm et al., 2002). No differences related to sex were observed for distribution of PCN homolog groups, and most of the PCN TEQ was due to two hexa-CN congeners, CN-66 and CN-67. The contribution to TEQ by PCNs was negligible (<0.10–2.4 fg/g ww) due to an absence of the more toxic hexa-CN congeners. Blubber from six beluga whales collected in 1994 near Pangnirtung were also analyzed for PCNs (Helm et al., 2002), and  $\sum$ PCN ranged from 40 to 384 pg/g ww. Penta-CN congeners dominated, followed by hexa- and tetra-CN congeners.  $\sum$ PCN levels were lower in females than in males. Most of the PCN TEQ was due to two hexa-CN congeners, CN-66 and CN-67. Although  $\sum$ CoPCB concentrations averaged about 500 times greater than  $\sum$ PCN in beluga blubber,

PCNs contributed 11% of the TEQ relative to CoPCBs, about the same as the contribution from mono-*ortho* PCBs CB105 and CB118. Thus, PCNs may be significant contributors to TEQ in beluga.

**3.3.3.4. Polybrominated diphenyl ethers (PBDEs).** PBDEs have been determined in beluga (Stern and Ikonomou, 2001) and in ringed seals (Ikonomou et al., 2002) in the Canadian Arctic. In general, concentrations of  $\sum$ PBDEs were orders of magnitude lower than legacy OCs such as PCBs and DDTs. PBDEs consist of a much smaller number of individual compounds, however, so the difference between concentrations of individual PBDE and individual PCB congeners is smaller than the sum concentrations suggest. BDE 47 is the most common congener measured, followed by BDE 99 and 153. Other congeners, such as BDE 100 and 49 have been measured in Canadian Arctic beluga (Stern and Ikonomou, 2001). These congeners are found at lower concentrations in the technical PBDE standards but may also be less prevalent due to biotransformation.

Although it is premature to draw conclusions about spatial trends of PBDE in Arctic marine mammals, there is sufficient data to suggest that concentrations are higher in the European Arctic than in the North American Arctic. The mean concentration of  $\sum$ PBDE were 92.9 ± 56.5 ng/g ww in Svalbard beluga blubber (van Bavel et al., 2001) compared with 15.5 ng/g in beluga from the western Canadian Arctic (Stern and Ikonomou, 2001). As well,  $\sum$ PBDE concentrations in the blubber of ringed seals from northeast Greenland (58 ± 23 ng/g) were an order magnitude higher than in western Greenland (3.6 ± 1.1 ng/g) (Johansen et al., 2004) and in the western Canadian Arctic (4.6 ng/g ww) (Ikonomou et al., 2002). Higher  $\sum$ PBDE concentrations were also detected in ringed seals from Svalbard (averaging 18 ng/g lipid wt) than in the Canadian Arctic or western Greenland (Wolkers et al., 2004).

PBDEs were determined in fat samples of female polar bears between the ages of 5 and 15 years collected in 2001–2002 from six populations spanning Nunavut and N.W.T. (Letcher et al., 2003). Mean  $\sum$ PBDE concentrations (ng/g, lipid wt) of 13, 35, 33, 13, 12 and 12 were found in bears from the Beaufort Sea, western Hudson Bay, southeastern Baffin Bay, Foxe Basin/Gulf of Boothia and Lancaster/Jones Sounds, respectively. These results demon-



strated that  $\sum$ PBDE concentrations in polar bears were higher than in the blubber of ringed seal from the same regions (Ikonomou et al., 2002). Furthermore, in female polar bears, BDE-47 accounted for 50% or more of the  $\sum$ BDE levels across all populations examined. BDE-153 was the next most abundant congener followed by BDE-99. Ikonomou et al. (2002) found an even greater dominance of BDE-47 in Canadian ringed seal blubber. This may indicate that higher brominated BDE congeners are bioaccumulating in polar bear relative to ringed seal, and/or that lower brominated BDE congeners are more rapidly metabolized in polar bear relative to ringed seal (Hakk and Letcher, 2003).  $\sum$ PBDE concentrations in polar bears from the Canadian Arctic were generally lower than those from Svalbard where Wolkers et al. (2004) found mean concentrations of 46 ng/g in females.

**3.3.3.5. Chlorinated paraffins (SCCPs).** Chlorinated paraffins (SCCPs) were detected at ng/g levels in beluga and ringed seals in the Canadian Arctic (Muir et al., 1999a; Tomy et al., 2000). Concentrations were similar in beluga from Mackenzie Bay, NWT, and Kimmirut in Hudson Strait. Ringed seals from Eureka had higher concentrations (mean 527 ng/g ww) than seals from Pangnirtung (mean for both sexes was 95 ng/g). The seals from Eureka were much older than those from Pangnirtung, and it is possible that SCCP levels increase with age as do other recalcitrant OCs, although further measurements are needed to confirm this. Both species had slightly higher mean levels in males than females, suggesting lactational or placental transfer of SCCPs. The SCCPs profile consisted of similar proportions of chlorodecanes, undecanes and dodecanes with very low or negligible amounts of tridecanes. Few other published data exist on SCCPs in marine mammals and differences in analytical methodology make comparisons problematic. Jansson et al. (1993) reported 130 ng/g SCCPs in ringed seal blubber from Svalbard, which is similar to levels found in the Canadian Arctic. St. Lawrence beluga had 6–8 times higher levels of SCCPs than Arctic beluga (Tomy et al., 2000) which illustrates the importance of local sources for this southern population of whales. The St. Lawrence animals had a pattern of SCCPs which resembled the pattern found in commercial products, while the Arctic beluga had higher proportions of the more volatile

chlorodecanes and undecanes. This pattern is consistent with long-range atmospheric transport to the Arctic being the most important source of SCCPs. There was no correlation between levels of PCBs or toxaphene and SCCPs in ringed seals and beluga. This was unexpected because levels of most recalcitrant OCs in beluga blubber are correlated. It implies that SCCPs undergo a greater degree of biotransformation or have quite different sources than PCBs.

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

Enantiomer fractions (EFs;  $EF = ER/(ER + 1)$ ) of  $\alpha$ -HCH and other chiral contaminants in seal blubber may not reflect the metabolic capability of seals. Wiberg et al. (1998b, 2000) noted near-racemic  $\alpha$ -HCH (EFs  $\approx 0.52$ ) in blubber of ringed seals but non-racemic values in liver (EFs  $\approx 0.6$ ). This phenomenon was observed with other chiral pollutants such as *trans*-chlordane, but in some cases the EF was greater in blubber (Wiberg et al., 2000). Wiberg et al. (1998b) attributed this difference to greater metabolic activity in the liver as compared with the blubber. This would imply that the proportion of the  $\alpha$ -HCH body burden that is transformed is small, and consequently that the EF in seal blubber is closer to that of the diet than is the EF of the liver. This is not always the case, since EFs of many chiral OCs in ringed seal blubber have been found that do not match those in their main prey item, Arctic cod (Wiberg et al., 2000; Moisey et al., 2001). In seven species of seabirds, animals that do not retain as large a fat reserve as ringed seals, there were no differences in EFs of chiral chlordanes between liver and fat (Fisk et al., 2001c). Differences

in EFs of chiral pollutants between tissues of seals, and potentially other marine mammals, require further study.

Enantiomeric fractions of chiral contaminants and stable isotopes of nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) were measured along with OCs in ringed seals collected from the east and west sides of the North Water Polynya in northern Baffin Bay (Fisk et al., 2002a). *Cis*- and *trans*-chlordane, oxychlordane and heptachlor epoxide were all non-racemic in the ringed seal blubber but did not vary with age, sex or collection site. *a*-HCH appeared racemic (enantiomeric fraction =  $0.50 \pm 0.01$ ) in the seals, although this EF is different than those previously observed in their prey species, and was found to vary significantly with age. An overall food web assessment of *a*-HCH in the North Water Polynya concluded that ringed seals did not metabolize *a*-HCH efficiently (Moisey et al., 2001). EF values in the seals varied considerably from other Arctic marine mammals and seabirds, providing additional evidence that the type(s) and characteristic(s) of the enzymes involved in biotransformation of chiral OCs vary among these organisms.

**3.3.3.7. Organochlorine metabolites.** Two classes of OC metabolites that have been established as significant contaminant residues, often at levels comparable to known classes of OCs, in the tissues of Arctic marine mammals are hydroxylated (HO) and methyl sulfone ( $\text{MeSO}_2$ ) metabolites of PCBs (Letcher et al., 2000a).  $\text{MeSO}_2$ -PCBs and -DDEs have been reported in marine mammals, birds and humans from a number of regions globally; however, most reports have concerned marine mammals from the Arctic and other northern latitudes (e.g., the North and Baltic Seas) (Letcher et al., 2000a).

Knowledge is limited concerning methylsulfone ( $\text{MeSO}_2$ )-PCBs and  $\text{MeSO}_2$ -DDE in cetaceans, including species and populations from the Canadian Arctic. 28  $\text{MeSO}_2$  metabolites of 14 *meta*-*para* chlorine-unsubstituted PCBs, four  $\text{MeSO}_2$  metabolites of 4,4'- and 2,4'-DDE, as well as PCBs and DDTs, were determined in blubber biopsies of beluga from western Hudson Bay (sampled during 1993–1994). These data were compared to those for beluga from the St. Lawrence River sampled between 1994 and 1996, two populations which are contrasted by their exposure to different levels of chlorinated hydrocarbon

contaminants (Letcher et al., 2000b). The mean concentration of  $\sum \text{MeSO}_2$ -PCB in male beluga from western Hudson Bay (159 ng/g), and the ratios of  $\sum \text{MeSO}_2$ -PCB to  $\sum \text{PCB}$  (0.03) and  $\sum \text{precursor-PCB}$  (0.08) were approximately 2-fold lower than in St. Lawrence River males, whereas the  $\sum \text{precursor-PCB}$  to  $\sum \text{PCB}$  ratio was approximately 2-fold higher. Both populations had a low formation capacity for  $\text{MeSO}_2$ -PCBs with  $\geq 6$  chlorines ( $<4\%$  of  $\sum \text{MeSO}_2$ -PCB). The congener patterns were dominated by trichloro- and tetrachloro- $\text{MeSO}_2$ -PCBs in western Hudson Bay animals, whereas they were dominated by tetra- and pentachloro- $\text{MeSO}_2$ -PCBs in St. Lawrence beluga. In addition to 2- and 3- $\text{MeSO}_2$ -4,4'-DDE, two unknown  $\text{MeSO}_2$ -2,4'-DDEs were detected. The mean 3- $\text{MeSO}_2$ -DDE concentration ( $<0.01$  ng/g) in the western Hudson Bay animals was much lower than in St. Lawrence animals (1.2 ng/g). The data suggested that sulfone formation and clearance was related to contaminant-induced metabolic capacity, and thus PCB, DDE and  $\text{MeSO}_2$ -PCB and -DDE toxicokinetics differed between beluga in the two regions.

Polar bears have a superior ability to biotransform OCs. For example, the PCB burden in polar bears is dominated by a small number of congeners, much fewer than are observed in their major prey item, the ringed seal (Muir et al., 1988). There have been several recent studies on  $\text{MeSO}_2$ - and HO-PCB metabolites in Canadian polar bears.  $\text{MeSO}_2$ -PCBs were examined in fat samples collected in 2001–2002 from female polar bears between the ages of 5 and 15 years from six populations spanning the Nunavut and Northwest Territories (Letcher et al., 2003). Mean  $\sum \text{MeSO}_2$ -PCB concentrations ranged from 90 to 157 ng/g (lw), which were markedly lower than those in eastern Greenland polar bears (699 ng/g lw; Sandala et al., 2004).  $\text{MeSO}_2$ -PCB and  $\text{MeSO}_2$ -4,4'-DDE metabolites were examined in the tissues of bears collected from Resolute Bay in 1993 (Letcher, 1996; Letcher et al., 1996, 1998; Norstrom, 1997; Wiberg et al., 1998a)). Concentrations of  $\sum \text{MeSO}_2$ -PCB were highest in liver ( $3049 \pm 1290$  ng/g lw) and were about 10% of the concentrations of  $\sum \text{PCB}$ . In fat, testes, lung and brain,  $\sum \text{MeSO}_2$ -PCB was 2, 9, 13 and 60 times lower than in liver on a lipid weight basis. The major congeners in all tissues were 3- and 4- $\text{MeSO}_2$ -CB87 and 3- and 4- $\text{MeSO}_2$ -CB101.  $\text{MeSO}_2$ -PCB uptake

from seals appeared to be the most important source of MeSO<sub>2</sub>-PCBs in bears. 3-MeSO<sub>2</sub>-4,4'-DDE concentrations (303 ± 85 ng/g lw) in liver were nearly half those of DDE and were 126 to 337 times higher than in the testes, adipose and lung. The highly asymmetric tissue distribution of ∑MeSO<sub>2</sub>-4,4'-DDE may be due to several factors, such as the liver being the site of formation or because of highly selective binding in liver cells. It is not possible to clarify the relative importance of MeSO<sub>2</sub>-DDE bioaccumulation versus formation in the bear, based on adipose tissue concentrations. MeSO<sub>2</sub>-PCBs are efficiently transferred from polar bear females to cubs via milk, resulting in concentrations that are about 3 times higher in the cubs than in their mothers, compared to about 2 times higher for PCBs (Letcher, 1996). The 3-MeSO<sub>2</sub>-4,4'-DDE was not preferentially transferred to cubs.

There are increasing reports of chlorinated phenolic compounds (CPCs) that are metabolically derived from PCBs (HO-PCBs) (Letcher et al., 2000a), as well as other primary and metabolite CPCs, in the blood of Arctic biota. In one study, phenolic compounds were identified and determined along with neutral OCs in blood plasma of polar bears collected in 1997 from the Resolute Bay area and from Svalbard (Sandau, 2000; Sandau et al., 2000). In the phenolic fraction, 35 CPCs were identified as HO-PCBs in the two populations. In addition to HO-PCBs, a previously unidentified metabolite of octachlorostyrene (OCS), 4-hydroxyheptachlorostyrene (4-OH-HpCS) was identified. Traces of pentachlorophenol (PCP) at about 0.2 ng/g were also

found. The rank order concentrations of the first 50 individual OCs, both neutral and phenolic, were determined in the combined Resolute Bay/Svalbard data set. Out of the 10 most abundant OCs in plasma, 6 were phenolic compounds, 3 PCBs, and 1 oxychlordane. These 6 phenolic compounds, including 4-HO-HpCS, constituted 42% by weight of all OCs in plasma. Of the remaining 40 compounds, 9 were chlordane-related compounds, 17 were PCBs, 5 HO-PCBs, and the rest were chlorobenzenes, DDTs, HCHs and PCP.

