*Chapter 6*

# Persistent Organic Pollutants

## Contents

## 6.1. Introduction

Interest in the presence of persistent organic pollutants (POPs) in Arctic aquatic and terrestrial ecosystems arises mainly from concerns that northern and indigenous peoples depending upon traditional food for most of their diet may be adversely affected by chronic exposure to these pollutants (Dewailly *et al.* 1989, Kinloch *et al.* 1992, chapter 12). This chapter deals mainly with persistent organochlorine (OC) pollutants as well as organotins. Methylmercury is dealt with in chapter 7 and polycyclic aromatic hydrocarbons (PAHs), another important group of POPs, are discussed in chapter 10. The persistent organochlorine contaminants are of special concern because 1) they biomagnify and persist in the environment for long periods of time, 2) indigenous people in the Arctic depend on a high fat diet which is a source of these contaminants, and 3) most northern residents have not used or directly benefited from the activities associated with the production and use of these chemicals. There are also concerns that the health of top predators such as small-toothed whales, seals, predatory birds, and polar bears may be affected because of chronic exposure to organochlorines and other persistent organic pollutants in their diet.

Information on the presence of persistent organochlorines in Arctic ecosystems is not new. Their presence in Arctic ecosystems was well known among scientists interested in the global distribution of these chemicals, but information on spatial trends in these contaminants was very limited. Most studies regarded the Arctic as a relatively pristine site for comparison with more contaminated southern locations. The first report of chlorinated organic chemical contamination of marine mammals in the Arctic was by Holden (1970) who detected the pesticides dieldrin and DDT as well as PCBs in blubber of ringed seals (*Phoca hispida*) from the Canadian and Norwegian Arctic. More detailed reports on DDT-related compounds and PCBs in ringed seal and beluga (*Delphinapterus leucas*) from Canada and Greenland were published in the early 1970s (Addison and Brodie 1973, Addison and Smith 1974, Clausen *et al.* 1974). Bowes and Jonkel (1975) were the first to report OCs in polar bears (*Ursus maritimus*). OC contaminants were also determined in freshwater and anadromous fish in the early 1970s (Reinke *et al.* 1972, Bowes and Jonkel 1975).

The biological effects of the organochlorines in Arctic ecosystems were not investigated in earlier work with the exception of effects on birds of prey. The decline of populations of birds of prey in Arctic and temperate areas of the northern hemisphere since the introduction of OC pesticides has been well documented (Ratcliffe 1967, Peakall 1976a, Newton 1979, Peakall *et al.* 1990). The human health aspects of contamination in the Arctic were brought to the world's attention in the late 1980s when it was discovered that mother's milk from northern Quebec contained unacceptably high levels of PCBs (Dewailly *et al.* 1989) (chapter 12). The results of studies on these contaminants in the human diet (Kinloch *et al.* 1992), and in mother's milk, demonstrated the transfer of bioaccumulating contaminants to humans and created a demand for more information on contaminants in 'traditional foods'.

Several developments in the late 1980s helped to accelerate the assessment of contaminants in Arctic food webs. Improvements in analytical chemistry techniques in the 1980s permitted a more comprehensive search for individual organochlorine and hydrocarbon contaminants. Development of biochemical effects tests, such as cytochrome P450 mixed function oxidase (MFO) enzyme activity, DNA oxidative damage, and immunosuppression, also provided tools for assessing biological effects of the contaminants in wildlife.

Also discussed briefly in this chapter are other pesticide-related compounds that have been shown to be present in the Arctic, especially air and seawater. Less-persistent pesticides including atrazine, endosulfan, chlorpyrifos, chlorothalonil, tetra- and pentachlorophenol (as the anisole), trifluralin, and methoxychlor have been detected in Arctic air, fog, and seawater, and in some cases in plants and aquatic biota. These chemicals are still registered for use as pesticides in most circumpolar countries and elsewhere in the world. The organotins, particularly tributyltin (TBT), a broad-spectrum algicide, miticide, fungicide, and insecticide, are a concern in the marine environment in general, and have been found in Arctic waters. Another group of POPs, the brominated aromatic compounds, which are used primarily as fire retardants, have been detected in Arctic biota and may be of concern in the future.

### 6.1.1. Physical and chemical characteristics of persistent organic pollutants (POPs)

The POPs dealt with in this chapter belong to several groups depending on their use or origin. Except for the organotins, they are all halogenated, the majority with chlorine. Although they have diverse chemical structures, the common characteristics for most of them are low water solubilities, high lipophilicity (accumulation in fat), and resistance to biodegradation (recalcitrance). These combined characteristics lead to uptake and accumulation in the fatty tissues of living organisms. Those that are or have been used as pesticides are designed to be toxic, particularly to invertebrates or fungi, but may also be toxic to fish, birds, and mammals. These include DDT, hexachlorocyclohexanes (HCH), chlordanes, aldrin, dieldrin, mirex, toxaphene, endosulfan, and hexachlorobenzene (HCB).

Commercial PCB (polychlorinated biphenyl), chlorobenzenes, and PBDE (polybrominated diphenyl ether) mixtures are technical products with widespread uses. Although open use of PCBs was banned in many countries during the 1970s, they are still used in some closed systems (e.g., until recently in fluorescent light ballasts) and are present in the environment as a result of past disposal practices. PCDD/Fs (polychlorinated dibenzo-*p*-dioxins and dibenzofurans) are by-products formed in the production of chlorinated substances, in high temperature thermal reactions such as incineration, metallurgical processes, and in direct chlorination reactions such as chlorine bleaching of wood pulp.

All of the POPs mentioned have been detected in Arctic abiotic or biotic samples and most of them are on lists of priority pollutants established by circumpolar and other governments for regulation of environmental contaminants. Many of these contaminants are associated with biological effects observed in more polluted environments such as the Baltic Sea, the North Sea, and the Great Lakes. They have been selected for the AMAP assessment because they would be expected to have effects on Arctic biota if exposures were similar to those in areas farther south.

The toxicological characteristics of POPs are discussed in more detail in section 6.5. In general terms, studies with laboratory animals and an increasing number of environmentally exposed organisms, have shown that exposure to organochlorine pollutants leads to immunosuppression, reduced reproduction rates, and induction of liver detoxification enzymes, for example, cytochrome P450 mixed function oxidases. Some organochlorines act as promoters, increasing the tumorigenic response to a carcinogen.

Selected chemical and physical properties, and chemical structures of most POPs considered in this chapter are given in Annex Tables 6·A1 and 6·A2. The properties of organic chemicals that are critical to understanding their movement through the abiotic and biotic environment include water solubility, vapor pressure (VP), Henry's Law constant (H), partition coefficients including octanol/air {$K_{oa}$}, octanol/water {$K_{ow}$}, and sediment or soil (organic carbon)/water sorption {$K_{oc}$}. The subcooled liquid vapor pressure ($VP_L$; see section 6.2.1.1), together with the surface area of atmospheric aerosols, determines particle/gas partitioning. H is defined as the ratio of the VP of a substance to its solubility in water and describes the equilibrium air–water partitioning. $K_{ow}$ is used to describe the equilibrium distribution of organic contaminants between lipid phases and water and is widely applied as a correlation parameter for bioaccumulation. The relationship of these properties to accumulation and metabolism is discussed in section 6.4. $K_{oa}$ describes the distribution of organic contaminants between air and lipid-like phases such as plant waxes (Harner and Mackay 1995) and organic films on aerosols. For additional information on physical properties and pathways, readers should consult reference texts of Howard (1991) and Mackay *et al.* (1992b, 1992c).

The importance of physical and chemical characteristics in the movement of OCs in the Arctic food web is demonstrated in Figure 6·1, taken from Norstrom and Muir (1994).

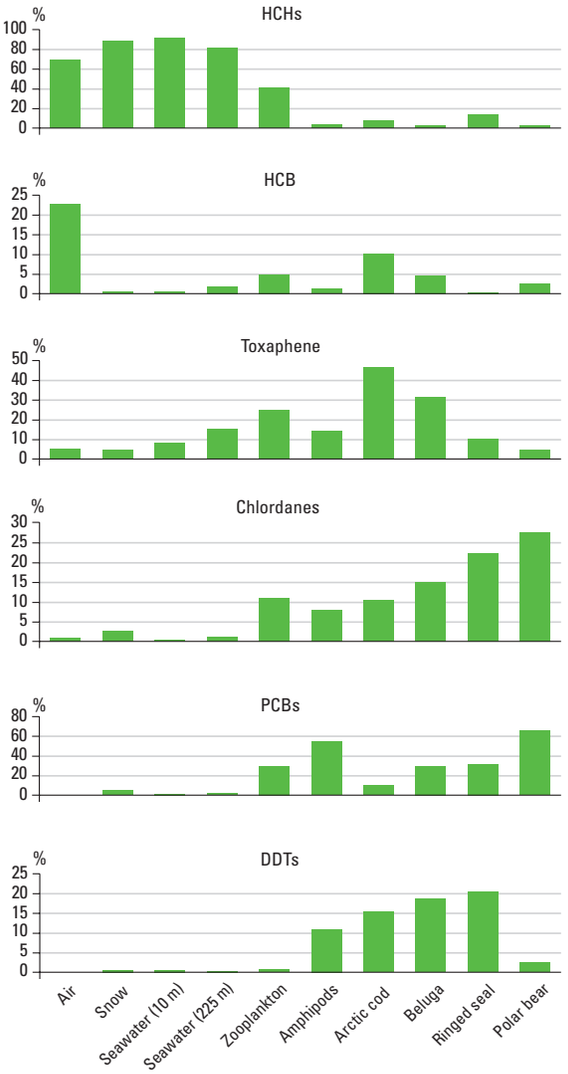



Figure 6·1. Distribution of organochlorine contaminants (OCs) in Arctic air, snow, seawater, and the marine mammals food chain (Norstrom and Muir 1994). Data for the six major classes of OCs are plotted for each compartment or species as the percent of OCs in that compartment or species to demonstrate the changing importance of residue classes in the process of transfer between compartments and bioaccumulation in the marine mammals food chain.

Table 6·1. Current legal status within the eight circumpolar countries of persistent organic pollutants (POPs) covered in this chapter.

| | USA (Alaska) | Canada | Russia | Iceland |
|---|---|---|---|---|
| Aldrin | Banned 1987 | Voluntarily withdrawn 1990 | n.i. [a] | Never registered as a pesticide. Banned 1996 |
| Dieldrin | Banned 1971 | Restricted 1987 | n.i. | Never registered as a pesticide Banned 1996 |
| Chlordanes | Restricted use 1988 | Registration discontinued 1990 | Use prohibited | Banned 1996 |
| DDT | Banned 1972. Products with more than 0.1% banned 1986 (Dicofol) | Voluntarily withdrawn 1985 | Banned 1970; unofficial use may continue | Never registered as a pesticide. Banned 1996 |
| HCB | Registration cancelled 1984 | Registration not renewed 1976 | n.i. | Never registered as a pesticide. Banned 1996 |
| α-HCH | Mixed isomers cancelled 1977 | Mixed isomers discontinued 1976 | Technical product probably still in use | Never registered as a pesticide |
| β-HCH | See α-HCH | See α-HCH | n.i. | See α-HCH |
| γ-HCH (lindane) | Cancelled for most uses 1983 | Restricted use | Still in use | Restricted use |
| Mirex | Use cancelled 1988 | Never registered as a pesticide | n.i. | Never registered as a pesticide |
| Toxaphene | Banned 1982, use ceased 1986 | Withdrawn 1982 | Severely restricted 1992 | Never registered as a pesticide. Banned 1996 |
| Endosulfan | Still in use | Restricted use for commercial pest control | n.i. | Restricted use in greenhouses |
| TBT | Restricted use 1988 | Restricted use – aluminum hulled boats in salt water | n.i. | Banned on vessels <25 m, docks and fishing gear |
| PCBs | New use cancelled 1970; closed use still allowed | Severely restricted 1985; open and new uses banned, closed use allowed | Limited use and manufacturing continues | >0.2% banned 1988; >0.005% banned 1996 |

a. n.i. = no information.

This shows the relative burden of six major OCs in air, snow, seawater, and the marine mammal food chain. In air, the principal medium of transport, the most volatile OCs, HCB and HCHs, predominate. The OCs which are most significant toxicologically, PCBs and DDT, are barely detectable. The HCHs take an even more dominant role in snow and seawater. Toxaphene (polychlorinated bornanes) behaves in a similar fashion to HCHs in the abiotic environment, while HCB decreases in importance. The reason for the changed distribution has to do with relative volatility and water solubility. The Henry's Law constant of HCHs and toxaphene are similar, and over 100 times lower than that for HCB. Thus, HCB partitions into water far less readily than HCHs and toxaphene. PCBs, chlordane, and DDT-related compounds have H values intermediate to HCHs and HCB. Incorporation into particulates and invertebrates at the bottom of the food chain can be predicted from $K_{ow}$. HCH has a $K_{ow}$ of about $10^4$, while $K_{ow}$'s of HCB, chlordanes, and PCBs are in the range of $10^5$-$10^8$. In the higher trophic levels of the food web, OCs with higher $K_{ow}$'s generally predominate, however, the extent of metabolism of individual components becomes important in determining actual levels of biomagnification. Thus, DDT does not biomagnify from seals to polar bears because of the capability of the bears to metabolize *p,p'*-DDE, a persistent OC in seals and whales.

Similarly, different PCB congeners are dominant at different levels of the food web, their concentrations depending on physical characteristics and metabolism. An example is given in Figure 6·12 (section 6.6.2.5), which shows levels of different PCB congeners in the lichen → caribou → wolf food chain.

### 6.1.1.1. **Industrial products**

#### 6.1.1.1.1. **Chlorinated industrial chemicals and by-products**

Persistent organochlorine contaminants, such as HCB and PCDD/F, are often produced as unwanted by-products of chemical processes. Others, such as PCBs and brominated diphenyl ethers, have been manufactured and used in large quantities because of their stability and flame-retardant properties.

*Hexachlorobenzene (HCB)*

HCB is produced as a by-product in the production of a large number of chlorinated compounds, particularly lower chlorinated benzenes, and in the production of several pesticides. It had limited use in the 1960s as a fungicide. HCB is emitted to the atmosphere in flue gases generated by waste incineration facilities and metallurgical industries. HCB has an estimated 'field half-life' of 2.7-5.7 years (Howard 1991). HCB has a relatively high bioaccumulation potential because of high lipophilicity (log $K_{ow}$ = 5.5) and long half-life in biota (Niimi 1987). Other chlorobenzenes, tetra- and penta-substituted, are also relatively lipophilic, semi-volatile, and persistent, especially in the abiotic environment (Mackay *et al.* 1992b, 1992c).

| | Denmark | Norway | Sweden | Finland |
|---|---|---|---|---|
| Aldrin | Prohibited for plant protection use | Not marketed since 1970 | Banned 1970 | Banned 1972 |
| Dieldrin | Prohibited for plant protection use | Not marketed since 1970 | Banned 1970 | Banned 1969 |
| Chlordanes | Prohibited for plant protection use | Not marketed since 1967 | Banned 1971 | Banned 1969 |
| DDT | Prohibited for plant protection use | Restricted 1969. Banned 1988 | Banned 1975. Dicofol banned 1991 | Banned 1976 |
| HCB | Banned | Withdrawn 1987 | Withdrawn 1980 | Ceased use on voluntary basis 1977 |
| α-HCH | Mixed isomers prohibited for plant protection use | n.i. | See γ-HCH | Mixed isomers cancelled 1977 |
| β-HCH | See α-HCH | n.i. | See γ-HCH | Mixed isomers cancelled 1977 |
| γ-HCH (lindane) | n.i. | Banned 1991 | Banned 1989 | Banned 1987 |
| Mirex | n.i. | Never used | Never used | Never used |
| Toxaphene | Banned 1987 | Never used | Slight use in 1950s; never used since | Banned 1969 |
| Endosulfan | n.i. | Still in use | Withdrawn 1995 | Still in use |
| TBT | n.i. | Restricted use – aluminum-hulled boats >25 m. All other use banned | Use banned in freshwater, the Baltic Sea and on ocean-going vessels <25 m | Banned on vessels <25 m |
| PCBs | Prohibited | New use banned 1980. All use banned 1995 | Open use banned 1972; new use banned 1978; closed use banned 1995 | Import and use banned 1990; all machines with PCB banned 1995 |

*Polychlorinated biphenyls* (*PCBs*)

PCBs were introduced in 1929 by the Monsanto Chemical Corporation and were manufactured in the USA, Japan, the former Soviet Union, and eastern and western Europe under various trade names (e.g., Aroclor, Clophen, Phenoclor). They are chemically stable and heat resistant, and were used worldwide as transformer and capacitor oils, hydraulic and heat exchange fluids, and lubricating and cutting oils (Verschueren 1983). Information about the legal status of their use and production is given in Table 6·1. Open use is currently banned in all circumpolar countries, but there are still large amounts in permitted use in large capacitors and transformers. Current uses and disposal practices in the developing world are not well documented.

There are 209 chlorinated biphenyl congeners, with different chlorine substitutions on the biphenyl ring (Mackay *et al*. 1992c). The number of chlorines, as well as positioning on the rings, influences the physical properties and biological activity of PCB congeners. The physical and chemical properties of selected congeners and homologues are given in Annex Table 6·A1. As with other POPs, the molecular weights and $K_{ow}$ of PCB congeners are correlated with their Henry's Law constant, vapor pressure, water solubility, and tendency to adsorb to plant and soil surfaces. PCB congeners with 3,3',4,4' chlorine substitutions, CBs 77, 126, and 169 (co-planar or non-*ortho* PCBs; abbreviated nPCB), as well as some 2,3,3',4,4' -substituted congeners (mono-*ortho* substituted congeners, for example, CBs 118 and 105) are the most biologically active (Ahlborg *et al*. 1992, 1994). The lack of chlorine substituents in the 2 and 6 (or *ortho*) positions permits nPCBs to assume a planar configuration similar to that of polychlorinated dibenzo-*p*-dioxins and dibenzofurans. The nPCBs and mono-*ortho* CBs (CBs 105 and 118) are collectively referred to as 'planar' PCBs in this chapter.

Most PCB congeners, particularly those lacking adjacent unsubstituted positions on the biphenyl rings (e.g., 2,4,5-, 2,3,5- or 2,3,6-substituted on both rings) are extremely persistent in the environment. They are estimated to have half-lives ranging from three weeks to two years in air and, with the exception of mono- and di-chlorobiphenyls, are essentially non-biodegradable in aerobic soils or sediments (Mackay *et al*. 1992a). Highly chlorinated PCBs have been shown to be dechlorinated in anaerobic sediments, but only where present at relatively high concentrations (>10 μg/g dw) (Brown *et al*. 1987, Rhee *et al*. 1993). PCBs also have extremely long half-lives in adult fish, for example, an eight-year study of eels found that the half-life of CB153 was more than ten years (de Boer *et al*. 1994).

*Polychlorinated dibenzo-p-dioxins and furans* (*PCDD/Fs*)

Polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) enter the environment as by-products of industrial processes. The most significant sources are low-temperature, incomplete incineration of chlorine-containing materials such as plastics. It has been reported that, in Russia, some portion

of the 30 million tons of halogenated sludge produced annually is incinerated, thus creating a potential source of PCDD/Fs (Federov 1993). Other major sources include thermal processes, such as motor vehicle fuel combustion in countries where leaded fuel containing chlorine scavengers is still used, and metallurgical industries. Pulp and paper mills using chlorine in the bleaching process have been important sources to the aquatic environment of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) and 2,3,7,8-tetrachlorodibenzofuran (2,3,7,8-TCDF). Emissions of 2,3,7,8-TCDD/TCDF by the pulp and paper industry in circumpolar countries, particularly in Sweden and Canada, have been significantly reduced in recent years because of the substitution of molecular chlorine by other bleaching agents. PCDD/Fs are also trace contaminants in chlorophenoxy herbicides, PCB formulations, and chlorophenol wood preservatives.

Most PCDD/F congeners, like PCBs, are extremely hydrophobic and resistant to biodegradation in soils and sediments. Historical profiles of PCDD/Fs in sediment cores from large lakes show no evidence of transformation of congeners (such as anaerobic dechlorination) over time (Hites 1990). The tetra- to octa-chlorinated PCDD/Fs have lower vapor pressures and Henry's Law constants than PCBs and are therefore not expected to undergo long-range transport to the same extent (Mackay *et al.* 1992c). PCDD/Fs are rapidly photodegraded in air, water, and on surfaces (Buser 1988). The 2,3,7,8-substituted PCDD/F congeners are known to bioaccumulate in fish and invertebrates, however non-2,3,7,8-substituted congeners (which predominate in combustion sources) are readily degraded by vertebrates (Opperhuizen and Sijm 1990).

#### 6.1.1.1.2. **Polybrominated diphenyl ethers (PBDEs)**

PBDEs (polybrominated diphenyl ethers) are aromatic compounds that are structurally related to the PCBs, but with bromine substitution instead of chlorine. Several brominated organic compounds are used as flame retardants in polymeric materials (Bergman 1989). There are two groups of chemicals, those that are covalently bound to a polymer and those that are mixed into materials. Additives may migrate out of the products and cause a diffuse contamination of the environment during the entire lifetime of the flame-retardant product. Some products that are flame-retarded are textiles, plastics, electrical equipment, building materials, and linings of vehicles. The increasing use of flame retardants in modern societies has led to increases of PBDEs in the environment (Sellström 1996), and may lead to increases in the Arctic as well. Concerns about the effects of polybrominated diphenyl ethers are similar to those for PCBs. Very little is known about their environmental concentrations and sources.