Mean concentrations of ∑HO-PCB in polar bear plasma collected in 1997 ranged from 57 ng/g in males from Resolute Bay to 218 ng/g in females from Svalbard (Sandau, 2000). Females had significantly higher concentrations of ∑HO-PCB than males. The ratio of ∑HO-PCB/∑PCB in plasma was also significantly higher in females (mean 4.08) than males (mean 1.49). It appears that females either have a higher binding capacity for HO-PCBs in plasma than males, probably due to higher TTR (transthyretin) concentrations, or a higher capacity to form HO-PCBs. The concentration of ∑HO-PCB in subadults was the same as that in females. ∑HO-PCB concentration was 2–3 times higher than any other residue class in female plasma, and in other age groups and males, it was equal to or higher than the concentration of the next highest residue class, ∑PCB. Thus, it appears that there is no selective transfer of HO-PCBs in polar bear milk, unlike MeSO<sub>2</sub>-PCBs (Letcher, 1996).

Recent investigations of 4-OH-HpCS in the blood of other Canadian Arctic marine mammals include

Table 2

Concentrations (mean ± S.D., ww) of contaminants in Arctic marine mammal plasma (Sandala et al., 2004; Sandau et al., Carleton University, unpublished data; Letcher et al., University of Windsor, unpublished data)

| Species   | 4-HO-CB187<br>(pg/g ww) | 4-HO-HpCS<br>(pg/g ww) | OCS<br>(pg/g ww) | CB153<br>(pg/g ww) | 4-HO-HpCS:OCS |
|---|-------------------------|------------------------|------------------|--------------------|---------------|
| Walrus ( <i>n</i> =9)   | 66.9 ± 113              | 15.7 ± 14.1            | 216 ± 144        | 662 ± 898          | 0.08 ± 0.08   |
| Ringed seal ( <i>n</i> =5)  | 17.1 ± 12.8             | 45.1 ± 18.4            | 293 ± 95.3       | 568 ± 409          | 0.17 ± 0.09   |
| Beluga whale ( <i>n</i> =6)                                       | 8.60 ± 11.7             | 1.47 ± 0.67            | 31.6 ± 13.9      | 2910 ± 1530        | 0.05 ± 0.02   |
|   | (ng/g ww)               | (ng/g ww)              | (ng/g ww)        | (ng/g ww)          |               |
| Polar Bear (Svalbard, <i>n</i> =32)                               | 54.2 ± 67.2             | 7.34 ± 4.18            | 0.493 ± 0.314    | 31.3 ± 19.9        | 20.1 ± 16.3   |
| Polar Bear (Barrow Str, <i>n</i> =24)                             | 25.5 ± 14.9             | 7.82 ± 3.60            | 0.342 ± 0.195    | 14.7 ± 8.71        | 31.0 ± 27.9   |
| Polar bear <sup>a</sup> (East Greenland,<br><i>n</i> =12 females) | 31.8 ± 17.1             | 7.71 ± 3.33            | 0.15 ± 0.18      | 12.4 ± 8.4         | 93.6 ± 59.1   |
| Polar bear <sup>a</sup> (East Greenland,<br><i>n</i> =7 males)    | 36.5 ± 11.8             | 8.17 ± 2.41            | 0.08 ± 0.02      | 10.6 ± 5.6         | 114.5 ± 35.4  |

<sup>a</sup> Whole blood.

beluga, walrus and ringed seal (Sandau et al., Carleton University, unpublished data; Letcher et al., University of Windsor, unpublished data). Concentrations of CB153, 4-HO-CB187 (the main HO-PCB metabolite in polar bears), OCS and 4-OH-HpCS in Arctic marine mammals are given in Table 2. The concentration of 4-HO-HpCS was 20–30 times higher than OCS in the plasma of polar bear from Barrow Strait, which is consistent with the metabolic capability of polar bears for a wide range of PCBs and OCs. The other marine mammal species from the Canadian Arctic have much lower concentrations of the metabolite in their blood, which may be indicative of lower metabolic capacity for OCS, and subsequent 4-HO-HpCS formation.

#### 4. Arctic marine food web studies

A number of studies have been carried out on the food web transfer of OCs and Hg in marine ecosystems since the first CACAR. These studies encompassed a larger number of species and trophic levels than was previously available for single Arctic marine food webs, and also incorporated the measurement of  $\delta^{15}\text{N}$  to discern trophic position. These recent studies provide an advantage over food web relationships for contaminants developed for the original CACAR assessment in that all samples were collected at the same time and in the same region, and the analytical methods were consistent for all samples. A case study assessing a local PCB-contaminated marine food chain near a former military radar site at Saglek Bay in northern Labrador is discussed elsewhere in this issue (Kuzyk et al., 2005).

##### 4.1. Trophic transfer of mercury in marine food webs

###### 4.1.1. Mercury in the Resolute Bay food web

Atwell et al. (1998) examined the concentration of total Hg in tissues from 27 species from the Arctic marine food web of Lancaster Sound, Nunavut. Samples included particulate organic matter as well as a wide range of species from invertebrates through to polar bears.  $\delta^{15}\text{N}$  values showed that total Hg in muscle tissue biomagnified in this food web. Polar bears were a notable exception, having a lower mean Hg concentration than their main prey, ringed seals. This is likely due to the fact that the bears tend to eat

only the blubber of seals, which is lower in Hg concentrations than muscle or liver (Atwell et al., 1998). Most vertebrates showed greater variance in muscle Hg content than whole body Hg content of invertebrates, and there was a trend in seabirds toward increased variability with trophic position.

###### 4.1.2. Mercury in the food web of northern Baffin Bay

Total Hg and MeHg concentrations and  $\delta^{15}\text{N}$  values were determined in mixed zooplankton, *C. hyperboreus* (a herbaceous copepod), eight species of seabirds, and ringed seals from the North Water (NOW) Polynya to examine the dynamics of Hg in an Arctic marine food web (Campbell et al., this issue). The NOW Polynya is located in northern Baffin Bay and is the largest and most productive polynya in the Canadian Arctic. All samples were collected in 1998. Concentrations of total Hg and MeHg were positively correlated with trophic level ( $r^2 > 0.85$ ), based on  $\delta^{15}\text{N}$ , demonstrating the occurrence of biomagnification (Fig. 11).

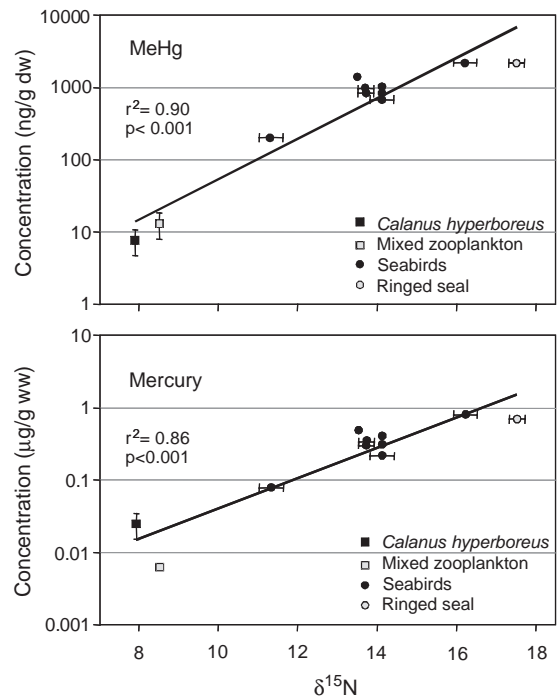


Fig. 11. Relationships between total and methylmercury concentrations and  $\delta^{15}\text{N}$  in the marine food web of northern Baffin Bay. Mercury and  $\delta^{15}\text{N}$  determined in muscle tissue of seabirds and ringed seals and whole body for zooplankton. Each point is the mean  $\pm 1$  S.E.

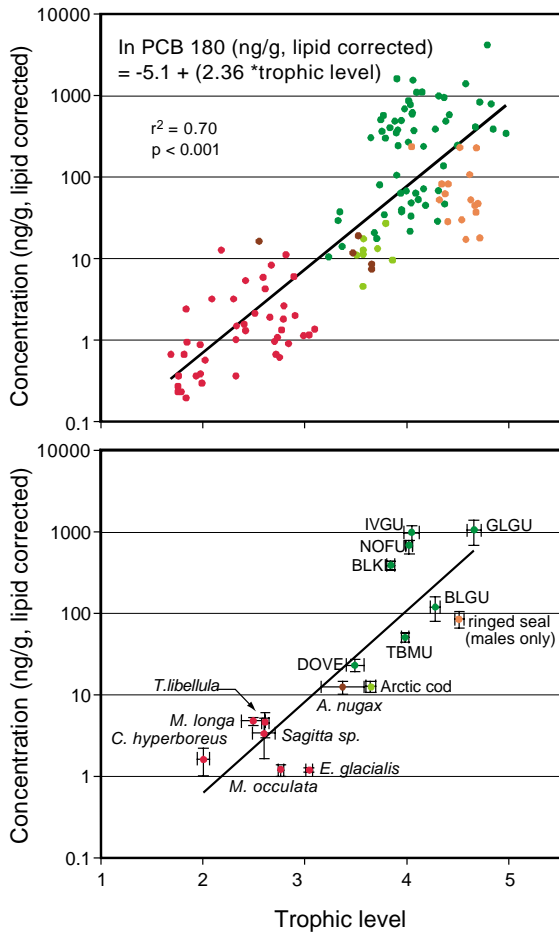


Fig. 12. PCB 180 concentration (ng/g, lipid-corrected)–trophic level relationships for the North Water (NOW) Polynya marine food web (Fisk et al., 2001a). The top graph contains all data points and the bottom graph contains mean ( $\pm$  S.E.) values for each species. Lines are linear regression. Trophic level based on  $\delta^{15}\text{N}$ . Symbols: Red circle—pelagic zooplankton; brown circle—benthic amphipods; light green circle—Arctic cod; orange circle—ringed seals; dark green circle—seabirds.

Atwell et al. (1998) reported a slope of 0.2 for the log Hg concentration– $\delta^{15}\text{N}$  regression for the Resolute Bay food web, which was close to the slope of 0.197 found in the NOW study and many other aquatic food webs (Campbell et al., this issue). These results show that mercury and  $\delta^{15}\text{N}$  track proteins, and the rate of trophic transfer of Hg will follow protein turnover. The log MeHg concentration– $\delta^{15}\text{N}$  relationship slope of 0.223 suggests that MeHg accounts for a large percentage of the biomagnification of Hg.

#### 4.2. Trophic transfer of organochlorines in marine food webs

In a companion study to that of Hg in the NOW Polynya, OC concentrations were also measured in zooplankton (six species), a benthic invertebrate (*A. nugax*), Arctic cod, seabirds (six species) and ringed seals collected in 1998 (Fisk et al., 2001a; Moisey et al., 2001). These samples were the same or similar to those used in the analysis of Hg described in Section 4.1.2 but include more species and samples. The trophic relationships derived from  $\delta^{15}\text{N}$  analysis for the NOW food web agreed with those determined in another Arctic polynya food web (Hobson et al., 1995), with seabirds and ringed seal at the top and zooplankton species occupying lower trophic levels.

Strong positive relationships were found between recalcitrant OC concentrations (lipid-corrected) and trophic level based on  $\delta^{15}\text{N}$  values, providing clear evidence of OC biomagnification (Fig. 12; Table 3). Trophic magnification factors (TMFs), determined from the exponential of the slope of  $\ln[\text{OC}]$ –trophic

Table 3  
Biomagnification factors (BMFs) and trophic magnification factors (TMFs) determined for key fauna in the marine food web of northern Baffin Bay (Fisk et al., 2001a)

| Predator/prey          | HCB  | $\sum\text{HCH}$ | $\sum\text{CHL}$ | <i>p,p'</i> -DDE | $\sum\text{PCB}$ |
|------------------------|------|------------------|------------------|------------------|------------------|
| Amphipod/copepod       | 3.8  | 4.5              | 26.5             | 16               | 4.6              |
| Arctic cod/amphipod    | 6.1  | 1.1              | 1.6              | 3.1              | 0.9              |
| <i>Seabirds</i>        |      |                  |                  |                  |                  |
| TBMU/Arctic cod        | 10.9 | 2.1              | 1.8              | 19               | 8.2              |
| BLGU/Arctic cod        | 5.0  | 3.5              | 4.0              | 18.5             | 8.9              |
| BLKI/Arctic cod        | 21.6 | 4.2              | 11.6             | 56               | 60.5             |
| GLGU/Arctic cod        | 6.7  | 5.2              | 80               | 49               | 28               |
| <i>Marine mammals</i>  |      |                  |                  |                  |                  |
| Ringed seal/Arctic cod | 0.5  | 2.0              | 2.4              | 7.0              | 5.5              |
| TMF                    | 4.1  | 2.7              | 7.0              | 14               | 4.6              |

BMFs based on lipid-corrected concentrations, corrected to one full trophic level based on trophic levels derived from stable nitrogen isotopes. TBMU—thick-billed murre, BLGU—black guillemot, BLKI—black-legged kittiwake, GLGU—glaucous gull.



level relationships, in this work are in good agreement with values obtained for food webs from temperate and Arctic ecosystems involving marine birds and mammals (Norstrom, 1994; Jarman et al., 1996).

Biomagnification factors (BMFs = lipid-adjusted concentration in predator/lipid-adjusted concentration in prey) values for the NOW food web are summarized in Table 3. BMFs were corrected for trophic level differences based on  $\delta^{15}\text{N}$  because many of these species had varied diets, and so for many of these comparisons the predator was not a full trophic level above the prey according to  $\delta^{15}\text{N}$  values (Fisk et al., 2001a). Generally, PCBs and *p,p'*-DDE had the highest BMFs, followed by the chlordanes ( $\sum\text{CHL}$ ) and hexachlorobenzene (HCB). Compared to other compound classes, the BMFs for hexachlorocyclohexanes ( $\sum\text{HCH}$ ) were low, reflecting their lower  $K_{ow}$  and greater susceptibility to biotransformation (Moisey et al., 2001). BMFs determined for amphipods that consumed copepods were very high, but Fisk et al. (2001a) suggested that these BMFs were not realistic because concentrations of OCs in zooplankton may be controlled by OC concentrations in water and not prey. Rather, large body size may be driving the higher concentrations found in the amphipods (Borgå et al., 2004). BMFs for Arctic cod/amphipod were slightly below and above one but are in the range of BMFs reported for similar-sized fish in laboratory experiments (Fisk et al., 1998) and field studies (Rasmussen et al., 1990). BMFs calculated for NOW ringed seals (Table 3) were slightly lower but within the range of those reported for male ringed seals from the east central Canadian Arctic (Muir et al., 1988). The BMFs of the seabirds appeared to be related to whether or not they scavenged on marine mammals and/or migrated to more contaminated regions. The highest BMFs were found in scavenging seabirds (glaucous gulls, ivory gulls and northern fulmars) and those that migrate to more contaminated southern regions (black-legged kittiwakes and glaucous gulls).

One of the most striking differences in BMFs was between poikilotherms (fish) and homeotherms (seabirds and mammals) (Table 3). The large differences in BMFs between poikilotherms and homeotherms of similar size and eating the same prey was first demonstrated in herring gulls (*Larus argentatus*) and salmon from Lake Ontario (Braune and Norstrom, 1989). Greater BMFs, and hence exposure to OCs, in home-

otherms has been attributed to their greater energy requirements and feeding rates (Braune and Norstrom, 1989; Fisk et al., 2001a).

A strong relationship between trophic magnification factors (TMFs), determined from concentration–trophic level relationships, and  $\log K_{ow}$  was found for recalcitrant OCs in the NOW food web (Fig. 13). It is clear that increasing  $\log K_{ow}$  is associated with greater trophic transfer of recalcitrant OCs. This relationship provides insight into the behavior of a number of OCs. For example, DDE and heptachlor epoxide have TMFs that are much greater than predicted based on the TMF– $\log K_{ow}$  relationships. DDE has been well established as a metabolite of DDT formed in animals. These results suggest that a large percentage of the high concentrations of DDE in upper trophic-level Arctic organisms are due to *in vivo* metabolic formation. Heptachlor epoxide, which is not found in technical mixtures, can be formed from heptachlor by photo-oxidation and has been found in rat livers exposed to heptachlor only (Buser and Müller, 1993). Fisk et al. (2001a) suggested that heptachlor epoxide was formed in upper trophic-level Arctic organisms, and may account for a large percentage of its concentration.

BMFs of  $\sum\text{OC}$  groups vary considerably from those calculated for individual compounds of these groups (Fisk et al., 2001a). This is due to a combination of differential susceptibility to biotransformation and variation in kinetics due to different physical–chemical properties. For example,  $\sum\text{HCH}$  was observed to biomagnify in seabirds and throughout the entire food web although neither  $\alpha$ - nor  $\gamma$ -HCH were found to biomagnify; the biomagnification was driven mainly by  $\beta$ -HCH (Moisey et al., 2001). These results suggest that caution should be exercised when interpreting patterns of  $\sum\text{OC}$  groups across food webs.

#### 4.3. Trophic transfer of “new” and chiral chemicals in marine food webs

Concentrations of four possibly naturally produced organohalogenes (collectively termed HDBPs)–1,1'-dimethyl-3,3',4-tribromo-4,5,5'-trichloro-2,2'-bipyrrole (DBP-Br<sub>3</sub>Cl<sub>3</sub>), 1,1'-dimethyl-3,3',4,4'-tetrabromo-5,5'-dichloro-2,2'-bipyrrole (DBP-Br<sub>4</sub>Cl<sub>2</sub>), 1,1'-dimethyl-3,3',4,4',5-pentabromo-5'-chloro-2,2'-

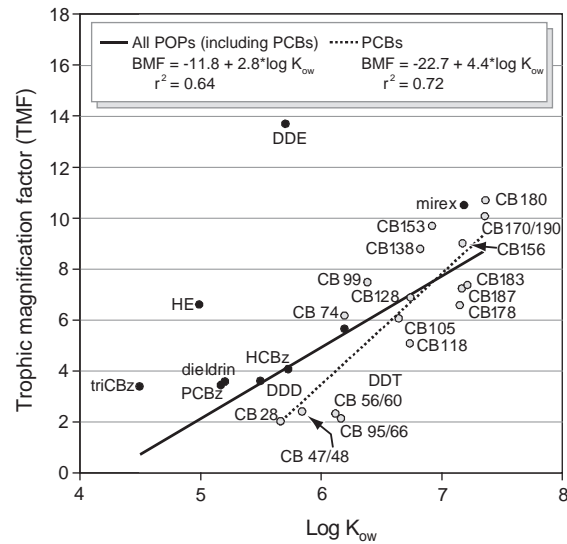


Fig. 13. Relationship between trophic magnification factors (TMF) and log  $K_{ow}$  of recalcitrant POPs in the North Water (NOW) Polynya marine food web (from Fisk et al., 2001a). TMFs were determined from the relationship between concentration (lipid-corrected) and trophic level.

bipyrrole (DBP-Br<sub>5</sub>Cl) and 1,1'-dimethyl-3,3',4,4',5,5'-hexabromo-2,2'-bipyrrole (DBP-Br<sub>6</sub>)—were determined in the NOW food web of Baffin Bay (Tittemier et al., 2002). All HDBP congeners were found to significantly biomagnify in the invertebrate–fish–seabird food chain. None of the four HDBP congeners in ringed seals agreed with this general trend, which was likely due to the ability of the seals to metabolize HDBPs.

Chiral contaminants have the potential to identify differences in species biotransformation ability and provide insights into the fate of OCs in food webs (see Sections 3.2.4 and 3.3.3.6). Concentrations of HCH isomers ( $\alpha$ ,  $\beta$  and  $\gamma$ ) and enantiomer fractions (EFs) of  $\alpha$ -HCH were also determined in the NOW food web (Moisey et al., 2001). For invertebrates and fish, the BMFs of the three isomers were  $>1$  and the proportion of each isomer and the EFs of  $\alpha$ -HCH were similar to water, suggesting minimal biotransformation. Seabirds appeared to readily metabolize  $\gamma$ - and  $\alpha$ -HCH, based on low BMFs for these isomers, high proportions of  $\beta$ -HCH (62–96%), and high EFs (0.65 to 0.97) for  $\alpha$ -HCH. The  $\alpha$ - and  $\beta$ -HCH isomers appear to be recalcitrant in ringed seals, based on BMFs  $>1$  and near racemic EFs for  $\alpha$ -HCH. The  $\beta$ -HCH isomer appeared to be recalcitrant in all species examined, and had an overall food web magnification

factor of 3.9. EFs of  $\alpha$ -HCH provided conclusive evidence that biotransformation accounted for much of the HCH isomer patterns observed in the NOW food web.

#### 4.4. Trophic transfer of organochlorine metabolites in marine food webs

With respect to Arctic food webs, there is only one published study on MeSO<sub>2</sub>-PCB and -4,4'-DDE metabolites relative to precursor PCBs and 4,4'-DDE. The Arctic cod–ringed seal–polar bear food chain in Resolute Bay was assessed to determine the relative importance of metabolite bioaccumulation from the food chain, versus metabolic formation from bioaccumulated precursor contaminants (Letcher, 1996; Norstrom, 1997; Letcher et al., 1998; Wiberg et al., 1998a). Overall, there was little difference in the PCB pattern in cod relative to the Aroclor standard (1242:1254:1260, 1:1:1); however, the PCB patterns changed noticeably in seal and bear, especially PCB congeners possessing hydrogen at the *meta-para* (3,4) position on one or more ring. *Meta-para* PCBs were present in seal blubber but notably absent in polar bear ( $<0.05$  ng/g, lw). These PCBs included CB31, CB49, CB64, CB70, CB91, CB101, CB110, CB141, CB132 and CB174, all of which were also

present in the form of their 3- and 4-MeSO<sub>2</sub>-PCB metabolites in seals and bears. Metabolites of CB52 and CB95 were not found in the bears. These PCB congeners exhibit hydrogen at both *meta-para* positions, and may be metabolized to bis-MeSO<sub>2</sub>-PCBs or to HO-PCBs. The results of this study demonstrated that both bioaccumulation of and precursor biotransformation to MeSO<sub>2</sub>-PCBs and -DDEs were potentially important in a food web where at least one species has the capacity to metabolically generate the metabolites.

Patterns of accumulation of CPCs and CPC metabolites and their precursors were studied in ringed seal and compared to those in polar bear to determine the potential for bioaccumulation of HO-PCBs and 4-HO-HpCS (Sandau, 2000; Sandau et al., 2000). Concentrations of  $\sum$ HO-PCB were 1000 times lower in ringed seal than in polar bear plasma, whereas  $\sum$ PCB was only 2 times lower. Considering that most of the HO-PCB metabolites in ringed seal were probably in blood and at low concentrations, it seems likely that the potential BAF and BMF values from ringed seal to polar bear were very low for HO-PCBs and 4-HO-HpCS.

## 5. Temporal trends of contaminants in biota

One of the major data gaps identified in the first CACAR assessment was the lack of temporal trend data sets for both metals and OCs (Muir et al., 1997). An exception was the data set for OCs in Holman Island ringed seals that spanned 1972 to 1991 (Addison and Smith, 1998). The number of sampling years was limited, however, and comparison of recent data with data from the 1970s was problematical because of changes in analytical methodology.

Two types of temporal trend data—short-term and long-term—are presented below, and the distinction between the two parallels important differences between Hg and POPs concerning sources in the environment and regulatory information needs. Virtually all POPs of toxicological significance are entirely industrial in origin and most have existed for no more than about 50 years. Thus, short-term data (i.e., covering the past few decades) are the only data of relevance for assessment and policy objectives related to POPs. For metals, in addition to establishing

the short-term trends, long-term trend data are also required. Metals in the environment and biota may come from both natural and anthropogenic sources, and distinguishing between these sources in remote regions such as the Arctic is especially difficult using geographic trend data alone. Estimating the anthropogenic component of Hg in present-day Arctic biota requires a long-term perspective, which can be gained by establishing the pre-industrial background levels. Only then can the current degree of Hg pollution in biota be estimated and subsequent regulatory actions be based on this knowledge.

The potential regulatory uses and knowledge needs for short-term data are similar for both POPs and Hg. Bans on the use of many of the legacy OCs, e.g., PCBs and DDT, have been in place in North America and western Europe since the 1970s but only recently have agreements been signed that will completely eliminate the use of these chemicals throughout the world. Releases of Hg, through coal combustion and other industrial and mining activities, continue to be a major concern. The continued emission of legacy chemicals from use areas (e.g., toxaphene and DDT in agricultural soils; PCBs from buildings, waste disposal sites) in temperate regions, their transport in air and seawater, and the slower rate of transformation of organic chemicals in the Arctic, make temporal trends and predictions about future levels of OCs and metals in biota an important issue. Wildlife, and in particular higher trophic level organisms that tend to have high levels of POPs and Hg, are a significant component of the Inuit diet, making temporal trend data sets particularly important information at the community level. As well, the discovery of new chemicals, some of which have been rapidly increasing in the Arctic in recent years (e.g., PBDEs), demonstrates the value of temporal trend data sets and particularly of archiving samples for future research.

One of the major accomplishments of the Northern Contaminants Program since the first CACAR assessment has been initiating or continuing a number of temporal trend studies. Because of the short time span of these studies, however, in most cases they presently have only limited ability to statistically validate observed trends (see Bignert et al., 2004). Other studies have used archived and archeological samples to generate new data sets that encompass a longer period of time.

### 5.1. Temporal trends of mercury in invertebrates

Long-term Hg time-series data for Arctic bivalves have recently been generated using sequences of shells collected from Holocene (1000–9000 years before present) beach ridges and as living modern specimens (Outridge et al., 2000). Significant north–south variations in the relative inputs of anthropogenic and natural Hg in bivalve species at sites across the Canadian Arctic were reported. At two high-latitude sites, Expedition Fiord on Axel Heiberg Island, and Resolute Bay on Cornwallis Island, Holocene shells contained similar concentrations of Hg to modern (1980s–1990s) specimens, suggesting an absence of industrial inputs in those areas. At a low-latitude site in southeast Hudson Bay, however, concentrations of Hg were significantly higher in modern (1970s) shells of blue mussels (*M. edulis*).

### 5.2. Temporal trends of contaminants in seabirds

Retrospective analyses of OCs, Hg as well as new chemicals (e.g., PBDEs) in archived Arctic seabird eggs from Prince Leopold Island in the Canadian High Arctic (between 1975 and 2003) have provided one of the most comprehensive contaminant temporal trend data sets available for the Canadian Arctic (Braune et al., 2001; Braune, 2003; Braune, Canadian Wildlife Service, unpublished data). At the time of

egg formation in birds, OCs are transferred along with fat to the eggs (Mineau et al., 1984). Contaminant burdens in the egg reflect residues assimilated over a long time period by the female and, particularly in migratory species, may integrate exposure from a number of different locations (Hebert, 1998; Monteiro et al., 1999). Eggs from three species of seabirds, thick-billed murres, northern fulmars and black-legged kittiwakes, have been used for this work.