### 6.1.1.2. **Chlorinated pesticides**

#### 6.1.1.2.1. **Persistent pesticides**

*Polychlorobornanes and camphenes* (*Toxaphene*)

The complex mixtures of polychlorobornanes and camphenes known as toxaphene were widely used in the USA on cotton crops. Use peaked between 1972 and 1975. Manufacture was banned in the USA in 1982 and uses ceased in 1986 (Voldner and Li 1993). Similar products have been, and may continue to be used in Mexico, Central America, eastern Europe, and the former Soviet Union. Toxaphene is extremely persistent in soils following pest control application, with reported half-lives ranging from one to 14 years (Howard 1991). Losses from soil are mainly via volatilization and runoff (Goltfelty *et al.* 1989). The lack of degradation, combined with relatively high vapor pressure ($VP_L$ = $1.9 \times 10^{-3}$ Pa at 25°C), have resulted in the global dispersion of toxaphene via atmospheric transport, as for several other chlorinated pesticides.

Toxaphene is produced by the chlorination of technical camphene or α-pinene and can consist of over 300 congeners, mainly bornanes and camphenes substituted with 6-10 chlorines, with an average composition of $C_{10}H_{10}Cl_8$. Analysis has been difficult because of the mixture's complexity, because it occurs in the presence of other OCs (PCBs, DDTs, HCHs), and because of lack of standards for individual components. Analytical standards for some chlorinated bornanes have recently become available (Xu *et al.* 1994). Nevertheless, the levels and effects of toxaphene in the Arctic are not well studied even though it is a significant contaminant.

*Chlordanes* (*CHL*)

Technical grade chlordane is a mixture of at least 120 compounds, with the major constituents being *cis* (or α)- and *trans* (or γ)-chlordane, heptachlor, *cis*- and *trans*-nonachlor, α-, β-, and γ-chlordene, and others (National Research Council of Canada 1974, Dearth and Hites 1991, Howard 1991). In the past, chlordane was released into the environment primarily from its application as an insecticide and for seed dressings and coatings (CCREM 1987). In the USA, it was used extensively prior to 1983, and from 1983 to 1988 it was registered for termite control. It was cancelled for this use in 1988. Heptachlor is of particular interest since its oxidation product, heptachlor epoxide, is carcinogenic, and has been found in the Arctic abiotic and biotic environments. Oxychlordane is another toxic metabolite of chlordane.

Chlordane is very persistent in the environment, with an estimated half-life in soil of 1-4 years (Augustijn-Beckers *et al.* 1994). *Cis*- and *trans*-chlordane have high Henry's Law constants (87 and 132 Pa $m^3$/mol, respectively) compared to most other chlorinated pesticides, and are therefore readily volatilized from water and wet soils.

*Hexachlorocyclohexanes* (*HCH*)

Technical HCH consists of a number of isomers: α-HCH, β-HCH, and γ-HCH (also called lindane). The approximate composition of technical HCH is 55-70% α-HCH, 5-14% β-HCH, 10-18% γ-HCH, and δ-HCH and impurities. Lindane (γ-HCH), the most biologically active insecticidal isomer, is the only form of HCH currently used in its pure form in North America, Japan and Europe, where it is used mainly in seed treatment (Bidleman *et al.* 1989). Other isomers have been banned for use in the USA and most other circumpolar countries since the late 1970s. Technical HCH is still used in China as an insecticide on hardwood logs and lumber, seeds, vegetables and fruits, and on existing buildings and structures. Lindane and other HCH isomers are relatively persistent in soils, with half-lives generally greater than one year (Wauchope *et al.* 1992). HCH is much less bioaccumulative than other organochlorines because of its relatively low lipophilicity (log $K_{ow}$ = 3.8) and short half-life in biota (Niimi 1987). Relatively high H's and $VP_L$'s characterize the HCH isomers as volatile compounds capable of long-range transport in the atmosphere. The α-HCH isomer is more volatile and has a higher Henry's Law constant than the other isomers.

*Mirex*

Mirex is of interest because of its high $K_{ow}$ and its persistence. It was used as an insecticide and fire retardant, mainly in the USA and Canada. Its presence in the Lake Ontario

food web has been well documented. Mirex is extremely persistent in soils and sediment with an estimated 'field half-life' of five to ten years (Augustijn-Beckers *et al*. 1994). Although mirex has a very high molecular weight, it has the physical properties of a relatively volatile compound ($VP_L$ = 4.76 Pa; H = 52 Pa $m^3$/mol) capable of undergoing long-range transport. High levels of mirex are found in Lake Ontario and St. Lawrence River biota as a result of past industrial releases into the Niagara and Oswega Rivers during its manufacture. Its presence in the Arctic at low levels is consistent with its volatility and persistence.

*Dieldrin*

Dieldrin was mainly used as a soil insecticide. It is no longer manufactured in Canada and the USA, and its use is now restricted for termite control. Manufacture in Europe, especially for export to developing countries, continued until the late 1980s. It is also a degradation product of aldrin, also no longer in use in circumpolar countries. It is extremely persistent in soil (half-life greater than seven years) and has a long half-life in biota (Howard 1991). It is the most potent carcinogen of the major organochlorine pesticides.

*Dichlorodiphenyltrichloroethane* (*DDT*)

DDT was introduced in 1945 as an insecticide. The technical product consists of 4,4'-DDT (or *p, p*'-substituted) and its *o,p*'-DDT isomer as well as their dechlorinated analogs (*p,p*'- and *o,p*'-DDD). Its use has been restricted in Canada, the USA, and western Europe for nearly two decades, however, it is used in pest control programs in southern Asia, Africa, and Central and South America (Voldner and Ellenton 1988) and may be used in China and Russia. DDT, especially its metabolite *p,p*'-DDE, is extremely persistent in soils and sediments and has a long half-life in biota. DDT-related compounds have lower H's (1-10 Pa $m^3$/mol) and $VP_L$'s ($1.5 \times 10^{-4}$ - $5 \times 10^{-3}$ Pa) compared to other more persistent, semi-volatile OCs such as chlordane and PCBs. Levels of DDT and its principal metabolite, DDE, have decreased in fish and wildlife of western Europe, North America, and Japan in the past 15 years due to bans on use.

#### 6.1.1.2.2. **Less persistent chlorinated pesticides**

A large number of chlorinated organic chemicals are still registered for use as pesticides in circumpolar countries and elsewhere in the world. Many of these pesticides are nitrogen- and phosphorus-based, while others are analogs of the persistent OCs but with oxygen and sulfur substituents. These compounds are characterized by low biomagnification in food webs (due to degradation and elimination by biota), and in most cases relatively short half-lives in water, soil, and sediment. Nevertheless, these less persistent organochlorines have been shown to be present in the Arctic, especially in air and seawater. Their presence reflects large-scale use (in some cases greater than 1 000 000 kg per year), combined with semi-volatile characteristics similar to the persistent organochlorines. It is likely that these current use pesticides are more persistent in most Arctic abiotic matrices than in temperate zones due to low temperatures and limited sunlight, although little is known about this. Examples of currently used chlorinated pesticides detected in Arctic environments are atrazine, endosulfan, chlorpyrifos, chlorothalonil, tetra- and pentachlorophenol (as the anisole), and methoxychlor. A number of other current use, but non-chlorinated, pesticides have also been detected in Arctic air, seawater, and snow, including the organophosphate terbufos, the phenylamide herbicide metolachlor, and the dinitroaniline herbicide trifluralin (Chernyak *et al*. 1996).

Atrazine (1-chloro-3-(ethylamino)-5-(isopropylamino)-s-triazine) is a widely used herbicide, especially on maize (corn). It is currently banned in Norway, Sweden, and Germany, but heavily used in the USA (29 000 000 kg in 1988; Aspelin 1994) and Canada. Atrazine has a relatively low subcooled liquid vapor pressure ($VP_L = 1.4 \times 10^{-3}$ Pa) and a very low Henry's Law constant ($2.9 \times 10^{-4}$ Pa $m^3$/mol) compared with many insecticides such as endosulfan and lindane. Nevertheless, based on air and precipitation measurements, it has been shown to undergo regional long-range transport from major use areas in the USA and Europe (Richards *et al*. 1987, Buser 1988, Muir and Grift 1995). Atrazine was detected at low levels in ice and air in the Bering Sea by Chernyak *et al*. (1996).

Endosulfan is a very toxic insecticide, still widely used against a variety of insects especially on high value crops. Isomers of endosulfan are contaminants in air, water, sediments, soil, fish, other aquatic organisms, and food. The physical properties of endosulfan are characteristic of a compound capable of long-range transport (H = 2.98 Pa $m^3$/mol; $VP_L = 5 \times 10^{-3}$ Pa) (Suntio *et al*. 1988). It has a higher H than α-HCH, but lower $VP_L$. It has been measured and detected in the Arctic in a few studies, including recent reports in Bering/Chukchi Sea water and Arctic air by Chernyak *et al*. (1996) and Bidleman *et al*. (1995b), respectively. An estimated 57 000 tonnes have been used globally since the mid-1950s (Barrie *et al*. 1992).

Pentachlorophenol (PCP) is a widely used fungicide, especially for wood preservation. It is banned in Canada and Scandinavia, and registered only for restricted use in the USA and western Europe. Some pentachlorophenol products also contain tetrachlorophenols. PCDD/Fs, particularly hepta- and octa-chlorinated dioxins, are trace contaminants in PCP. It is very persistent in water and sediments under aerobic conditions, but degrades rapidly under anaerobic conditions such as occur in flooded soils (Augustijn-Beckers *et al*. 1994). PCP has limited capability to volatilize from water (H = 0.044 Pa $m^3$/mol; $VP_L$ = 0.11 Pa), especially near neutral pH where it exists in anionic form (Suntio *et al*. 1988, USEPA 1991). PCP is slowly dechlorinated in anaerobic sediments and is also converted to pentachloroanisole (PCA) by biomethylation. PCA is quite volatile (estimated $VP_L$ = 1 Pa; USEPA 1991b) relative to many other organochlorines. PCA is detectable in Arctic air, water, plants, and lower food web organisms.

Chlorothalonil (2,4,5,6-tetrachloro-1,3-benzenedicarbonitrile) is a fungicide widely used against moulds in cereal crops. Approximately 1 800 000 kg were used annually in the USA in the late 1980s (Aspelin 1994). Chlorothalonil is thought to have low bioaccumulation potential in aquatic environments (log $K_{ow}$ = 2.9; Tomlin 1994) and has an (average) field half-life of 30 days (Wauchope *et al*. 1992). Detection of chlorothalonil in Arctic air (Barrie and Muir unpubl. data 1996, Chernyak *et al*. 1996) and seawater indicates its potential for long-range transport. Chlorothalonil has a lower H (0.023 Pa $m^3$/mol) than other semi-volatile OCs such as lindane, but similar $VP_L$ (0.015 Pa).

Chlorpyrifos (O,O-diethyl-O-(3,5,6-trichloro-2-pyridinyl)phosphorothioate) is a widely used organophosphorus insecticide, first registered for use in the mid-1960s. Accurate global production figures are lacking, but 5 000 000 kg were used annually in the USA in the late 1980s (Aspelin 1994). Chlorpyrifos has a relatively high Henry's Law constant (1.75 Pa $m^3$/mol), similar to endosulfan, indicative of a compound capable of partitioning from water to air. It is considered moderately persistent in agricultural environments with field half-lives ranging from 12-90 days (Wauchope *et al*. 1992).

Methoxychlor (1,1,1-trichloro-2,2-bis(4-methoxyphenyl)ethane) is a widely used chlorinated insecticide, used for control of a variety of insect pests. It has a similar structure to DDT, but has much lower biomagnification potential because it can be metabolized by most higher organisms. Methoxychlor is relatively persistent in soils (estimated half-life 120 days; Wauchope *et al.* 1992). Methoxychlor has a Henry's Law constant of 0.6 Pa $m^3$/mol, similar to many other semi-volatile organochlorines such as lindane, but a much lower $VP_L$ (estimated $8.3 \times 10^{-4}$; USEPA 1991).

#### 6.1.1.3. **Other pesticides**

##### 6.1.1.3.1. **Tributyltin (TBT)**

Tributyltin (TBT) is a broad-spectrum algicide, miticide, fungicide, and insecticide (Aboul Dahab *et al.* 1990). TBT and other organotin compounds were first used in agriculture; subsequently TBT has had wide application as a marine antifoulant starting in the 1960s. Its most important entry route to the sea is directly from boats, aquaculture pens, moorings, and industrial cooling pipes to which products containing TBT have been applied, but it may also enter the sea in runoff from agricultural areas, from boat repair yards, and through municipal waste water and sewage sludge. TBT is found to provide effective protection for boat hulls at release rates less than 4 $\mu g/cm^2$/day and has been a popular antifoulant because it maintains its efficacy for up to five years compared to about three years for other conventional applications.

Once released to water, TBT is degraded by sequential debutylation to dibutyltin (DBT), monobutyltin (MBT), and eventually to relatively non-toxic inorganic tin compounds, primarily by organisms. Sunlight irradiation may also contribute to TBT degradation. The degradation time in water is short, with half-lives reported from days to a few weeks (Stewart and de Mora 1990, Dowson *et al.* 1993). TBT may be concentrated in the sea surface microlayer (Dobson and Cabrindenc 1990) and is strongly particle-reactive with partition coefficients reported to be as high as $10^3$-$10^4$ (Langston and Pope 1995). The breakdown of TBT in anaerobic sediments is much slower than in water (half-life of two or more years; Clark *et al.* 1988). Therefore, contaminated sediments are potentially an important environmental reservoir for TBT, which can continue to provide a source long after the industrial use of TBT has been curtailed.

Many of the developed countries now partially regulate the use of TBT (France in 1982; USA, 1986; UK, 1987; Canada and New Zealand, 1989; Europe, 1991). Regulations vary, but generally, only controlled release formulations are permitted and TBT-based antifoulants are prohibited for boats smaller than 25 m (Table 6·1).

## 6.2. Sources of persistent organic pollutants

The distribution of POPs in the Arctic is determined by the spatial distribution of the sources (this section), and physical and chemical properties of the compounds (section 6.1.1), combined with global atmospheric circulation patterns, removal by precipitation, interactions with the Earth's surface (see chapter 3), and biological pathways (section 6.3).

### 6.2.1. Pathways

POPs can be transported to and within the Arctic via several pathways and in different media. These pathways are described in detail in chapters 3 and 4 and are briefly reviewed here.

Persistent OCs of concern in the Arctic mainly originate in temperate and warmer areas of the world. The pathways followed by individual molecules to the Arctic and ultimately to Arctic organisms, can be complex. For example, a common pathway is: volatilization from soil or plant surfaces → atmospheric transport → deposition to the terrestrial environment on snowpack → snowmelt → river transport to lakes or estuaries → transport to the ocean → ocean current transport in ice or water within the Arctic. Repetitive revolatilization is also a major factor increasing the complexity of pathways. The vast majority of these contaminants remain in the abiotic environment. However, a small fraction can be transferred to biota by direct exposure through water and/or biomagnified in complex food webs or by maternal transfer. Although the total quantities of OCs in biota are very small compared to the quantities in the abiotic environment, significant bioaccumulation occurs in some parts of the food web resulting in elevated levels in top predators, including humans.

In general, as discussed in chapter 3, air in the northern hemisphere flows parallel to isobars in a counterclockwise direction around low-pressure systems (cyclones) and clockwise around high-pressure systems (anticyclones). The lower tropospheric circulation of the northern polar region is enhanced during winter (January-May) by semi-stationary high-pressure systems over continents and low-pressure systems over the northern Pacific and Atlantic Oceans. In particular, the intense continental Siberian high-pressure cell tends to force air on its western side northward into the Arctic. Airborne pollution from Siberian and European sources is injected into the far north by this mechanism. The high-pressure ridge over central portions of northern Canada tends to drive Arctic air southward. The mean flow in winter is out of Eurasia into the Arctic and out of the Arctic into North America. Transport times of pollutants via air currents can be fairly rapid, and complete mixing in the troposphere of the northern hemisphere can occur within one month (Ballschmiter and Wittlinger 1990).

Measurements of Arctic airborne particulate matter described by Rahn (1981), and others (Leaitch *et al.* 1984, Maenhaut *et al.* 1989, Barrie *et al.* 1992), identified metals in ratios characteristic of emissions from specific industrial areas in Europe and Asia. This is consistent with the generic meteorological pathways described above and suggests that transport of particulate matter into the Arctic is primarily from Eurasian sources.

Significant quantities of organochlorine contaminants are present in the atmosphere, the surface oceans, and the terrestrial environment. The volume of the accessible terrestrial environment of soils and vegetation is relatively small (effective depth of 5-30 cm) compared to that of the atmosphere (≈10 000 m deep) and the oceans (effective depth of accumulation 200-500 m). However, the hydrophobic nature of most POPs results in their preferential partitioning into soil and plant surfaces from water or air. Although fish, marine and terrestrial mammals, and birds are critical from the viewpoint of human exposure and ecological effects, the biota probably play a minor role in the overall mass balance on global, hemispheric, and regional scales. The possible exceptions are microbial conversion in soils and partitioning to planktonic organisms in fresh and marine waters. Vegetation may also play a key role in atmosphere-surface transport, especially in forested systems (Simonich and Hites 1995). Snow and ice are significant components of the Arctic ecosystem, but the present ability to describe exchange between falling snow, snowpack and the atmosphere and terrestrial environments is primitive. Recent measurements of air-ice surface partitioning (Hoff *et al.* 1995) suggest that snow may be significant both as a medium of transport and as a reservoir.

Atmospheric transport pathways for semi-volatile organics are often divided into two types: 'one-hop' and 'multi-hop' pathways, although this is a simplification of events. For compounds emitted to the atmosphere, transported, and deposited to the surface never to return to the atmosphere ('one-hop' compounds), the dispersion of the contaminant would be simply defined by its initial source distribution, its lifetime in the atmosphere, and atmospheric circulation. This type may apply for the relatively non-volatile POPs, such as the higher chlorinated PCB congeners, which tend to be particle-associated at low temperatures. The pathways of these constituents follow that of Arctic haze from mid-latitudinal sources into the Arctic (Barrie 1986, 1994, 1996). This type is also distinguished by longer atmospheric residence times in winter (≈20-30 days) compared to summer (≈3-7 days) and stronger north-south transport into the Arctic, particularly from Eurasia, in winter than in summer (Barrie *et al*. 1997).

A compound that has a tendency to re-enter the atmosphere after initial deposition to the Earth's surface, can move through the environment in a series of 'hops'. Processes that facilitate this type of transport include volatilization from the surface under warmer temperatures than existed at the time of initial deposition, rapid exposure of ocean or freshwater to the atmosphere after prolonged periods of ice cover, resuspension of dust by wind, and re-distribution and morphological changes to fresh snow by wind. Most organochlorines fall into this group (Barrie *et al*. 1997). The polar regions are potentially cold traps for these compounds. Wania and Mackay (1993) and Strand and Hov (1996) have developed models to simulate this process and have been able to qualitatively reproduce observed patterns and concentrations of contaminants in the different compartments of the Arctic. Primarily due to their cold temperatures, the near surface waters of the oceans serve as sinks for some OCs in these models.

Rivers can be an important pathway of contaminants. North-flowing rivers drain an area of 10 000 000 km$^2$ of northern Eurasia and North America from as far south as 50° latitude (Barrie *et al*. 1992, Macdonald and Bewers 1996) (section 6.6.3.1). In draining vast areas of land, they may also carry significant quantities of contaminants, often from industries, to the ocean surface layer. Marine areas affected by rivers are highly productive and tend to have more detrital food chains.

Contaminants are also transported by ocean currents. Ocean circulation is driven by a combination of various forces (e.g., tides, wind stress, mixing of water masses) and a particular force may dominate in a particular area. Within the Arctic Ocean, the main surface circulation features are the clockwise circulation of the Beaufort Gyre, and the Transpolar Drift, which flows from Siberia, across the pole, and then southward to exit as the East Greenland Current (chapter 3). The major currents whereby water is exchanged between the Arctic Ocean and other oceans are found in Fram Strait. The West Spitsbergen Current flows northward off the west coast of Spitsbergen, transporting Atlantic water from the Norwegian Sea into the Arctic Ocean (chapter 3).

#### 6.2.1.1. Particle/gas partitioning and atmospheric deposition processes

Exchange of organic compounds between the atmosphere and the Earth's surface takes place by rain and snow scavenging of gaseous and particulate species and transfer of gaseous compounds across air–water interfaces (chapter 3). Flux equations used to describe these processes necessarily take into account the partitioning of the compound between the particle and gas phases in air (Cotham and Bidleman 1991). Estimates of the atmospheric phase distribution are frequently made with the Junge-Pankow adsorption model (Pankow 1987):

$$\phi = c\theta / (VP_L + c\theta)$$

where $\phi$ is the fraction of the compound adsorbed to aerosols, $VP_L$ is the sub-cooled liquid-vapor pressure (that is, the vapor pressure of the chemical in a hypothetical liquid state below its melting point) at the ambient temperature, $\theta$ is the aerosol surface area in cm$^2$ per cm$^3$ air, and the parameter c, often taken as 17.2 Pa-cm, depends on the thermodynamics of adsorption and the surface area occupied by a sorbate molecule. Values of $\theta$ that are representative of urban, rural, and clean air regimes are given by Bidleman (1988).