#### 5.2.1. Temporal trends of organochlorines in seabird eggs

With the exception of  $\sum$ HCH, OCs in Arctic seabird eggs have shown declines or, in some cases, no significant change in levels between 1975 and 1998 (Braune et al., 2001). Levels of  $\sum$ PCB and  $\sum$ DDT as well as  $\sum$ CBz decreased significantly in eggs of all three species while  $\sum$ CHL, dieldrin and mirex levels decreased in kittiwake eggs only. Recent data from 2003 indicate a continuation of these trends (Fig. 14; Braune, Canadian Wildlife Service, unpublished data). Kittiwakes, whose migration pattern would have historically brought them into closer contact with industrial sources of contaminants such as PCBs in more temperate latitudes, showed the most dramatic declines through to 2003. The significant declines in concentrations of  $\sum$ PCB and  $\sum$ DDT in this study have also been observed in seabirds from other areas including the Baltic Sea (Olsson and Reu-

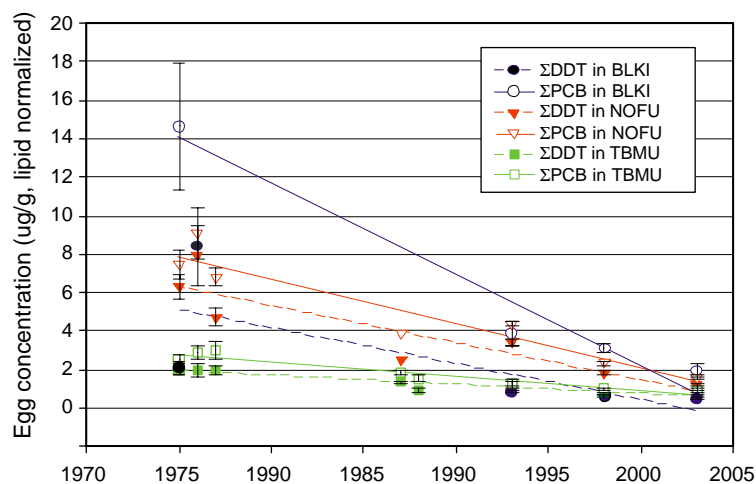


Fig. 14. Mean concentrations ( $\pm 1$  S.E.) of  $\sum$ DDT and  $\sum$ PCB in seabird eggs collected between 1975 and 2003 from Prince Leopold Island (Braune et al., 2001; Braune, Canadian Wildlife Service, unpublished data). BLKI=black-legged kittiwake, NOFU=northern fulmar and TBMU=thick-billed murre.

tergårdh, 1986; Andersson et al., 1988; Bignert et al., 1995), the Barents Sea (Barrett et al., 1996), and the Great Lakes (Hebert et al., 1997). The only OC in this study for which a significant increase in concentrations was seen was for  $\sum$ HCH, particularly for  $\beta$ -HCH in murre and fulmars.

Concentrations of  $\sum$ PCDD and  $\sum$ PCDF decreased in eggs of northern fulmars collected between 1975 and 1998 from Prince Leopold Island (Lancaster Sound) whereas in thick-billed murre, the pattern was less clear due mainly to a slight increase in concentrations in 1993 (Braune, 2003). Concentrations of total non-ortho PCBs also decreased in both the fulmars and the murre between 1975 and 1998 (Braune, 2003). Stable nitrogen isotope analyses indicate that the temporal trends observed in OC concentrations in seabird eggs were not the result of shifts in trophic level over time (Braune et al., 2001). More likely, the trends reflect changes in contaminant deposition into the various marine environments that these birds occupy throughout the year as well as the toxicokinetics of each contaminant as it is transported through the food chain.

### 5.2.2. Temporal trends of new chemicals in seabird eggs

Mean concentrations of total (Br<sub>3</sub>–Br<sub>7</sub>)-PBDEs ( $\sum$ PBDE) in eggs of northern fulmars and thick-billed murre from Prince Leopold Island (near Lancaster Sound) increased 9.1- and 4.4-fold, respectively, between 1975 and 1998 (Fig. 15). Increasing

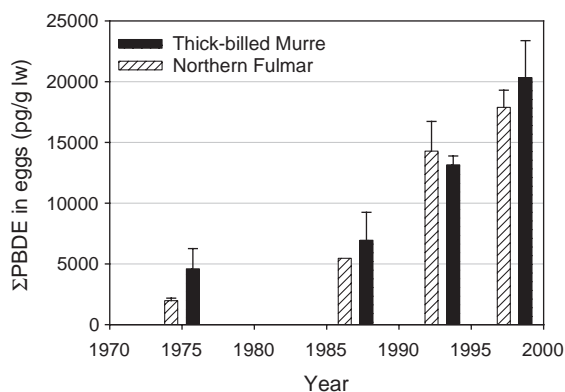


Fig. 15. Mean concentrations ( $\pm$  1 S.E.) of total PBDEs (pg/g lw) in eggs of northern fulmars and thick-billed murre collected between 1975 and 1998 from Prince Leopold Island.

concentrations of PBDEs have also been found in black guillemot eggs from Sweden (Sellström et al., 1993) and herring gull eggs from the Great Lakes (Norstrom et al., 2002). Total PBDE concentrations in fulmars and murre from the Canadian Arctic were several orders of magnitude lower (maximum of 2.9 ng/g ww in thick-billed murre eggs in 1998) than concentrations found in Great Lakes herring gull eggs (up to 1400 ng/g ww in 2000).

### 5.2.3. Temporal trends of mercury in seabird eggs

Total Hg concentrations almost doubled between 1975 and 1998 in eggs of thick-billed murre, and a 50% increase in levels was observed in northern fulmars (Braune et al., 2001). Recent data from 2003 indicate a continuation of this trend (Fig. 16; Braune, Canadian Wildlife Service, unpublished data). Data for marine mammals (Wagemann et al., 1996), seabird feathers (Thompson et al., 1992) and lake sediments (e.g., Lockhart et al., 1995) also indicate that Hg levels have increased in the North over the last few decades. The rate of increase of total Hg in black-legged kittiwake eggs appears to be less than in the other two species. This may be partially due to the fact that kittiwakes overwinter at more temperate latitudes whereas the murre and fulmars overwinter in northern waters.

### 5.3. Temporal trends of contaminants in marine mammals

#### 5.3.1. Temporal trends of organochlorines

5.3.1.1. Ringed seals. Temporal trends of PCBs, DDT,  $\beta$ -HCH and  $\gamma$ -HCH in ringed seals in the Canadian Arctic have been studied at the communities of Ikpiarjuk (Arctic Bay), Ausuittuq (Grise Fiord), and Holman from the early 1970s to the late 1990s or 2000/2001. Here we present a brief overview of the detailed new information presented in Addison et al. (2005) and Muir and Kwan (2003) together with the earlier studies at these sites (Addison et al., 1986; Muir et al., 1988; Weis and Muir, 1997; Addison and Smith, 1998; Letcher et al., 1998; Addison et al., 2000; Wiberg et al., 2000; Muir et al., 2001). Mean concentrations in female ringed seals of  $\sum$ DDT,  $\sum$ PCB,  $\sum$ HCH and  $\sum$ CHL, along with ratios of recalcitrant members of each class, are



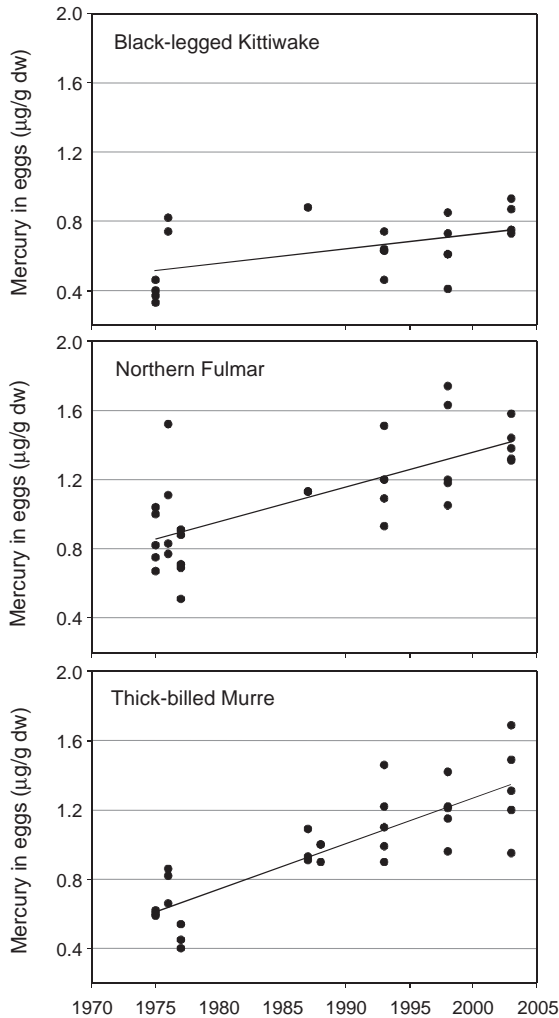


Fig. 16. Concentrations of total mercury in seabird eggs collected between 1975 and 2003 from Prince Leopold Island (Braune et al., 2001; Braune, Canadian Wildlife Service, unpublished data). Each point represents a pool of three eggs.

shown in Figs. 17, 18 and 19 for Ikpiarjuk, Ausuittuq and Holman, respectively. At Ikpiarjuk and Ausuittuq,  $\sum\text{PCB}_{10}$  (sum of 10 congeners) was used as the basis for comparison over time, whereas at Holman,  $\sum\text{PCB}$  was based on the conversion of Aroclor 1254 to a  $\sum\text{PCB}$  value consisting of 20 major congeners.  $\sum\text{PCB}$  in seals declined significantly at all three sites over the past three decades: at Ikpiarjuk by 2.4 times, at Ausuittuq by 1.5 times, and at Holman by 5.5 times, based on arithmetic means. At Holman, the

bulk of the decline occurred between 1972 and 1981, with no further decline between 1981 and 1991 but another significant decline between 1991 and 2001 (Fig. 17). PCBs were being phased out in North America and northern Europe from about the mid-1970s onwards. More recent analyses (Addison et al., 2005) showed that there was no significant decline in total non-ortho-, mono-ortho- and di-ortho-substituted-PCBs (NO, MO, DO-PCBs) between 1981, 1991, 1996 and 2000, although there was a shift in PCB congener distribution over the interval which was consistent with atmospheric transport processes becoming increasingly important in the introduction of PCBs to the Arctic in recent years.

$\sum\text{DDT}$  declined significantly in female ringed seals from all three locations between the early/mid-1970s and the late 1990s/2000.  $\sum\text{DDT}$  exhibited the largest decline of any “legacy” OC—2.5 times at Ausuittuq and 3.3 times at Ikpiarjuk and Holman over 25–30 years. Significant increases of the  $p,p'$ -DDE/ $\sum\text{DDT}$  ratio were found at all three locations, reflecting the shift from fresh DDT to “weathered” or degraded older sources. Between 1972 and 1982, the  $p,p'$ -DDE/ $\sum\text{DDT}$  ratio at Holman declined slightly, even though total DDT levels fell by about 40% (Fig. 19). This was interpreted as indicating a continuing minor input into the western Arctic of “new” DDT, possibly by atmospheric transport from the Far East where DDT was still being used, combined with cessation of use in North America. After 1981, the decline of total DDT at Holman slowed, and  $p,p'$ -DDE/ $\sum\text{DDT}$  increased, indicating that negligible fresh DDT entered the region (Addison and Smith, 1998).

$\sum\text{HCH}$  concentrations showed no significant changes in concentrations from the 1970s to 2001 (1981 in the case of Holman). However,  $\beta\text{HCH}$  as a fraction of  $\sum\text{HCH}$  increased (4-fold at Ikpiarjuk). This shift in the composition of HCH to higher proportions of  $\beta\text{HCH}$  has also been reported in seawater in the western Canadian Arctic during the 1980s and 1990s (Li et al., 2002).

$\sum\text{CHL}$  in female seals showed quite a different trend from  $\sum\text{DDT}$  and differences between sites, with increasing concentrations at Holman and Ausuittuq, and a slow decline (2.1 times in 25 years) at Ikpiarjuk. Proportions of oxychlordan, a recalcitrant metabolite of chlordane in mammals, increased at all three locations.

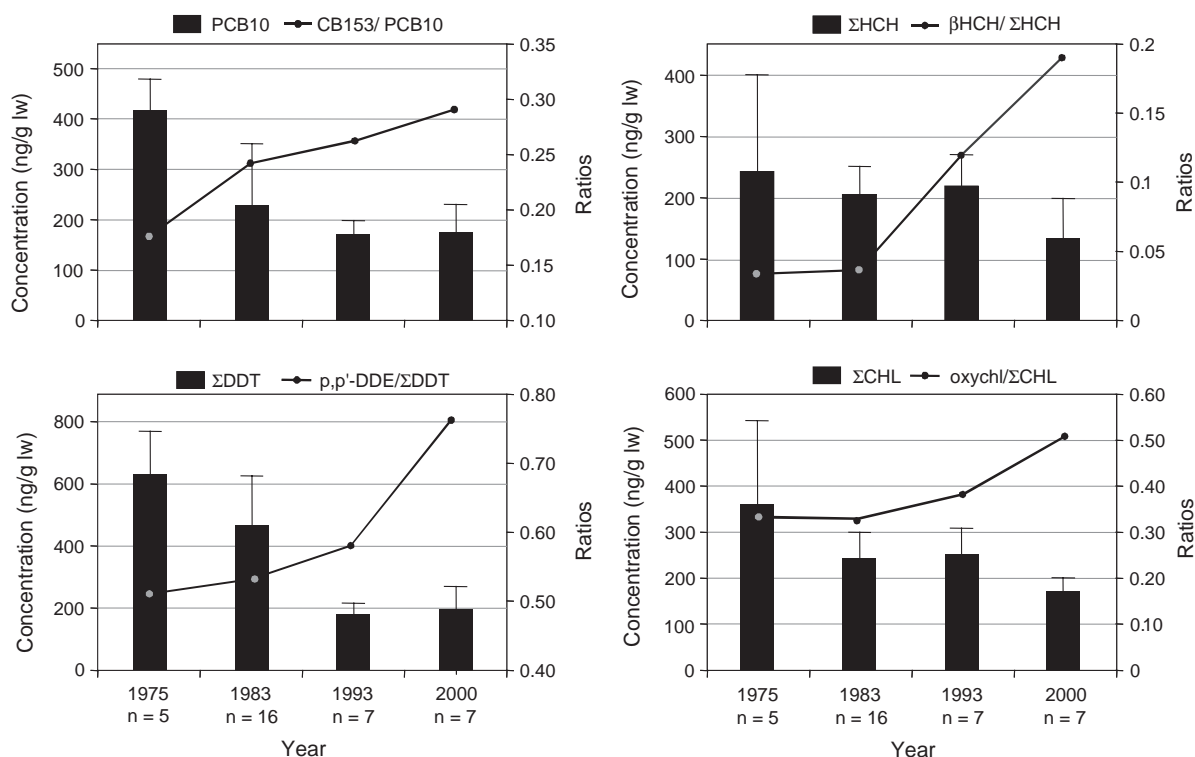


Fig. 17. Trends in concentrations and proportions of major components in blubber of female ringed seals from Ikpiarjuk (Arctic Bay). Concentrations are presented as arithmetic means and 95% confidence intervals. Significant differences between 1975 and 1993 were found for  $\sum$ PCB10 and  $\sum$ DDT.