The Junge-Pankow model has been applied to Arctic aerosols by Cotham and Bidleman (1991) and Patton *et al*. (1991), using values of $\theta$ measured in the atmospheric monitoring program at Alert on Ellesmere Island, Canada. The adsorption of vapor-phase substances to Arctic aerosols is most important during the winter haze season, when the air temperature is low (approximately –30 to –40°C) and the aerosol concentration is relatively high. Generally, substances that have $VP_L \leq 10^{-3}$ Pa at Arctic winter temperatures will be 50% or more in aerosol form under typical haze conditions. During winter, the expected percentages of γ-HCH, *p,p'*-DDE, and benz[a]anthracene on haze aerosols are 2, 50, and 90%, respectively. In summer, when the Arctic air temperature warms to 0°C and above and the aerosol concentration is less than one-tenth that of winter values, these percentages fall to < 0.001, 0.5, and 4%, respectively, for the three compounds. Thus, many of the organic contaminants of interest are transported in the gas phase during summer, but become sorbed to some extent to haze aerosols in winter.

### 6.2.2. Global sources

A database of historical, present, and predicted global usage or sale of selected persistent pesticides including aldrin, dieldrin, endrin, technical HCH, lindane, DDT, chlordane, endosulfan, heptachlor, and toxaphene was prepared by Voldner and Li (1993, 1995), Li *et al*. (1996, 1997), and Barrie *et al*. (1997). This information was linked with the global distribution of agricultural activities to determine usage distribution. The reported global cumulative usage for selected pesticides is presented in Table 6·2. When usage was estimated by interpolating to non-reporting times and areas, the

Table 6·2. Global cumulative usage of selected pesticides for various periods of time (Barrie *et al.* 1997).

| Pesticide | Usage, tonnes | Period | Source |
|---|---|---|---|
| *Reported* | | | |
| DDT | 1500000 | 1948-1993 | Voldner and Li 1995 |
| Technical HCH | 550000 | 1948-1993 | Voldner and Li 1995 |
| Technical lindane | 720000 | 1948-1993 | Voldner and Li 1995 |
| Toxaphene | 450000 | 1948-1993 | Voldner and Li 1993 |
| Technical HCH | 40000 | 1980 | Li *et al.* 1996 |
| | 29000 | 1990 | |
| Technical lindane | 5900 | 1980 | Li *et al.* 1996 |
| | 4000 | 1990 | |
| α-HCH | 28000 | 1980 | Li *et al.* 1996 |
| | 20400 | 1990 | |
| γ-HCH | 11900 | 1980 | Li *et al.* 1996 |
| | 8400 | 1990 | |
| *Interpolated* | | | |
| DDT | 2600000 | 1950-1993 | Voldner and Li 1995 |
| | 990000 | 1970-1993 | |
| Toxaphene | 1330000 | 1950-1993 | Voldner and Li 1993 |
| | 670000 | 1970-1993 | |

expected usage was considerably larger (1.7 and 3 times more for DDT and toxaphene, respectively) (Voldner and Li 1993, 1995).

A number of countries have made attempts to identify sources of PCDD/Fs and to estimate the amounts emitted annually to air, and in some cases even to water and in wastes. The Netherlands has estimated its PCDD/F emissions to air to be 484 g TCDD equivalents (TEQ) for 1990 (de Koning *et al*. 1994). The United Kingdom estimated its PCDD/F emissions to air to be 560-1100 g TEQ/y (HMIP 1995). The Federal Republic of Germany has estimated its PCDD/F emissions to air to be 1166-1646 g TEQ/y for 1985-1990 and 452-656 g TEQ/y for 1993-1995 (Lahl and Zeschmar-Lahl 1995). Austria estimated its emissions to be 50-320 g TEQ/y in 1987/88 and Japan estimated its emissions to be 4000-8400 g TEQ/y for 1990 (Liem and van Zorge 1995). For all surveys, the major PCDD/F sources to air are combustion-related, including municipal waste incinerators, hospital incinerators, metallurgical processes (sintering, smelting, die-casting, etc.), automobiles, and woodburning. A major source to water is bleached kraft pulp and paper mills using elemental chlorine. All countries with these types of industries have emissions of PCDD/Fs to air and water, but it is not possible to quantify the global amounts released.

Several surveys indicate declines in emissions for some countries, which is primarily due to improved technologies for flue gas cleaning, optimized combustion technology for complete combustion in incinerators, and other measures that have been taken to reduce PCDD/F formation in high temperature processes. Continued implementation of these technologies will lead to continued declines. PCDD/F levels have probably also declined due to bans on PCBs, chlorophenols, and phenoxy acid herbicides that contained PCDD/Fs. Some international agreements exist where countries aim to reduce PCDD/F emissions. One such agreement, made within the North Sea Conference, instructed parties to reduce dioxin inputs to the North Sea by 70% or more between 1985 and 1995 (North Sea Conference 1995).

#### 6.2.2.1. HCH case study

The most extensive database for a persistent organic pollutant in the abiotic environment exists for technical HCH and its isomer γ-HCH (lindane). The HCH mass balance in the Arctic Ocean is described in section 6.6.4.3.1.

The leading HCH-utilizing countries are listed in Table 6·3. In 1980, the annual utilization of α-HCH in India, China, and the former Soviet Union accounted for more than 95% of the total global α-HCH consumption. By 1990, although α-HCH usage continued to increase in India, the former Soviet Union, and Mexico, usage decreased dramatically among other countries. γ-HCH consumption in India, China, and the former Soviet Union in 1980 accounted for approximately 90% of the global usage. This dropped considerably by 1990 due to China's ban on technical HCH usage (Li 1996, Barrie *et al*. 1997, Li *et al*. 1997).

### 6.2.3. Sources in circumpolar countries

The majority of circumpolar countries have taken action to reduce the inputs to the environment of the POPs discussed in this chapter. This information has been summarized in Table 6·1. In some cases, certain POPs were never used, in other cases, they were used until bans were put in place. Certain POPs are still allowed for restricted use, for example the use of TBT on ocean-going ships longer than 25 m.

Table 6·3. Estimated annual usage of α-HCH and γ-HCH in 1980 and 1990 for the top-consuming countries (Li 1996, Li *et al*. 1997, modified from Barrie *et al*. 1997).

| | Usage, tonnes/year | |
|---|---|---|
| α-HCH | 1980 | 1990 |
| China | 200521 | 0 |
| India | 15100 | 32900 |
| Former Soviet Union | 11718 | 17528 |
| Former East Germany | 2626 | 84 |
| Argentina | 1470 | 0 |
| Sri Lanka | 770 | n.a. |
| Turkey | 628 | n.a. |
| Nigeria | 358 | n.a. |
| Mexico | 105 | 1218 |
| South Korea | 84 | 0 |
| γ-HCH | 1980 | 1990 |
| China | 42969 | 100 |
| India | 3376 | 7050 |
| Former Soviet Union | 2511 | 3756 |
| Italy | 1580 | 600 |
| France | 1172 | 1863 |
| Former East Germany | 563 | 18 |
| Argentina | 315 | n.a. |
| United States | 268 | 114 |
| Canada | 200 | 284 |
| Sri Lanka | 165 | n.a. |
| Turkey | 135 | n.a. |
| Nigeria | 77 | 397 |
| Mexico | 23 | 261 |
| South Korea | 18 | n.a. |
| Former Yugoslavia | n.a. | 151 |
| Spain | n.a. | 96 |
| Pakistan | n.a. | 3 |

n.a.: not available.

In the USA, lindane is no longer produced, but use is allowed for structural treatment, hardwood logs and lumber, moth sprays, seed treatment, dog shampoos, homeowner ornamentals, and other household uses. Chlordanes are only allowed for fire ant control in power plants. DDT was banned in 1972, however, due to a loophole in the pesticide law, it was allowed as an inert ingredient in another pesticide, Dicofol, which could contain up to 12% DDT. As of 1986, all products containing more than 0.1% DDT were cancelled in the USA (Sherman 1994). Dicofol was also banned in Sweden in 1991. Lindane use is restricted in Canada, where its major use is as a seed dressing for cereal crops. Mirex was never registered as a pesticide in Canada, but was used as a flame retardant between 1960 and 1970.

In Iceland, dieldrin, HCB, α- and β-HCH, mirex, and toxaphene use has not been allowed since 1975 and available documentation suggests that these were never used (S. Gisladottír, Hollustuvernd Ríkisins, pers. comm.). Chlordane was used from the 1940s up until 1975, when use was prohibited. DDT was used as a pesticide up until 1975. After 1975, it has been used as a drug for scabies in horses. Although one lindane product is still registered, it has not been used for several years. Lindane is also used to treat head lice. In Norway, lindane was used before it was banned in 1991, but no information is available for α- and β-HCH. DDT was used fairly extensively until 1969. Limited use was allowed until 1988 for trees in plant nurseries. Chlordane was marketed and used until 1967 and dieldrin and aldrin were marketed and used up to 1970.

In Russia, household and institutional insecticides containing concentrated solutions of DDT and HCH are still being used (Valery Klopov, pers. comm.).

Although PCB formulations have been banned for open use in most circumpolar countries, they were used in several countries until recently in closed systems. A few circumpolar countries still allow PCB use in closed systems that existed prior to bans.

In Norway, approximately 1500 tonnes of technical PCB have been used. About 650 tonnes of PCB are still contained in products that are still in use. These include 350-410 tonnes in sealing compound in windows, 70 tonnes in joint sealants between concrete elements, 200 tonnes in low voltage condensers in lighting, and 10 tonnes in high voltage condensers/transformers. Approximately 850-880 tonnes have been disposed of, stored, or destroyed. It is believed that PCB-containing products corresponding to 400-600 tonnes of technical PCB have been disposed of in such a way that it may represent a threat due to eventual/possible leakage to the environment. However, plans are being made to minimize this threat (SFT 1996).

In Sweden, approximately 8000-10 000 tonnes of PCB were imported between 1957-1980 (Reutergårdh 1988) and probably more than half of this amount was re-exported in goods. The major use was in condensers and transformers. Open use of PCBs was banned in 1972, and new use cancelled in 1978, however, PCBs were allowed in existing closed condensers and transformers until 31 December 1994, after which they were banned completely. PCB was also used widely in many building products such as paints, plastics, window putties, window sealing compounds, and sealants that are still in use. PCB-containing joint sealants were the main product used, but floor paint and window sealing compounds were also used extensively. The joint sealants were used between prefabricated building elements in apartment buildings erected from 1950-1972 in Sweden. The polysulfide Thiokol-rubber joint sealants have been found to contain up to 20% PCB, which was used as a plasticizer. It is estimated that this is equivalent to 100-500 tonnes of PCB in unintended open use in Sweden (Hammar 1992). Current studies show that the PCBs are leaking from the joint sealants to outside air (Jansson *et al*. 1997). The emission rate is estimated to be 0.1-0.2% or 0.1-1 tonnes/y to air. The sealant itself is also eroding, leading to elevated PCB levels in soil near the buildings.

Some sealant has ended up in landfills when buildings were renovated before it was recognized that the sealant contained PCB, making the wastes an added source to the environment. As the sealant was imported to Sweden, it is not unlikely that other countries, including the circumpolar countries, also have buildings with PCB-containing sealants. It is estimated that a further 50-100 tonnes of PCB are still in use in existing insulated window-glass and 20-30 tonnes in floor paints in buildings in Sweden (KEMI 1996a).

In the USA and in Canada, new use and open use of PCBs are forbidden, but they are still used in closed systems that existed before the ban took effect. Canada is currently phasing out these PCBs. Landfills and dumps are also probably sources of PCB, due to previous disposal of PCB-containing products. After banning PCB in 1988, Iceland decommissioned all equipment and products containing PCB and sent them abroad for destruction.

Three circumpolar countries have carried out surveys to estimate PCDD/F emissions – Sweden, the USA, and Norway. For Sweden, PCDD/F emissions to air for 1990 were estimated to be 32-115 g TEQ, and for 1993, 22-88 g TEQ (C. de Wit unpubl. data). Estimated PCDD/F emissions to air for the USA are 3774-34 278 g TEQ/y (Schaum *et al*. 1994). For Norway, PCDD/F emissions to air are estimated to be 63 g TEQ for 1994 (B. Kvæven, Norwegian State Pollution Control Authority, pers. comm.). Emissions to water in Norway have declined from 504 g TEQ in 1985 to 3 g TEQ in 1994. Sweden, Norway, and Denmark are parties to the North Sea Conference.

Canada is currently undertaking a re-evaluation of PCDD/F sources, but current information shows that paper and pulp mills have reduced their annual emissions from 350 g TEQ in 1989 to 9 g TEQ in 1994 (J. Van Oostdam, Health Canada, pers. comm.). Other sources are hospital incinerators and, to a lesser extent, municipal waste incinerators.

Although most circumpolar countries have banned HCB as a pesticide, it is also produced as a by-product during thermal processes. Two known large point sources of HCB in Norway are a magnesium factory in Telemark in the southeast and a nickel smelter on the south coast. These released 126 tonnes HCB to air and 5 tonnes to water as late as 1994, but extensive measures reduced these releases considerably by 1995 (B. Kvæven, Norwegian State Pollution Control Authority, pers. comm.).

Besides the pesticides listed in Table 6·1, Sweden has banned the following current-use pesticides that have been detected in the Arctic: atrazine (1989), chlorothalonil (1994), pentachlorophenol (1978), methoxychlor (1990), and trifluralin (1990). Use of chlorpyrifos is severely restricted (KEMI 1996b). Terbufos and metolachlor have never been registered as pesticides in Sweden. Atrazine, chlorpyrifos, metolachlor, trifluralin, chlorothalonil, methoxychlor and terbufos are currently in use in the USA Pentachlorophenol use is severely restricted in the USA. In Canada, atrazine, chlorpyrifos, chlorothalonil, methoxychlor, terbufos, metalochlor, and trifluralin are all currently in use, but pentachlorophenol use was banned in the early 1990s.

In Finland, terbufos, metolachlor, and tetra/pentachlorophenol as anisoles have never been used. Use of atrazine and methoxychlor has ceased voluntarily. Use of chlorpyrifos and chlorothalonil is restricted. Trifluralin is currently in use in Finland (J. Malm, Finnish Environment Institute, pers. comm.). Norway has banned atrazine (1988) and trifluralin (1992) and the following are not approved: chlorpyrifos, tetra/pentachlorophenol, methoxychlor, terbufos, and metolachlor. Chlorothalonil is currently in use and endosulfan is allowed only in integrated pest management (B. Kvæven, Norwegian State Pollution Control Authority, pers. comm.).

In Iceland, atrazine was used between 1976 and 1992. Chlorpyrifos is not a registered pesticide, but some exemptions for use have been made in 1995 and two applications for registration are currently being handled. Chlorothalonil is not registered, but exemption for use was given in 1995. Terbufos and metolachlor have never been registered or used in Iceland. Trifluralin is registered and used. Pentachlorophenol was banned from Iceland in 1996 (S. Gisladottír, Hollustuvernd Ríkisins, pers. comm.).

### 6.2.4. Local/regional sources within the Arctic

Sources of POPs in the Arctic are not well documented in most cases, but could be important especially in terms of exposure of humans and wildlife living near a use site. In most cases, Arctic sources are the result of accidental spills or deliberate and inappropriate disposal of contaminants. Combustion, especially of municipal garbage, is a common sight in the Arctic and could be a source of PCDD/Fs and HCB as well as PAHs. The topic of combustion sources for PAHs within the Arctic is dealt with in chapter 10. Combustion or deliberately disposed chemicals are often distributed over a wider area than the initial disposal areas through the interaction of different dispersal processes. In other cases, pesticides have deliberately been used for insect control (section 6.2.4.4).

#### 6.2.4.1. **PCBs at military sites**

PCBs were in use in electrical equipment at the Arctic radar stations in North America, Greenland, and Scandinavia, and presumably also in Russian facilities. In North America, disposal practices resulted in PCBs entering the local terrestrial and aquatic environments either through disposal at landfills, down-the-drain disposal, and accidental or intentional spills. There were few concerns about the disposal of the PCB fluids during much of the time of operation of the stations. Similar problems may have occurred in Russian facilities, but they are currently undocumented.

*Canada*

The DEW (Distant Early Warning) Line, started in 1955, consisted of 63 radar stations across Alaska, Canada, and Greenland along the 66th parallel. Over the life of the DEW Line, which was replaced in the early 1990s by another radar system with fewer stations (the North Warning System, NWS), stations were closed and added, so they were used to varying degrees (Fletcher 1990). The largest change occurred in 1963 when all 31 gap-filler or 'I' (Intermediate) sites, 20 of which were in Canada, were closed. A large, but as yet unquantified portion of the estimated 30 tonnes of PCBs imported to the radar sites, may be fugitive in the environment.

An extensive effort to assess the potential for site contamination was undertaken (Holtz *et al*. 1987), which addressed primarily drums of contaminated oils, discarded electronic equipment, and contaminated soils and other material. However, ongoing environmental assessment and impact studies at the northern radar stations and at Arctic reference sites have also resulted in sampling and analysis of over 3500 soil, 1600 plant, and various small mammals and marine invertebrate and fish samples, from abandoned and soon-to-be abandoned DEW Line and NWS sites across Canada (Reimer *et al*. 1991, 1993a, 1993b, 1993c, 1994, Dodd and Reimer 1992, Dushenko and Reimer 1994, Bright *et al*. 1995a, 1995b, 1995c). In general, levels in soils and plants are 1-3 orders of magnitude higher than at remote sampling sites. Results from the soil/plant studies are discussed elsewhere (section 6.6.2.1).

Concentrations in samples taken from remote locations (more than 20 km distant), when compared to local background locations, provided evidence for short-range redistribution of PCBs (Bright *et al*. 1995a). In addition, the PCB congener signatures for background samples correlated well with PCB signatures from actual contaminated locations at radar sites. At more remote sites, the congener signature changed, and could be attributed to atmospheric transport, which contains a relatively higher proportion of more volatile congeners.

Comparison of PCB levels and congener profiles in contaminated soil from selected DEW Line sites and in background soils (Bright *et al*. 1995a), with levels in fresh snow, snowpack, glacial snow, and background soils and sediments from across the Canadian Arctic (Gregor 1995), suggests that the impact of the DEW Line sites is limited to the immediate vicinity of the site. At Cambridge Bay, Bright *et al*. (1995a) concluded that PCBs were found only at a distance of 5 km or less from the site. The studies suggest that there is a 'halo' of impacted soils around each contaminated site, which gradually blends into the background soil PCB signature determined by atmospheric deposition. Gregor (1995) estimates that while approximately 300 km$^2$ may be affected at each site, this represents less than 0.2% of the total surface area of the Yukon and Northwest Territories.

A study of the use, transportation, and disposal of PCBs in the Yukon (Canada) concluded that the major use of PCBs in the Yukon was limited to the wartime construction era in the period from 1941 to 1946 (Nordin *et al*. 1993).

*USA*

Over 600 formerly used defense sites (FUDS), including 49 DEW Line sites, have been identified in the state of Alaska for possible environmental assessment and remediation (US Department of Defense 1992). Most of the sites are located within the broad context of the USA Arctic as defined under the Arctic Research and Policy Act. Contaminated sites include those used for military training exercises, those used for major industrial operations and production facilities, and those where only minimal impact occurred. Based on preliminary contamination and site inspection, a vast majority of FUDS may no longer have any significant contamination. However, some of the existing military sites, such as Eielson Air Force Base, Fort Wainwright, and Adak Naval Air Station, are heavily contaminated (petroleum and lubricants, heavy metals, chlorinated solvents, transformer oils containing PCBs, pesticides, etc.) and listed in the National Priority List for long-term clean-up and remedial action.

*Denmark* (*Greenland*)

Contamination associated with military activities was also investigated during the 1980s near the Thule Air Base in northwest Greenland. PCBs were monitored in sediments in the Wolstenholme Fjord in Greenland to investigate the influence of the Thule Air Base, which was the suspected source of these contaminants (Kjølholt and Hansen 1986). PCB levels in sediments were highest in surface samples (0-1 cm), ranging from 10 to 65 ng/g (dw), and showed a decreasing concentration with distance from the pier at the air base. These levels were much higher than in most Arctic marine sediments where PCB levels are generally below 1 ng/g dw (section 6.6.4.2.1).

*Iceland*

Iceland has ten abandoned military sites, which were used for radar surveillance and communication. Some were built in the 1940s and a few of these were closed after World War II, but others were built later. The last site was closed down in 1994. The level of possible local contamination has not been extensively studied, however, the information available indicates only minor pollution (PAME 1996). It is not clear if studies of POPs, particularly PCBs have been performed.

*Russia*

Military-industrial complexes exist in Murmansk, Arkhangelsk, and the Taimyr Autonomous Territory. No studies have been carried out pertaining to possible POP contamination, but such contamination, particularly for PCB is considered likely.

#### 6.2.4.2. **Other PCB sources**

Electrical capacitors and fluorescent light ballasts, manufactured prior to 1979, contained small quantities of PCBs. These may have been disposed of at military and industrial sites, including the DEW Line sites within the Yukon, and municipal landfills, as equipment was replaced at least until 1977. Contaminated soils exist at a number of sites in the Yukon while equipment burial or contamination is expected at other sites. Another possible source has been the use of waste oil, contaminated with PCB, to control dust on Whitehorse streets. It has not been possible to estimate the volume

of PCB lost to the Yukon environment (Nordin *et al.* 1993). Similar local sources could be present in other large northern communities in Canada.

As stated above, some buildings in Sweden have PCB-containing joint sealants that are leaking PCB to the environment. There are several small cities in the Swedish Arctic, and it is not unlikely that they also have buildings with PCB-containing joint sealants. This may also be true for the other circumpolar countries. However, currently no estimate can be made of the amounts of PCBs in sealants in buildings located specifically in the Swedish Arctic.