Ikonomou et al. (2002) examined temporal trends of MO+NO PCBs and PCDD/F in male ringed seals from Holman. Concentrations of MO+NO PCBs (149–174 ng/g) and PCDD/F (8.6–14.6 pg/g) in ringed seals aged 0 to 15 years remained approximately constant from 1981 to 2000. Total PCB concentrations did not change significantly in male Holman seals between 1981 and 1991 (Addison and Smith, 1998).

5.3.1.2. *Beluga*. Stern and Addison (1999) and Stern and Ikonomou (2003) examined temporal trends of “legacy” OCs in blubber of beluga harvested in Cumberland Sound (southeast Baffin Island) between 1982 and 2002. A significant decline of  $\alpha$ - and  $\beta$ -HCH concentrations occurred over this period, while no significant difference was observed for the  $\gamma$ -HCH isomer (Fig. 20). A number of chemicals showed greater declines in the latter

half of the 1990s than in the previous 15 years.  $\sum$ DDT concentrations, for example, did not change between 1982 and 1997, although a 2.2-fold decline and a 1.3-fold increase in the (age-adjusted) concentration of *p,p'*-DDT and its metabolite *p,p'*-DDE, respectively, were observed (Fig. 20). But from 1996 to 2002, a 33% decline occurred in age-adjusted  $\sum$ DDT concentrations (Stern and Ikonomou, 2003). These changes translated into a significant increase in the *p,p'*-DDE/ $\sum$ DDT ratio from 0.37 to 0.48 and suggests “old” rather than recent-use DDT sources. Oxychlordan, the principal metabolite of *cis*- and *trans*-chlordan, and second only to *trans*-nonachlor as the most abundant chlordan-related residue in the southeast Baffin beluga blubber, also did not change significantly from 1982 to 1996 but declined by 38% from 1996 to 2002.

Likewise, no clear trends were evident in total toxaphene and toxaphene congeners 26 and 50 from

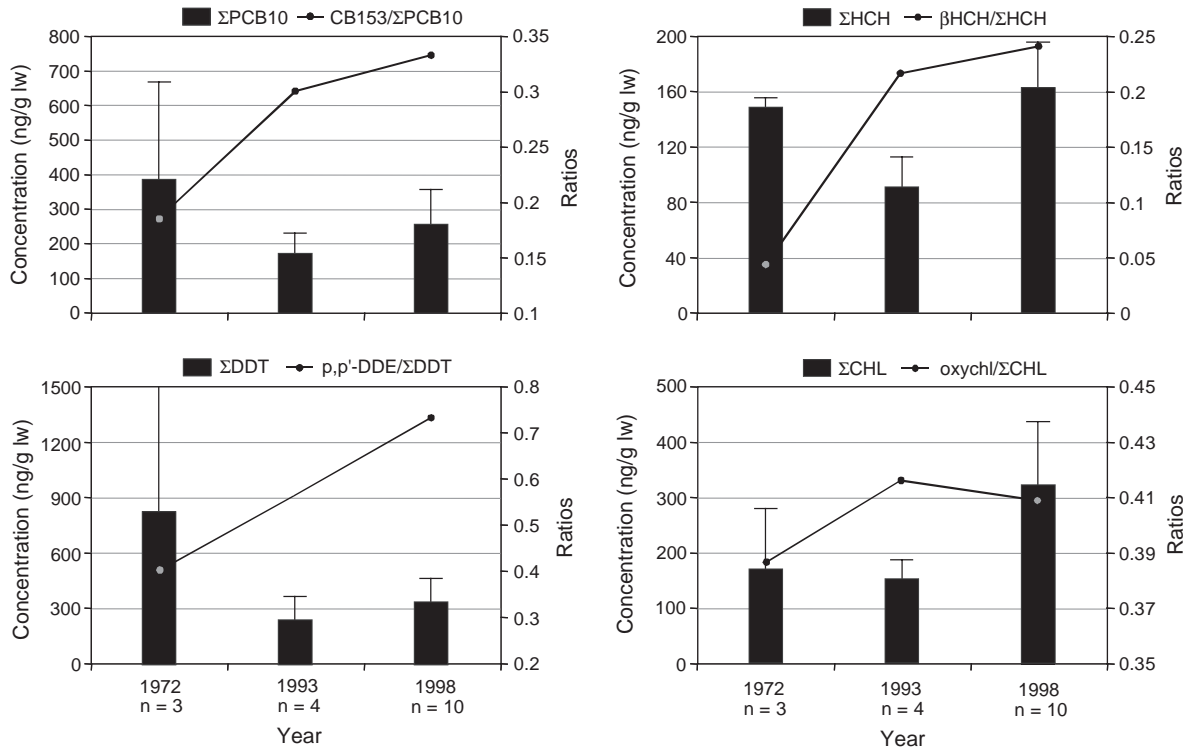


Fig. 18. Trends in concentration and proportions of major components in blubber of female ringed seals from Ausuittuq (Grise Fiord). Because of the small sample sizes from 1972 and 1993 no significant differences over time could be found. Nevertheless, the results suggest similar trends to those observed at Ikpiarjuk for concentrations and proportions of major components. Data are represented as in Fig. 17.

1982 to 1996 (Fig. 20) but more recent measurements suggest a 40% decline from 1996 to 2002 (Stern and Ikonou, 2003). A 3-fold increase in age-adjusted mean concentrations of endosulfan (sulfate) and a 2-fold decline in dieldrin were observed over the 20-year period (Stern and Ikonou, 2003). Total PCBs as well as age-adjusted concentrations of major PCB homolog groups did not show a consistent decline over the period 1982–2002 (Stern, 1999; Stern and Ikonou, 2003) (Fig. 20). For coplanar PCB congeners, significant declines ranging from 1.7-fold for CB81 to 2.8-fold for CB126, were observed. Non-ortho PCB TEQs (CB77, 126 and 169) declined from 16 to 6.1 pg/g (2.6-fold) between 1982 and 1997 (Stern and Addison, 1999).

Stern and Addison (1999) found that tetrachlorobenzene (1,2,3,4-TeCBz) increased 2-fold from 1982 to 1997. Total chlorobenzenes also increased over the period 1982 to 2002 (Stern and Ikonou, 2003).

HCB concentrations declined between 1982 and 1992 but were higher in 1996.

5.3.1.3. *Narwhal*. Results for OCs in blubber of narwhal from the Baffin Bay–Lancaster Sound region (Pond Inlet) were available from previous reports (Muir et al., 1992a) and more recently from Stern (Fisheries and Oceans Canada, unpublished data). Only combined results for 1992 and 1999 were available; therefore, only a qualitative assessment of temporal changes over the period 1982–1983 to 1999 was possible. Male narwhal were selected for temporal trend comparison because, unlike seals, they show little trend in PCB contamination with age (Muir et al., 1992a). Comparison of the combined 1992–1999 data with previous results suggests no major changes in concentrations of  $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHL or toxaphene. This lack of change in concentrations of  $\Sigma$ DDT,  $\Sigma$ CHL,  $\Sigma$ PCB and toxaphene in narwhal

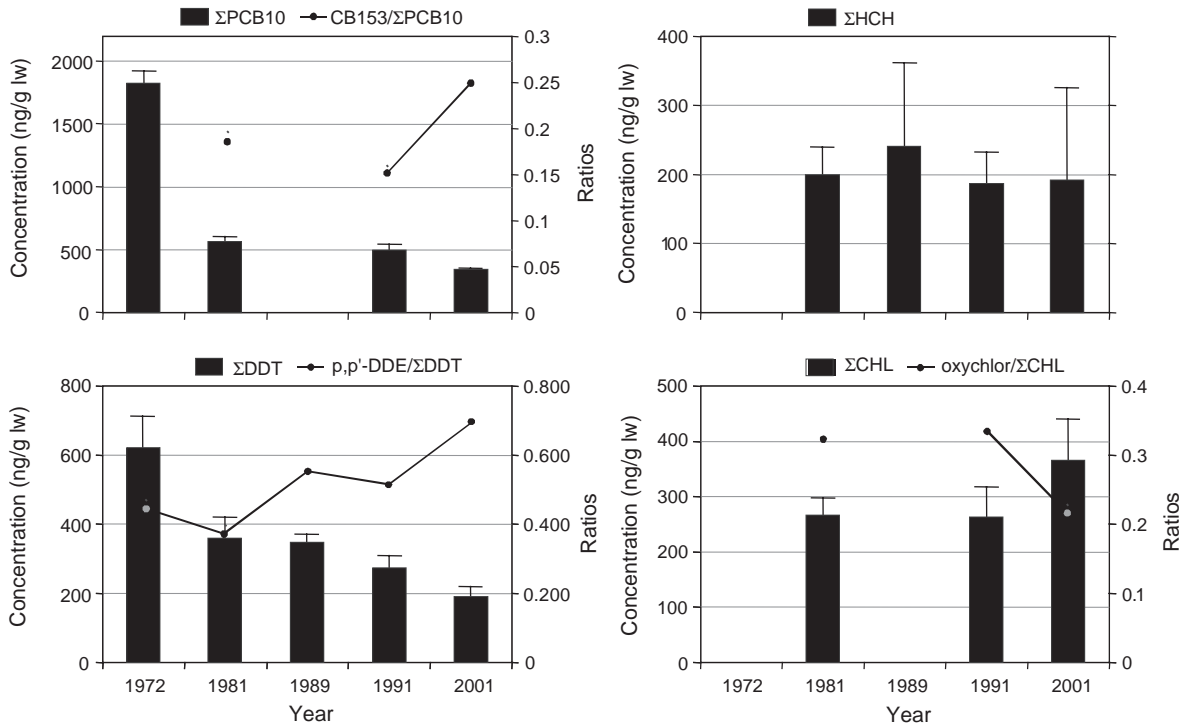


Fig. 19. Trends in concentration and proportions of major components in blubber of adult female ringed seals from Holman Island, from 1972 to 2001. Data are represented as in Fig. 17.

agrees with results for beluga in the southeastern Baffin Island area. One difference between these two odontocetes is the decline of  $\Sigma$ HCH in beluga, while no decline occurred in narwhal.

**5.3.1.4. Polar bears.** The longest temporal record of the major OC groups in polar bears anywhere in the world concerns the population near Churchill in western Hudson Bay. OC data are presented in Fig. 21 for the period 1968 to 2002 for adult females. This group was chosen for the study of temporal trends because there is no significant age–concentration effect for any OC in females, whereas there is a significant increase of highly chlorinated PCBs with age in males (Bernhoff et al., 1996; Norstrom et al., 1998). Also, bears less than 5 years old were excluded because the levels of some OCs were higher in younger animals, probably due to high exposure during nursing. The trends in adult female bears are therefore more likely to represent actual changes in the polar bear food chain.

For many, but not all, OCs in female bear fat, concentrations had declined significantly from maximum values in 1968 and/or 1984 up to the 1990s (Fig. 21). Levels of  $\alpha$ -HCH in female bears were about 2–3 times higher in 1968 and 1984 than the average in the 1990s and 2002. By contrast,  $\beta$ -HCH concentrations were lower in 1968 than in any subsequent year, and the overall trend in the 1980s and 1990s is very flat. As a consequence, a significantly higher proportion (approximately 50%) of present day  $\Sigma$ HCH in polar bears from Hudson Bay is  $\beta$ -HCH compared to 1984 (25%) and 1968 (17%). This trend in  $\beta$ -HCH has also been observed in ringed seals at two High Arctic locations (see Figs. 17 and 18), and in beluga in Cumberland Sound, Baffin Island (see Fig. 20). There was a significant decline in  $\Sigma$ HCH between 1984 and the 1990s.