Larger urban settlements, particularly those along the Arctic coasts and major Arctic rivers discharge waste water containing domestic sewage and industrial effluents that end up in the limnic and marine environments. In some cases, solid waste is placed in landfills or is incinerated. Landfills may be rudimentary and due to the slow decomposition rate, there is a risk of long-term leaching of contaminants. The scale of local pollution from large Arctic towns and cities is not known, but it may be most acute in Russia, in harbors and ports such as Murmansk, Severomorsk, Arkhangelsk, Severodvinsk, Amderma, Dikson, and Salekhard (PAME 1996).

Leakage from solid waste disposal sites at the coal-mining settlements of Barentsburg and Longyearbyen on Svalbard is known to occur. The landfills probably contain industrial wastes as well as general garbage. Bottom sediments collected on the coast near these landfills have shown among other things, HCB contamination, and at Barentsburg, PCB contamination as well (Holte *et al.* 1996, PAME 1996). PCB has also been found in soil samples from the landfill at Ny-Ålesund on Svalbard.

Several circumpolar countries produce hydroelectricity from rivers in the Arctic, including Norway, Sweden, Finland, Iceland, the USA (Alaska), and Canada. A number of the power plants were built during the 1960s and early 1970s. No studies of PCB levels near hydroelectric plants have been done, although transformers at the plants most probably contained PCB at one time. Based on the experience from DEW line sites, it is probable that there have been leaks or spillage that have caused local contamination, but the extent of this can only be guessed. Trains running on electricity also had PCB-containing transformers, and leakage along rail lines in the Swedish Arctic has probably occurred, although no studies have been performed to check this. This may also have been a problem in other circumpolar countries with such trains.

PCBs may have also been used historically as hydraulic and drilling fluids in mines and at oil wells. A number of metal ore and coal mines have been, or are currently, active in the Arctic. These are found in Canada, Norway including Svalbard, Russia, Sweden, the USA, and Greenland. Tailings effluent has been discharged into the marine environment from several of these. Although they are known to be sources of heavy metals in the Arctic, the possible contamination by POPs from these sources has not been studied. Used drilling fluids have also been released in Alaska, Canada, and Russia (chapter 3), however, no information is available on possible POP contamination from these sources. Investigations of oil products delivered to Norwegian state-owned mining companies on Svalbard have shown that none of these contained PCB (SFT 1996).

Another potential source of PCB is transformers on oil drilling platforms. The oil well platform Piper Alpha released 5 tonnes of PCB into the North Sea when a fire destroyed it in 1988 (Wells *et al.* 1989). Oil exploration and exploitation is ongoing in the northern North Sea by, among others, Norway. Onshore wells are currently in use in the USA (Alaska), Canada, and Russia. Russia and Norway may be considering offshore oil development in the Arctic in the near future. PCB use on new platforms is unlikely because of use restrictions, however, older oil platforms and equipment could be potential sources if PCB-containing transformers or other equipment are still in use. This has not been studied. Ships and submarines, either those trafficking or those that have been dumped in the Arctic are also potential sources if they have or had transformers containing PCBs.

#### 6.2.4.3. **Specific PCDD/F sources**

As stated in section 6.2.2, the major sources of PCDD/Fs to air are waste incineration, most particularly where incomplete combustion occurs, wood burning and other combustion, and metallurgical industries. Such activities located in or near the Arctic are thus suspected as local sources of PCDD/Fs. In some cases, such local sources have been identified and studied.

##### 6.2.4.3.1. **PCDD/F contamination from smelters**

Known local PCDD/F sources in Arctic Sweden are iron ore pelletizing plants at Malmberget, Kiruna, and Svappavaara (C. de Wit unpubl. results). A source of air-bound PCDD/Fs to the Arctic, although not located within the Arctic itself, is the primary smelter of Rönnskärsverken in Skellefteå on Bothnian Bay.

The smelters and metallurgical plants on the Kola Peninsula, the Vorkuta area in the north Komi Republic, and the Norilsk area are suspected local PCDD/F sources in Arctic Russia. Suspected sources in Arctic Norway are a secondary iron and steel industry, an aluminum industry, and a ferroalloy industry. Suspected PCDD/F sources in Finland, Canada, and Alaska are waste incineration and wood burning for heating, and for Greenland, waste incineration.

Local and regional contamination by PCDD/Fs from the Syd-Varanger smelter works in Kirkenes, in Arctic Norway, was studied by Schlabach and Skotvold (1996a, 1996b). This smelter sinters iron ore, a process that is known to produce PCDD/F emissions. Marine sediments and biota were sampled at increasing distances from inner Bøkfjord, where the smelter works is located, as well as throughout the fjord system. Freshwater sediments were sampled at increasing distances from the emissions of the smelter works. Soil was sampled at several locations in the town of Kirkenes. The results of this study are described in more detail in sections 6.6.3.2.1.2, 6.6.3.3.3 and 6.6.4.2.2.

In brief, the top layer of sediment, as well as whitefish (*Coregonus*) from the lake nearest to the smelter works, were found to be markedly contaminated with PCDD/Fs. All the samples of marine sediments, including those taken in the vicinity of the plant, contained diffuse background levels of PCDD/Fs. Slightly elevated levels of PCDD/Fs were detected in the horse mussel (*Modiolus modiolus*) sampled in the vicinity of the plant, while those from the reference stations contained only background levels. The results of the study by Schlabach and Skotvold (1996a, 1996b) illustrate that combustion-related sources such as metal smelting are significant locally. The smelters on the Kola Peninsula and at Norilsk are probably also local PCDD/F sources in Russia. Overall, metal smelting and other combustion-related processes within the Arctic could be important contributors to the background level of contamination.

#### 6.2.4.3.2. PCDD/F contamination from chlorine-bleached kraft pulp and paper mills

Pulp and paper mills employing elemental chlorine for bleaching wood pulp are located within the Arctic Ocean drainage area. The pulp mills located in closest proximity to the Arctic Ocean are found in western Russia. Pulp mills in northern Alberta, on the Wapiti, Peace, and Athabasca Rivers also discharge into waters which reach the Arctic Ocean (over 2000 km north) via the Mackenzie River; pulp mills in northwestern Ontario and Manitoba are also within the Hudson Bay drainage area.

Studies in the Arkhangelsk area indicate local PCDD/F contamination along the Severnaya Dvina River and its tributaries where at least two chlorine-bleached kraft pulp and paper mills are located (Yufit and Khotuleva 1994). The PCDD/F concentrations appear to be low in the river sediments and water samples analyzed, indicating that these are not major PCDD/F sources to the Barents Sea via the White Sea. This study is described in more detail in section 6.6.3.2.1.3.

Studies of the emissions and ultimate fate of PCDD/Fs in the Peace-Athabasca River system were conducted from 1992-1996 (Pastershank and Muir 1995, 1996). PCDD/Fs were first detected in the effluent of three mills in the region in the late 1980s. Highest levels of PCDD/Fs were found in environmental samples within 50 km downstream from the mills on the Athabasca and Wapiti Rivers (Swanson *et al.* 1995, Pastershank and Muir 1996). PCDD/F levels returned to upstream (control site) levels in fish sampled more than 100 km downstream from the bleached kraft mill effluent source. All three mills upgraded their bleaching processes in the early 1990s and converted to the use of chlorine dioxide as a substitute for molecular chlorine by 1993. Following the introduction of these process changes, emissions of 2,3,7,8-TCDD and TCDF declined substantially (Swanson *et al.* 1995). Nevertheless, low concentrations of PCDD/Fs and other organochlorines (e.g., chloroveratroles), indicative of a chlorine-bleach pulp source, were found in the western basin of Great Slave Lake (Evans *et al.* 1996). Concentrations of PCDD/Fs in the lake sediments are discussed in more detail in section 6.6.3.2.1.2. The results from Great Slave Lake demonstrate that PCDD/Fs could be transported long distances in the Mackenzie River system. PCDD/Fs are also detectable in estuarine sediments in the Mackenzie Delta area (section 6.6.4.2.2; Macdonald unpubl. data 1996), although the sources of the contaminants at this site, which are more than 2000 km from the pulp mills, are likely to be diffuse.

#### 6.2.4.4. Pesticides

Persistent chlorinated pesticides have been used in the past in the Arctic in a variety of ways, especially for the control of insects such as biting flies and mosquitoes in or near populated areas. Other uses within urban areas may be important, such as use as rodenticides, treatment of stored grains and other foods, and medical uses. With the exception of agricultural areas at the southern extremes of the watersheds of the Mackenzie, Nelson, Ob, Yenisey, and other major north-flowing rivers in Russia, there has not been widespread use of pesticides for agricultural purposes in these watersheds.

In Canada, knowledge of pesticide usage in the Northwest Territories has been limited largely to anecdotal information, although there is evidence of DDT at DEW Line sites (Bright *et al.* 1995a). The main insecticide used in the Yukon for mosquito and black fly control in the late 1940s was DDT and it was first applied directly into the Yukon River in July 1948. Aerial spraying of DDT, and later ground fogging, to control mosquitoes was also conducted. There does not seem to be a complete, cumulative record of the total quantity of DDT applied over this period of time, although Nordin *et al.* (1993) estimated a total application of over 15.8 tonnes. Spraying of DDT continued until 1969 when DDT was replaced by other insecticides (Nordin *et al.* 1993).

Similar pest control activities occurred within the Arctic Ocean watershed in Canada, for example, treatment of the Athabasca and Saskatchewan Rivers with DDT, and later with methoxychlor, for control of blackflies (Fredeen 1972).

As far as can be determined, no OC pesticides were used in Arctic Norway or Sweden. Unofficial use of DDT may still be continuing in remote areas of Siberia, Russia, primarily for insect control (McConnell *et al.* 1996). Just as in Canada, DDT was probably used for insect control around DEW Line sites in Alaska.

## 6.3. Characteristics of Arctic ecosystems related to POP accumulation

A number of generalizations can be made about POP accumulation in Arctic climates compared to temperate climates. Several characteristics of Arctic ecosystems influence the extent and manner in which POPs bioaccumulate and/or cause stresses in biota that may make them more vulnerable to the effects of POPs (section 6.8.4). The details of the relationship between the tendency of organisms to accumulate POPs and the characteristics of their environments are discussed in section 6.6. The most important characteristics of the Arctic that relate to POP bioaccumulation in biota are the following.

*1. Cold*

Cold conditions influence physical characteristics of the abiotic environment, the chemical and physical characteristics of contaminants, the rates of biological processes, and a large number of physiological and behavioral adaptations of biota to cold. The most important of these adaptations is the metabolic use of lipids as an energy source and as stored energy. As a result, large amounts of lipids are transferred in Arctic food webs. This is the most important factor relating to OC accumulation and biomagnification in Arctic biota (section 6.3.1.1. below).

*2. Conspicuous species and humans at high trophic levels*

Arctic food chains, in general, are neither longer nor shorter than natural food chains in temperate regions. There are many species of first-level carnivores in both climates, but there are few third-level carnivores.