The trends for  $\Sigma$ CBz and  $\Sigma$ CHL were similar, and appeared to increase between 1968 and 1984 followed by a consistent downward trend after that

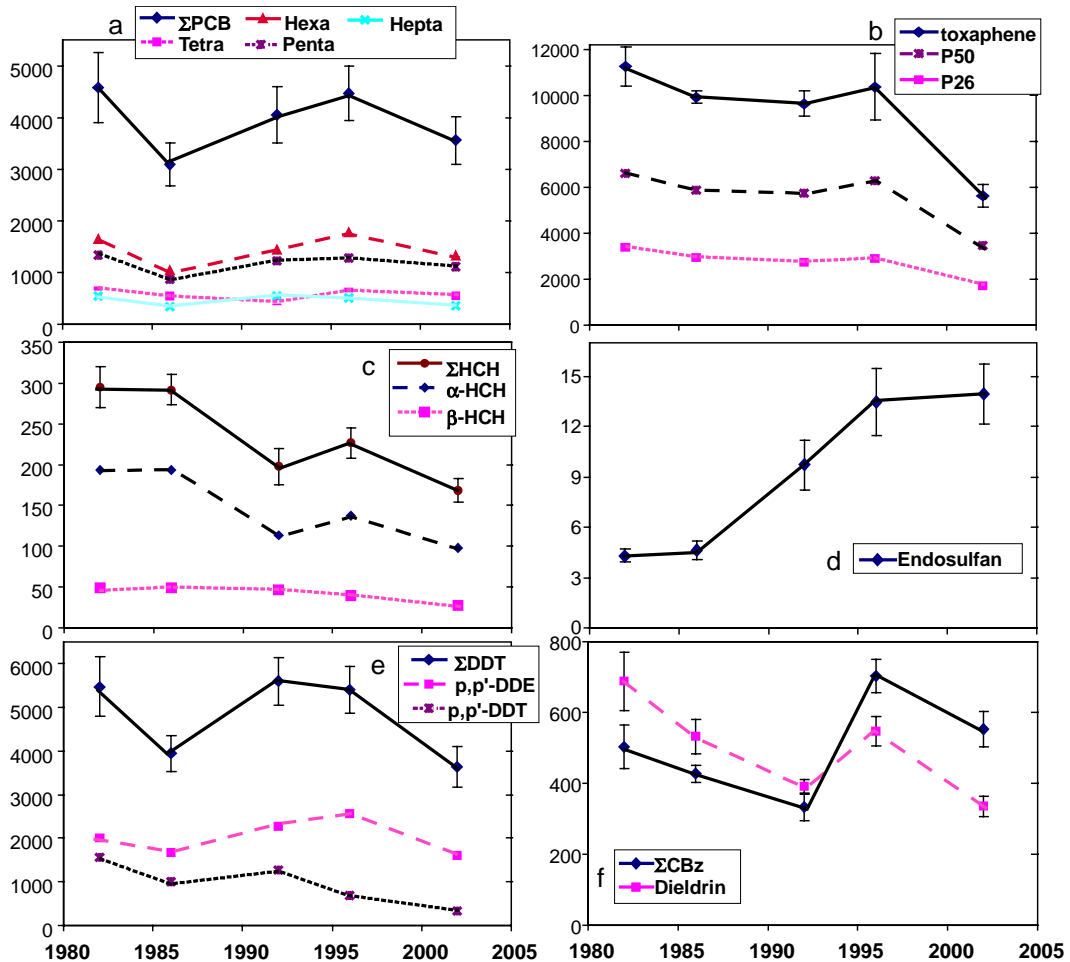


Fig. 20. Temporal trends of age-adjusted concentrations in blubber of male beluga from Pangnirtung in the eastern Canadian Arctic of: (a) sum of tetra-, penta-, hexa- and heptachlorobiphenyls ( $\Sigma$ PCB); (b)  $\Sigma$ toxaphene and congeners 50 and 26; (c)  $\Sigma$ HCH,  $\alpha$ - and  $\beta$ -HCH; (d) endosulfan; (e)  $\Sigma$ DDT,  $p,p'$ -DDE and  $p,p'$ -DDT; (f) sum of tetra-, penta- and hexachlorobenzene ( $\Sigma$ CBz) and dieldrin. Redrawn from Stern (1999) and Stern and Ikonoumou (2003).

time. Most of the decline in  $\Sigma$ CBz was due to HCB, which had a half-life in bear fat of approximately nine years during the 1990s. HCB and 1,2,4,5-TeCB were each roughly half of the total, with a minor contribution from PnCB. The proportion of 1,2,4,5-TeCB peaked at 53% between 1995 and 1997, and then decreased to values similar to the pre-1995 period (i.e., 40–45%).

There was a generally consistent (and significant) decrease of approximately 80% in  $\Sigma$ DDT over the 34 year study period. Declines of  $\Sigma$ DDT of a similar

magnitude have occurred since the early 1970s in ringed seal, the major prey of polar bears, though there are no directly comparable seal data from Hudson Bay (see Section 5.3.1.1). Concentration changes during the 1990s were less marked, and there was no significant change between 1991 and 2002.

There was less than a factor of 2 variation in  $\Sigma$ PCB levels in bears throughout the 1968–2002 period, and no long-term trend was apparent. Concentrations in the 1990s in Hudson Bay bears were similar to those in the late 1960s, in sharp contrast



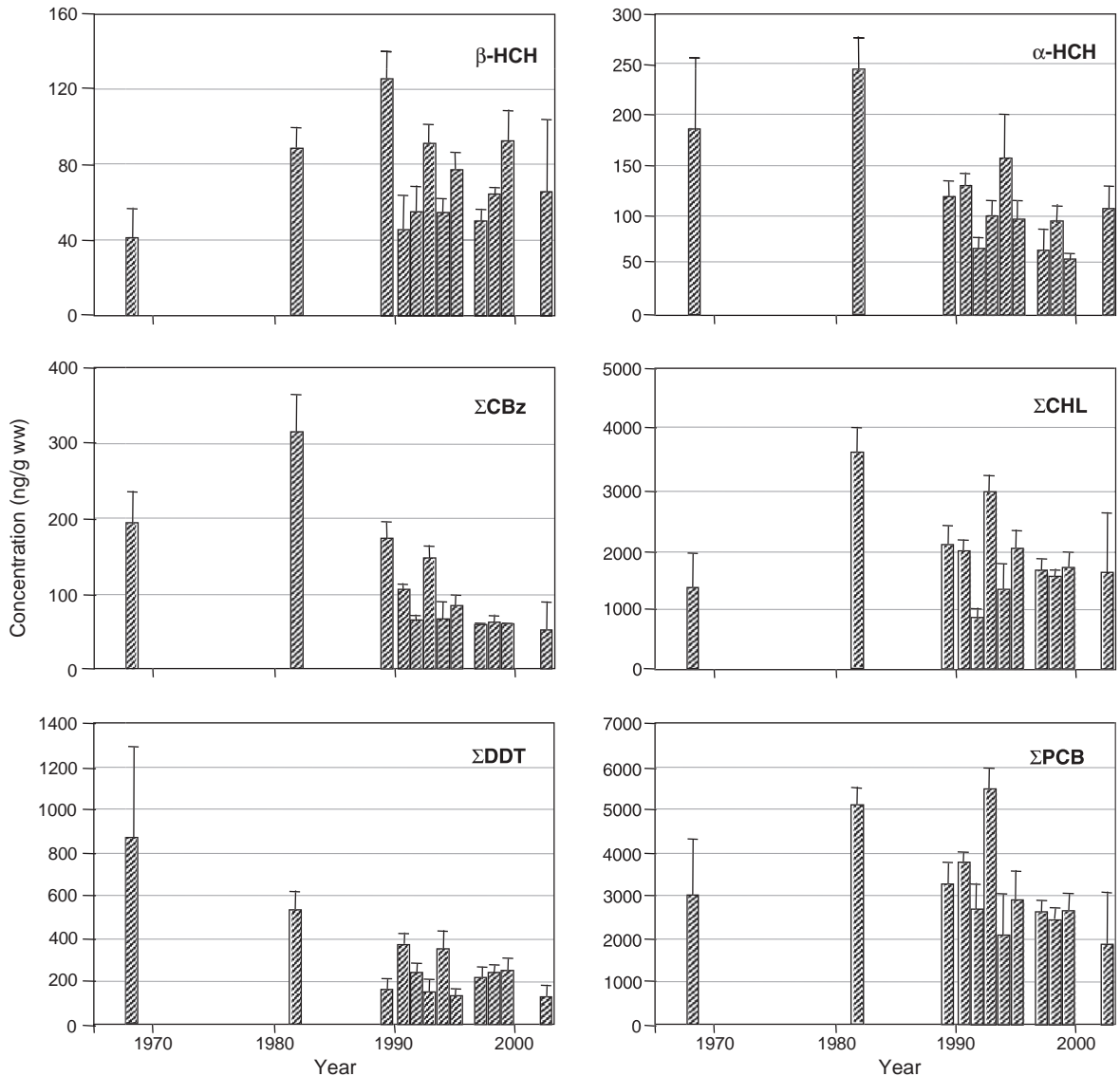


Fig. 21. Temporal trends of major organochlorines in Hudson Bay polar bear adipose tissue (Norstrom, 2001; Letcher et al., 2003). Samples are from the Churchill area of western Hudson Bay from 1968 to 2002. Samples from 1991 to 2002 are fat biopsies but earlier samples are adipose tissue.

to the situation in areas such as the Great Lakes or the North Atlantic, where PCBs in herring gull eggs have declined by about 10 times since the late 1960s and early 1970s (Hebert et al., 1997).  $\Sigma$ PCB decreased fairly steadily throughout the 1990s, but with a long half-life of approximately 18 years. The shift in composition of PCBs was subtle over the decade, but there

was a clear tendency for the proportion of less chlorinated congeners to increase, and the highly chlorinated congeners to decrease. CB99 increased from ca. 10% to 12% of  $\Sigma$ PCB, CB153 was relatively stable at about 35%, and CB180 decreased from 17% to 14%. The trends in these three congeners indicate that the half-life of CB153 (19 years) was similar to

that of  $\sum$ PCB. The half-life of CB180 (13 years) was shorter, and of CB99 (>50 years) was longer than  $\sum$ PCB. Thus, the decreasing trend in  $\sum$ PCB is driven by loss of the highly chlorinated congeners.

Shorter-term temporal data concerning OCs in polar bears were available from five other areas in the Canadian Arctic:

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

as reported by Muir and Norstrom (2000). In that comparison, 8–10 adult male polar bears per area in each of 1984 and 1989/1990 were used. Changes in PCDD/Fs were also determined in pooled samples. Organochlorine data were presented in Norstrom (1997) and Muir and Norstrom (2000). There was a significant decline of  $\sum$ HCH,  $\sum$ DDT and dieldrin in area I, a significant decrease of dieldrin in area II, no changes in area III, a significant increase of  $\sum$ CHL in area IV, and a significant decrease of  $\sum$ CBz,  $\sum$ DDT and  $\sum$ PCB in area V between 1984 and 1989.

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

PCB-153 concentrations decreased significantly (by about 25%) in plasma collected from polar bears sampled at Svalbard during the 1990s (Henriksen et al., 2001). This is a much faster decline than in the Canadian Arctic, but concentrations were

also much higher in Svalbard than in the Canadian Arctic in 1990 (Norstrom et al., 1998). Consequently, it appears that circumpolar concentrations of the more persistent contaminants in polar bears have been approaching a steady state over the past 15 years. It was estimated by Henriksen et al. (2001) that about 14 years of sampling PCBs in Svalbard polar bears would be needed to be >90% sure of detecting a change with an average rate of 5% per year. It was recommended that future monitoring efforts should sample annually at the same location, at the same time of year and analyze 10–25 samples per year. It was further recommended that blood samples be used in preference to other tissues. It would be difficult to implement these recommendations in the whole Canadian Arctic because of the much bigger geographical area; however, the sample size and frequency in the Hudson Bay study (Norstrom, 2001) met the recommendations.

Verreault et al. (2005) compared concentrations of PCBs in polar bear fat samples collected at 5 locations in the Canadian Arctic in 2002 with those from the same five locations in 1989–1991 and reported by Norstrom et al. (1998). In general,  $\sum$ PCB,  $\sum$ CHL and *p,p'*-DDE declined at all locations. The most consistent declines were observed for  $\sum$ PCB and *p,p'*-DDE, averaging 39% and 37%, respectively. The decline in  $\sum$ CHL ranged from 0 to 34% while dieldrin in 2002 exhibited higher or lower concentrations than in 1989–1991 depending upon location.

### 5.3.2. Temporal trends of new chemicals

Ikonomou et al. (2002) reported 9-fold increases of total (Br<sub>2</sub>–Br<sub>7</sub>)-PBDEs ( $\sum$ PBDEs) in male ringed seals aged 0–15 years from Holman over the period 1981 to 2000 (Fig. 22). Penta- and hexa-BDEs increased at approximately the same rate ( $t_2=4.7$  years and 4.3 years, respectively) and more rapidly than tetra-BDEs ( $t_2=8.6$  years) whereas tri-BDEs showed no increase in this age/sex grouping. The three most prevalent PBDE congeners (BDEs 47, 99 and 100) all increased over the 19-year period; however, only BDEs 47 and 100 increased in parallel with  $\sum$ PBDE. BDE-99 increased exponentially in a similar manner to  $\sum$ PBDE and BDEs 47 and 100 from 1981 to 1996; however, the levels of BDE-99 appeared to be stabilizing in 2000. This suggests a

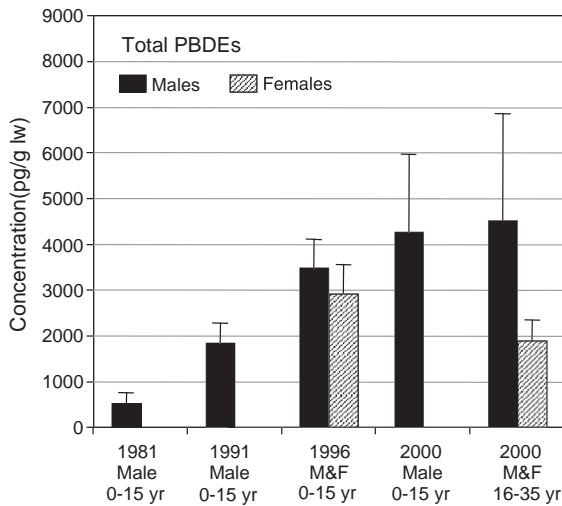


Fig. 22. Increasing concentrations of total Br<sub>2</sub>–Br<sub>7</sub> PBDE congeners in ringed seal blubber from Holman in the western Canadian Arctic (Ikonomou et al., 2002).

shift in sources or change in composition of PBDE products. No difference in PBDE levels (both total and of individual congeners,  $p=0.98$  for  $\sum$ PBDE) were observed between younger (0–15 years) and older (16–35 years) male seals in 2000, suggesting that recent PBDE accumulation dominates potential historic accumulation for the older seals.

Stern and Addison (1999) and Stern and Ikonomou (2000) studied temporal trends of PBDEs and PCDEs in beluga blubber samples from southeast Baffin Island. Levels of the total PBDEs (Br<sub>2</sub>–Br<sub>7</sub>) and major congeners increased significantly in southeast Baffin beluga over the period 1982 to 1997 (Fig. 23). Age-adjusted concentrations of BDE-47 (2,2',4,4'-TeBDE), the predominant PBDE congener, increased 6.5-fold over this 15-year period while BDE-154 increased 30 times. Concurrently, contributions of the tribromo-homologue group and PBDE-47 to total PBDEs declined by 7% and 3%, respectively. Conversely, pentabromo- and hexabromo-DE contributions increased by 20% and 80%, respectively. This compositional change in the beluga could be related to a shift in composition of commercial PBDEs to higher brominated mixtures (de Boer et al., 2000).

PCDE concentrations declined in these same beluga during 1982–1997 (Stern and Addison, 1999;

Stern and Ikonomou, 2000). The two most abundant congeners, CDE-99 (2,2',4,4',5-CDE) and CDE-154 (2,2',4,4',5,6'-CDE), declined 2.5-fold and 1.8 fold, respectively, over the 15-year period. These congeners are prominent contaminants in pentachlorophenol (PCP) wood preservative and the decline most likely reflects the ban on PCP use in Canada and Scandinavia and use restrictions in the USA.

MeSO<sub>2</sub>-PCBs were recently examined in fat samples collected in 2001–2002 from female polar bears between the ages of 5 and 15 years from six populations spanning the Nunavut and Northwest Territories (Letcher et al., 2003).  $\sum$ MeSO<sub>2</sub>-PCB decreased by about 50–60% relative to 1989–1991 levels for comparable Canadian populations (Letcher et al., 1995). Furthermore, the  $\sum$ MeSO<sub>2</sub>-PCB to  $\sum$ PCB ratios were virtually unchanged over approximately 10 years, suggesting that  $\sum$ MeSO<sub>2</sub>-PCB and  $\sum$ PCB concentrations are temporally co-variable in Canadian polar bears. Comparisons of mean 3-MeSO<sub>2</sub>-4,4'-DDE concentrations and 3-MeSO<sub>2</sub>-4,4'-DDE to 4,4'-DDE ratios for 2001–2002 with bear populations sampled in 1989–1991 showed similar declines to  $\sum$ MeSO<sub>2</sub>-PCB (Letcher et al., 1995). In the fat of polar bear collected from East Greenland in 1999 and 2001 (Sandala et al., 2004), mean  $\sum$ -MeSO<sub>2</sub>-PCB concentrations were  $699 \pm 836$  ng/g (lw) in fat ( $n=19$  males and females), and like those in Canadian

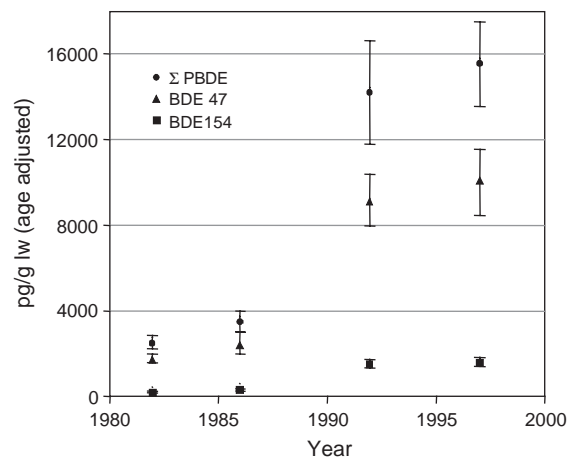


Fig. 23. Increasing concentrations of PBDE congeners 47 and 154 along with total Br<sub>2</sub>–Br<sub>7</sub> PBDE congeners in beluga blubber (SE Baffin Island, 1982–1996) (Stern and Ikonomou, 2000).

bears appear to have decreased relative to Scoresby Sound bears samples collected in 1989 and 1991 (Letcher et al., 1995).

### 5.3.3. Temporal trends of mercury

In the previous assessment, the data available to 1996 suggested increasing concentrations of Hg in seals, beluga and narwhal (Wagemann et al., 1996; Muir et al., 1999b) but the rates differed in the eastern and western Arctic, at least for beluga.

**5.3.3.1. Ringed seals.** Results for Hg in seal liver were available for several communities in the eastern Arctic (Qausuittuq (Resolute), Mittimatalik (Pond Inlet), Pangnirtung, Inukjuaq, Kangiqsujuaq (Wakeham), Kangiqsualujjuaq (George River)) and western Arctic (Holman, Sachs Harbour) from the 1970s to 1990s; however, they were presented only on a regional basis (Wagemann et al., 1996).

Recent (1998–2001) collection of ringed seal tissues from the same locations as those sampled in the 1970s, 1980s and early 1990s has enabled extension of the temporal data sets (Fisk et al., 2003a; Muir and Kwan, 2003). Results for both male and female ringed seals were combined because statistical analysis showed no effect of gender on hepatic Hg concentrations. The most complete data set was available from Holman Island, N.W.T. (Fisk et al., 2003a), where Hg concentration data along with corresponding animal ages were available for 1972, 1974 and 1977 from the original work of Smith and Armstrong (1978) as well as from Wagemann et al. (1996) and Muir and Kwan (2003). Trends in mean Hg concentrations in Holman ringed seals aged 5–15 years are shown in Fig. 24. Age-adjustment of the data was also conducted using analysis of covariance but it yielded essentially the same results. Average Hg concentrations in the seals varied markedly over the 30-year period, but not in a consistent temporal pattern. Significantly higher concentrations were found in 1974 and 1977 compared to 1993 and 1996, while levels in 2001 were also higher than in 1993.

Results for Hg in liver of ringed seals from seven other locations (Arviat, Hudson Strait, Inukjuaq, Pond Inlet (Mittimatalik), Resolute (Qausuittuq), Sachs Harbour, Ungava Bay) are shown in Fig. 25. Geometric mean concentrations from Wagemann et al. (1996 and related publications) were combined with

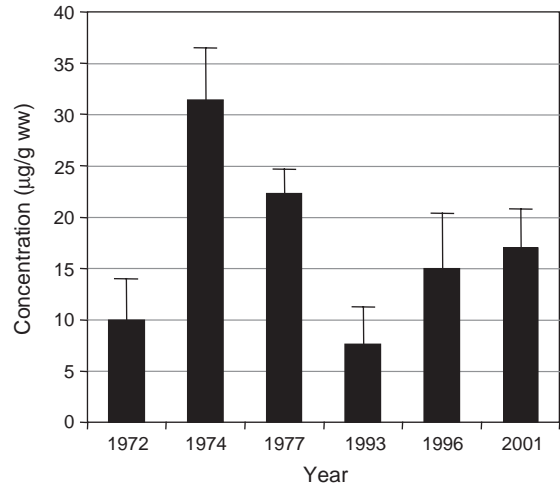


Fig. 24. Temporal trends of mercury in ringed seals aged 5–15 years from Holman, NWT (1972–2001). Bars represent geometric means and vertical lines represent upper 95% confidence intervals.

results from 1998 to 2000 (Muir et al., 2000b, 2001). The geometric means were not available from the 1970s, therefore for Mittimatalik (2000) and Qausuittuq, the arithmetic means in adult seals (5–15 years age) reported by Smith and Armstrong (1978) were used. The geometric means at Mittimatalik in 2000 were 3-fold higher than the arithmetic means in 1976, confirming previous conclusions by Wagemann et al. (1996) of increasing Hg over time at this location. Hg concentrations declined 1.5 to 2 times at Resolute, Ungava Bay and Sachs Harbour during the 1990s (Fig. 25) (Muir and Kwan, 2003). The year-to-year variation seen at several sites, e.g., Holman and Ungava, suggests that Hg levels in ringed seal liver can vary annually. Thus, the Mittimatalik data from 1976 and at Holman in 1993 may represent one of those low exposure years, presumably due to dietary shifts. To confirm this would require annual sampling and analysis of prey species.

**5.3.3.2. Beluga.** Long-term trend data for Hg in beluga, based on archeological and modern teeth samples, have recently become available which show the extent to which present Hg concentrations exceed the pre-industrial (i.e., natural sources only) background. The teeth of Beaufort Sea beluga harvested in 1993 as part of the traditional Inuit hunt contained significantly higher Hg concentrations than

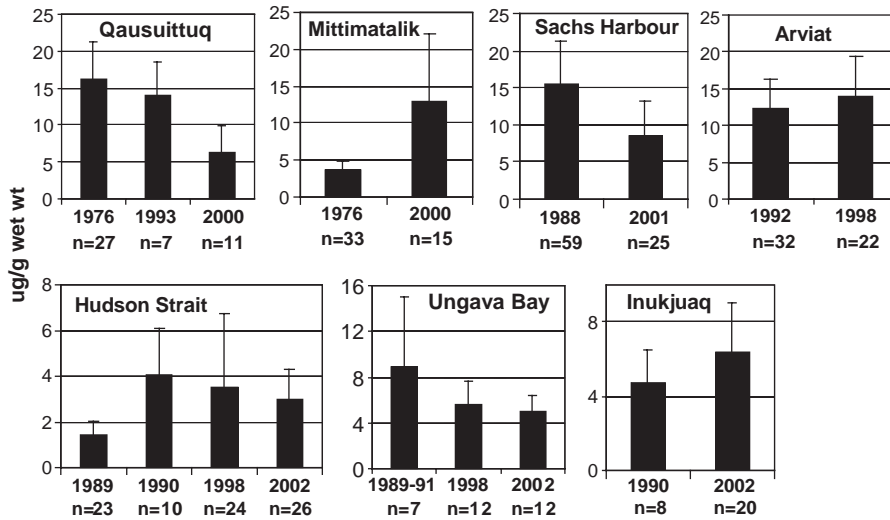


Fig. 25. Temporal trend analysis of mercury in ringed seal liver from seven locations in the Canadian Arctic. Bars represent geometric mean concentrations and vertical lines represent upper 95% confidence intervals. Results for 1976 from Pond Inlet and Resolute are arithmetic means reported by Smith and Armstrong (1978). For all other results, geometric means were calculated from the original data of Wagemann et al. (1996; and related publications) and Muir and Kwan (2003).

samples dated 1450–1650 AD from this region (Fig. 26). The concentration increases were age-dependent, ranging from 4-fold higher in 10-year-old beluga, to 17-fold higher in 30-year-old animals, a pattern which matches that predicted by Hg modeling work (Bern-

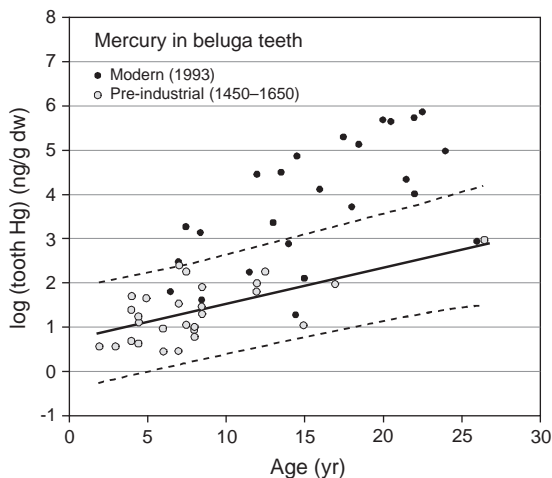


Fig. 26. Mercury in modern and pre-industrial teeth of Beaufort Sea beluga whales from the Mackenzie River delta (Outridge et al., 2002). Note that the scale of the vertical axis is in log-normal units. The maximum concentration in modern teeth was 360 ng/g dw.

hard and Andreae, 1984). Because tooth Hg in the modern beluga was significantly correlated with soft tissue Hg (including in the muscle and muktuk that Inuit traditionally consume), it is likely that soft tissue Hg has also increased. The Hg increase could not be explained by changes in diet (shown by stable C and N isotope analyses), sex differences in the harvested sample, or any other natural phenomenon; anthropogenic Hg inputs were therefore the most likely explanation. In contrast, pre-industrial and modern walrus at Igloolik in Foxe Basin contained similar concentrations of Hg in their teeth, suggesting an absence of anthropogenic Hg in this species at this location (Outridge et al., 2002).

Wagemann et al. (1996) showed that Hg in livers of beluga increased significantly in both the western and eastern Canadian Arctic from 1981–1984 to 1993–1994. Similarly, livers of narwhal sampled from Pond Inlet in the eastern Canadian Arctic in 1992–1994 had significantly higher mean concentrations of Hg than the narwhal sampled from the same site in 1978–1979 (Wagemann et al., 1996). Mercury data from the late 1990s and 2001 is now available for beluga from several sites in the Canadian Arctic, extending the time series particularly for the Beaufort coast (Lockhart et al., 2005b). Age-adjusted



mean concentrations of Hg in beluga liver reported by Lockhart et al. (2005b) from four locations for which three or more sampling times were available are presented in Fig. 27. Robust regression equations were used to calculate the level of Hg in a 13-year-old whale from each collection. For Mackenzie Bay, an increase in levels of mercury in males was found in all samples from 1993 and later relative to the levels found in 1984. Overall, there was a 4-fold increase of hepatic Hg in beluga from the Beaufort Sea coast between 1981 and 1996, with most of the increase occurring in the 1990s, but no further change between 1996 and 2002 (Lockhart et al., 2005b; Stern et al., 2003). Whales in the eastern Canadian Arctic also showed a somewhat consistent pattern of increase in Hg from the 1980s to the 1990s. Mercury levels in Pangnirtung beluga in 1993 and 1997 were greater than those measured in 1984. No significant change in Hg levels were found at Coral Harbour (northern Hudson Bay), Kimmirut (Hudson Strait) and Sanikiluaq (southeastern Hudson Bay) although samples spanned intervals of only 4 to 7 years. Beluga from Arviat (western Hudson Bay) showed much higher levels in 1999 than in 1984, with the age of whales in both years being similar. At the two locations where samples were taken 13 years or more apart (Beaufort coast, Arviat), an increased rate of Hg accumulation with age was evident in the more recent sample (Figs. 28 and 29). Beluga showing the largest Hg increases were

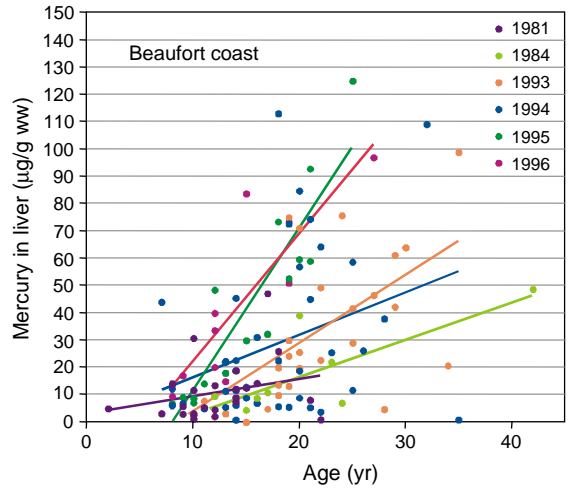


Fig. 28. Scatter plot of levels of mercury in liver of beluga whales collected in different years from the Beaufort sea coast (Lockhart et al., 2001). Least squares regression lines are shown.

collected in areas influenced by large freshwater drainages (Mackenzie River, Nelson River, James Bay rivers). This suggests that changes in Hg accumulation by the beluga may be related to freshwater inputs to these water bodies (Lockhart et al., 2001, 2005b).

5.3.3.3. Polar bears. In a long-term study using hair samples, Hg levels in modern polar bears were reported to be several times higher than in pre-indus-

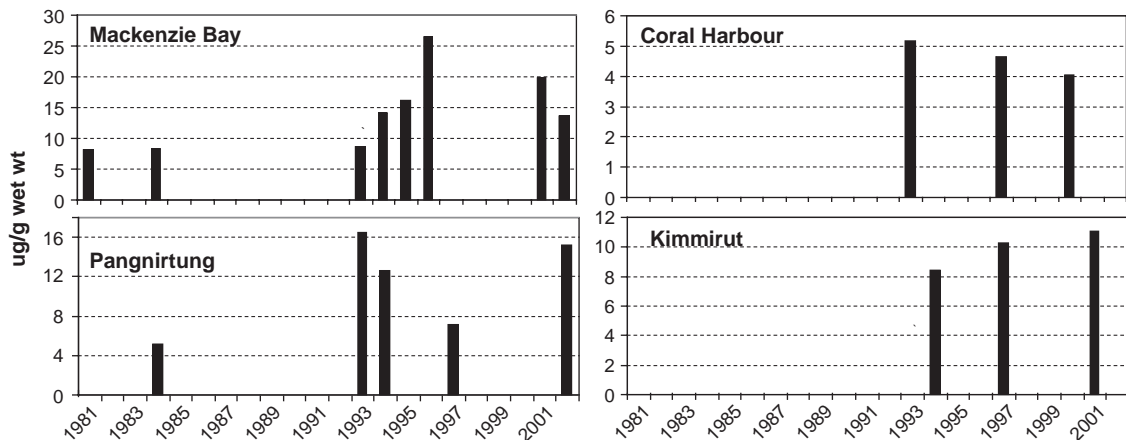


Fig. 27. Age-adjusted levels of total mercury (µg/g ww) in beluga whale liver at age 13 years from four locations in the Canadian Arctic (from Lockhart et al., this issue-b).

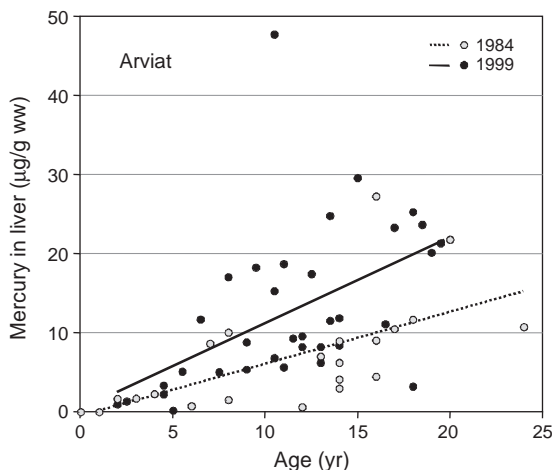


Fig. 29. Scatter plot of levels of mercury in liver of beluga whales collected in different years from Arviat on the western coast of Hudson Bay (Lockhart et al., 2001). Least squares regression lines are shown.

trial samples recovered from archeological sites (Wheatley and Wheatley, 1988). Possible differences in age (which is a significant factor in the Hg content of mammals) between modern and pre-industrial populations could not be assessed, and may have influenced the results. Letcher et al. (2003) reported that hepatic Hg concentrations in polar bears sampled in 2001–2002 in various regions of the Canadian Arctic were similar to those determined in the early 1980s, but cautioned that owing to a lack of age data for the recent samples, no definite conclusions could be drawn.

## 6. Summary and conclusions

Approximately 100 legacy OCs (including PCBs, DDTs, chlordanes, HCHs and CBz) as well as Hg and a number of new-use chemicals have been measured in Canadian Arctic marine wildlife. From a pan-Arctic perspective using ringed seals as a common indicator species, concentrations of many OCs in Canadian marine ecosystems are generally lower than in the European Arctic and eastern Greenland but are higher than in Alaska, whereas Hg concentrations at several Canadian sites are substantially higher than anywhere else.

The summary and conclusions below deal separately with the spatial and temporal trends of legacy OCs and Hg, and with new chemicals for which only relatively sparse data are available.

### 6.1. Spatial trends of legacy organochlorines and mercury

Within Canada, the spatial trends of most legacy OCs in marine biota are relatively subtle. For example, data on OCs in ringed seals from a number of locations show that concentrations generally range within a factor of 2. However, within this range there are consistently higher OC levels (for  $\sum$ PCBs,  $\sum$ DDT and their metabolites) in the eastern Arctic (Hudson Bay, Labrador Coast and around Baffin Island) than in the western Arctic (Beaufort Sea and Amundsen Gulf).  $\sum$ HCH is the only OC group to show the opposite trend with higher concentrations in the western Arctic. These geographic patterns are reflected at different levels in marine food webs: in several marine mammal species (ringed seals, beluga and narwhal) as well as zooplankton. In the eastern Arctic, more fine-scaled geographical differences seem to occur. Combined results from several different studies suggest higher levels of most OCs in narwhal sampled from northeastern Baffin Island (Pond Inlet) than eastern Baffin Island (Broughton Island), or southern Ellesmere Island (Grise Fiord). The reason for this is unclear.

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

Mercury data are considerably more limited in coverage, geographically and taxonomically, than OC data, and are notably sparse for marine fish. Significant geographic differences in Hg concentrations occur in beluga, being 3-fold higher in Beaufort Sea animals than in those around eastern Baffin Island and western Hudson Bay. There was also a large difference in mean Hg levels between beluga caught in Mackenzie Bay and at Paulatuk despite all nominally belonging to the same Beaufort Sea stock. The reason for this difference is unknown. It has been suggested that proximity to major river outflows may be a factor in the geographic pattern; this hypothesis should be tested in future studies because it may suggest a major influence of rivers on Hg speciation in the near-shore marine ecosystem, possibly mediated by extensive riverine wetlands. In ringed seals, however, there was no corresponding east–west difference in Hg body burden like that in beluga; highest concentrations were reported in western Hudson Bay but overall the differences were less pronounced than for beluga.

Biomagnification is the predominant process influencing animal body burdens of OCs and Hg, with the highest concentrations found in species at the top of marine food chains. Thus, generally the highest OC levels are found in polar bears and in seabirds such as glaucous gulls which scavenge marine mammal carcasses. This process has been studied quantitatively in several trophic web studies which linked contaminant levels in species with their dietary habits, as revealed by stable N isotope analyses. Consistent relationships were found between trophic status and Hg and OC levels in different Arctic marine food webs. The *in vivo* biotransformation of OCs by mammals has been shown to be a source of exposure to novel compounds, especially in polar bears. Organochlorine metabolites, particularly MeSO<sub>2</sub>-PCB and HO-PCB, are high in polar bears and are of concern due to their endocrine disrupting potential. Age and sex are also important variables in animal OC body burdens, with concentrations increasing with age, but there is conflicting evidence about whether gender-based differences exist. Organochlorine concentrations increase with age in male seals, for example, but not in female seals in some studies. Loss of OC body burden during lactation may explain the female pattern.

Mercury also increases with age in most mammals, but there is no apparent gender effect. For seabirds,

migration annually to more temperate latitudes may influence body burdens of OCs and Hg, generally leading to higher OC and lower Hg concentrations than in species which remain in northern latitudes year-round.

## 6.2. Temporal trends of legacy organochlorines and mercury

Temporal trend studies on biota have begun to address a major data gap identified for both the Canadian and circumpolar Arctic (de March et al., 1998; Muir et al., 1999b). The fact that these studies were, in virtually all cases, retrospective surveys based on tissue archives updated with samples from recent animal harvests shows the importance of continued monitoring of contaminants and of support for continuing tissue archiving for the future.

Long-term (i.e., century-scale) data for Hg, which form the basis for distinguishing and quantifying anthropogenic Hg inputs, remain sparse for Arctic biota. A north–south comparative study using bivalve shells found evidence of significant modern Hg increases only in Hudson Bay; elsewhere in the eastern Arctic, modern Hg levels appear to be similar to those thousands of years ago. In the western Arctic, Hg concentrations in modern Beaufort Sea beluga appear to be, on average, 10-fold higher than 500 years ago, based on analyses of teeth. There is also evidence of a several-times Hg increase in modern polar bears over the pre-industrial background, based on hair samples.

For temporal trends of OCs in biota, a 25- to 30-year perspective is available for polar bears, seabirds and ringed seals, and 10–15 years of data are available for beluga. Most legacy OCs significantly declined in marine biota between the 1970s and the 1990s. The one exception is  $\sum$ HCH, which has remained relatively constant in most species, probably due to the continued use of these chemicals in Asia until very recently. The general OC decline was most rapid in the 1970s and 1980s following their ban or reduced usage in the USA, Canada and western European countries. However, over the last decade, the rate of decrease has slowed or stopped in many instances. The rate of decline has also varied among species, being more rapid in ringed seals and seabirds, and slower in polar bears and beluga. Relative proportions

of individual chemicals have also changed with more recalcitrant compounds becoming more prevalent. For example, PCB 153, DDE and  $\beta$ -HCH now make up a greater proportion of their respective groups than in the past.

Mercury trend data are available for seabirds, beluga and ringed seals from a number of populations, spanning the 1970s through to 2000–2003. A 3-fold increase of Hg was observed in Pond Inlet ringed seals between 1976 and 2000 but no significant increase occurred in six other seal populations, including at Holman from which the longest data sets are available. Given the significant inter-annual variation seen at several sites, e.g., Holman and Ungava, the results from Pond Inlet should be treated cautiously. Increasing attention on possible dietary shifts over time, by simultaneously monitoring stable C and N isotope ratios in tissue samples, would help to avoid any confounding effects on the Hg time trend data.

Another explanation for the differences among seals at different sites is that the temporal patterns of Hg are specific to local areas. There is evidence that the Pond Inlet Hg increase in seals occurred in the surrounding region and in other species as well. Mercury increased in narwhal collected at Pond Inlet between 1978–1979 and 1992–1994, and a similar increase of Hg occurred in the eggs of northern fulmars and thick-billed murres from Lancaster Sound over the period 1975–1977 to 2003 and in ringed seals from Avanersuaq in northwest Greenland between 1984 and 1998 (Riget, Danish Environmental Research Institute, personal communication). Thus, there is consistent evidence across a number of species for significant Hg increases in the Lancaster Sound–northern Baffin Bay marine ecosystem over the past few decades.

Increases of Hg in beluga also appear to be localized, with significant recent increases only for the Beaufort Sea (during the mid-1990s) and at Arviat. In the Beaufort Sea, these increases were not paralleled in ringed seals from the same general region, e.g., Sachs Harbour and Holman, although seals from Mackenzie Bay have not been sampled.

Rivers are probably an important transport vehicle for Hg entering the Arctic marine environment although there are few measurements to confirm this. The large increases in Hg concentrations that have been observed recently in the Mackenzie Bay beluga

coincide with a warming trend in the western Arctic. Hypothetically, the recently observed melting of permafrost and likely release of organically bound Hg from Mackenzie River Valley peatlands may play a role in the Beaufort Sea beluga Hg story. The influence of climate warming on levels and ecosystem dynamics of OCs and Hg in the Arctic should be a priority area for future study.

### 6.3. New contaminants in the Arctic

New information on toxaphene has been generated for seabirds and marine mammals, and new data for PCDDs, PCDFs and non-*ortho* PCBs has been generated for seabirds. In addition to the legacy OCs which occur in the Arctic largely as a result of historical manufacturing and usage, numerous new contaminants, which were mostly developed in the past two decades, have recently been detected in Canadian Arctic marine biota. In particular, information on polybrominated diphenyl ethers (PBDEs) has been generated for several marine organisms while measurements of short chain chlorinated paraffins (SCCPs) and chlorinated naphthalenes (PCNs) were made in beluga and ringed seals. Although  $\sum$ PBDE levels are low compared with the legacy OCs, they are rapidly increasing as has been demonstrated in Holman ringed seals, southeastern Baffin Island and western Arctic beluga, as well as seabirds from Lancaster Sound. The discovery of perfluoro-octane sulfonic acid, PFOS, as well as PFCAs in Canadian Arctic biota, was not expected due to their physical–chemical properties. The presence of these chemicals at significant concentrations raises questions about our knowledge of the sources and pathways of these and other polar chemicals and possible volatile precursors. There is insufficient information on most new chemicals to assess species differences, spatial trends or food web dynamics; continued temporal trend monitoring of these chemicals must be regarded as a priority.

### Acknowledgements

The authors wish to thank the Northern Contaminants Program of Indian and Northern Affairs Canada for providing funding to most of the authors which

made possible the generation of much of the data presented in this report. The authors would like to thank Jill Watkins and Jason Stow for their support in the preparation of the assessment report version of this review and the preparation of this document. Xiaowa Wang (NWRI) provided valuable assistance on many logistical details in the initial preparation of the document. We also thank Russell Shearer for his never-ending support and enthusiasm for the Northern Contaminants Program (NCP). We would also like to acknowledge the Aboriginal organizations that have supported the NCP program and its contaminant measurement projects. In particular, we would like to thank the community councils and Hunters and Trappers Organizations of Yukon, NWT, Nunavut, Nunavik and Labrador. Their cooperation and active participation in the collection of biological samples made all of this work possible.

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