Nevertheless, many southern environments have simpler food webs than Arctic ecosystems for a variety of reasons, including the dominance of agriculture and livestock farming, past and present exploitation (e.g., logging, hunting), and contamination (e.g., fertilizers, pesticides). At southern and temperate latitudes, many of the large higher level carnivores (e.g., seals, other marine mammals, birds of prey, wolves, wild cats), particularly the ones that competed with humans for food, are relatively rare or have become extinct. While contaminant levels in these species may be a concern, the species are seldom eaten by humans. Thus, the opportunity for biomagnification of contaminants to humans is considerably reduced (section 6.4). In the Arctic, on the other hand, these third-level predators are more likely to be consumed by humans, and thus, transfer of contaminants to humans is more likely to occur.

Also, in the Arctic, some of the higher level carnivores that are consumed by humans, such as whales, are older than animals used for meat at southern latitudes. Such animals may have high contaminant levels due to many years of accumulation.

*3. Low species diversity*

The low species diversity in the Arctic is the result of recent repeated glaciations, and consequently, a short evolutionary history of ecosystems, and also often due to low absolute productivity. Polar areas that have not experienced such glaciations, such as the Antarctic marine environment, have considerably higher biological diversity and an accompanying higher degree of specialization (Dunbar 1968). Although listings of Arctic species may appear substantial, the number of species in any particular area is usually very limited. The complexity of food webs increases as Arctic ecosystems grade into temperate ecosystems.

Because of the low species diversity, some food chains may be very simple, for example, the lichen → caribou → wolf chain in Arctic Canada. This chain is of particular interest because of the importance of caribou as a food source to many northerners. Food webs are more complex in most freshwater habitats, and even more complex in marine habitats.

The low diversity in the Arctic is associated with opportunistic and invading species that are adapted to survive successfully under a range of conditions. Individuals in many Arctic species adjust their feeding habits, growth rates, migration patterns, and reproductive characteristics in response to climatic factors or the availability of food. Individuals or species in any given environment may be opportunistic feeders and, thus, may not have a well-defined position in the food web. For example, the freshwater amphipod *Gammarus* can be entirely herbivorous, but is carnivorous if possible; a few individuals in a population of freshwater fish may be cannibals; and walrus, which feed mainly on mollusks, may eat seals if desperate or if the opportunity is provided. Feeding strategies may also depend on the age and experience of an animal, and may also differ between years. Many species are adapted to withstand periods of starvation. Few population ecologists consider Arctic species to be 'sensitive', since the environmental tolerances of most species are broad.

*4. Low productivity*

Many areas of the Arctic are low in productivity primarily due to low levels of nutrient input and low levels of light, not necessarily due to the cold or short growing season (Dunbar 1968, 1986). Low productivity is most common in terrestrial and freshwater environments.

The levels of productivity in an area may be dependent on aerial deposition of nutrients and organic matter, and in aquatic systems, inputs from streams and rivers, and upwelling of nutrient-rich water from southern water masses. All inputs tend to carry organic matter, and associated contaminants.

In lakes, productivity level is correlated with food web complexity. Such relationships may partly account for different rates of bioaccumulation for a species when comparing systems.

Low productivity in the Arctic may result in slower-growing and longer-lived poikilotherms than in temperate climates (Dunbar 1968). Fish and invertebrates may be exposed to OCs for a long period of time before being consumed in the next level of the food web. Within populations, OC levels in individual fish have been shown to relate to age in both temperate and Arctic species.

*5. Cyclic annual productivity*

Arctic ecosystems are highly pulsed due to fluctuations in light levels, nutrient input, and temperature. OCs and nutrients deposited on snow, ice, soil, and plants during the Arctic winter can be mobilized and assimilated very quickly in the spring when light and warmer temperatures occur. In freshwater systems, the spring melt carries nutrients and some OCs into streams, ponds, and lakes. In the Arctic marine and freshwater environments, a burst of primary productivity occurs under the ice when light levels become high in the spring. At this time, nutrients, lipids, and associated contaminants can move into and through the food web very rapidly.

The cyclic productivity is related to many physiological and behavioral adaptations of plants and animals to their environment. The first major adaptation important to POP bioaccumulation is the ability to consume food, grow and store energy when food is available, and to metabolize stored energy when no food is available The importance of lipids in this process is further discussed in sections 6.3.1.1 and 6.4. The second adaptation is migration and/or dispersal to superior overwintering or feeding habitats, in most cases outside of the Arctic.

To interpret contaminant levels in many species, it must also be recognized that many Arctic animals disperse or migrate at some points in life, hence they are exposed to different levels of contaminants at different life stages or at different times of the year. A large variety of dispersal and migration patterns exist in Arctic animals, and the patterns for any species may differ between years. Thus, contaminants in some species and also in the predators that consume them may not directly relate to contaminant deposition in their summering ranges.

Some details about the dispersal and migration patterns of individual species are described in the discussion of species in section 6.6.

*6. Physical stressors in the Arctic*

There are numerous stressors, not directly related to chemical contamination, which do and will continue to affect the Arctic. Any of these could influence the effects of OCs on species or ecosystems since the stressors may affect their health in a variety of ways. Among these stressors are habitat destruction due to, e.g., hydroelectric development, increased human settlement and activity, resource extraction, climate change, and over-harvesting (Welch 1995). The Arctic environment is very sensitive to physical disturbance, recovering slowly due to the cold climate and low biological productivity. The terrestrial environment is particularly susceptible, with evidence of minor human activity often obvious after hundreds of years. The marine environment is also vulnerable. Ecotourism has already led to habitat destruction and harassment of animals in some cases. Seabirds are particularly susceptible to oil fouling.

### 6.3.1. The relationship between food webs and POP accumulation

Studies in both temperate and Arctic ecosystems have shown that the accumulation and metabolism of organic contaminants in fauna is dependent on the length of the food chain. For example, Rasmussen *et al.* (1990) reported that in lakes in Ontario, Canada, each trophic level contributed a 3.5-fold biomagnification factor to PCB concentrations in lake trout (*Salvelinus nanaycush*); the lipid content of trout also increased 1.5-fold for each additional trophic level. Also, food web characteristics relate to the type of OCs accumulated since some OCs are metabolized or excreted more efficiently than others.

Similar processes occur in Arctic systems. As an example, Figure 6·41 (see section 6.6.4.10) demonstrates the biomagnification of PCBs and chlordane-related compounds at different trophic levels in the marine food web. The logarithmic plot for PCBs has a slope of approximately 1, hence the increase at each level of the food web is approximately 10-fold.

Some Arctic food webs are not well defined due to the opportunistic feeding habits of Arctic species and a lack of knowledge of the food webs. One recent approach to quantifying the trophic status of individuals or species has been to measure ratios of stable carbon ($^{13}C/^{12}C$) or nitrogen ($^{15}N/^{14}N$) isotopes (Peterson and Fry 1987) (for an example, see Figure 6·20 in section 6.6.3.4.1). Carbon isotope ratios do not change much from prey to predator, enabling the original carbon source to be traced through the food web. The heavier isotope of nitrogen $^{15}N$ is usually enriched approximately 3.7 parts per thousand in an organism compared to its diet since the lighter isotope is preferentially metabolized. Stable isotopes integrate information from an organism's diet over the time period of tissue turnover.

#### 6.3.1.1. **Role of lipids in Arctic food webs**

Due to the hydrophobic nature of organic contaminants, their dynamics in Arctic food webs is closely related to the dynamics of lipids in Arctic organisms. OC levels in Arctic organisms are mostly determined by the OC levels in lipids that they consume or are in contact with, and the efficiency of lipid absorption (section 6.4). As discussed earlier, high lipid levels are adaptations to the cold and the cyclic annual productivity in many Arctic organisms. Also as discussed, long lipid-based food chains and complex food webs also contribute to high levels of OCs in the Arctic.

The transfer of lipids is particularly evident in the marine food web. The lipid content of primary producers, mainly diatoms, is high (Sargent and Henderson 1986). Pelagic marine organisms that feed on the phytoplankton have a strong propensity for converting phytoplankton into lipid stores (Falk-Petersen *et al.* 1981, Sargent and Falk-Petersen 1981). The lipids produced by phytoplankton during the spring bloom in March and April are evident as high lipid levels in the zooplankton species *Calanus finmarchicus* and *Thysanoessa* spp. (krill) (up to 70% and 50% of dry weight, respectively) in June to August, and then as high lipid levels (up to 50%) in a fish species, *Mallotus villosus* (capelin), which feeds on these zooplankton in September and October. Transfer to higher levels of the food web occurs over many years, since most species at the top of the food web, particularly marine mammals, are long-lived.

A large proportion of the body mass of marine mammals is fat. This is consolidated as a blubber layer that serves as both body insulation and energy storage. Many species, particularly carnivores, have a preference for lipid-rich tissues. For example, polar bears prefer to eat blubber and skin of seals (Stirling and McEwan 1975). This preference or necessity may exist for many Arctic species that survive at colder temperatures and/or accumulate energy reserves before an Arctic winter or for reproduction.

Lipids and associated OCs can be passed on from parent to offspring during several stages of development and during lactation in mammals. OC levels in bird eggs are usually 60% of the levels in the adults on a wet weight basis (Braune and Norstrom 1989). In birds that lay more than one egg, highest OC levels are usually found in the first egg and lower levels in subsequent eggs (Bignert *et al.* 1994). In migratory species, these lipids and OCs often originate from winter feeding areas at southern latitudes. In mammals, lipid reserves are also converted to lipid-rich milk, which may result in unusually high OC concentrations in young animals. For example, harp and hooded seal pups attain the PCB level in their mothers within two weeks after birth (Espeland *et al.* 1996).

Due to a lifetime of feeding on lipid-rich foods, older individuals usually have higher OC concentrations than younger animals. For example, in seals, older males often have the highest OC levels since females are able to excrete OCs in milk (Espeland *et al.* 1996).

Nutritional status can have dramatic effects on OC levels. When animals are starved or lose weight due to other stresses, the lipid content in their tissues necessarily decreases. However, the total amount of many contaminants in body tissues does not decrease, hence contaminant concentrations in the lipids or in vital organs increase. This has been observed in several species. Polar bears undergo a long period of fasting during which the adipose fat reserves are depleted. The lipid weight levels of PCB, chlordane, and chlorinated benzenes increase in the remaining fat and in the milk, thus increasing the amounts ingested by cubs (Polischuk *et al.* 1994, 1995). Harp seals from the Barents Sea showed seasonal changes in POPs levels related to changes in blubber thickness, with the highest levels found when the animals were leanest (Kleivane *et al.* 1995). PCB levels increase in female kittiwakes during the egg-laying period, partly as a result of mobilization of lipid reserves with consequent decreases in body mass (Henriksen *et al.* 1996). Also, in both freshwater and marine fish species, rapid decreases in lipid levels during the spawning season have been shown to be accompanied by sudden increases in PCB and DDT (Edgren *et al.* 1981, Bignert *et al.* 1993, Jacobsen *et al.* 1993).

The higher lipid weight levels of OCs caused by reduced amounts of body fats may have toxicological implications. Geyer *et al.* (1993a, 1993b) have found significant correlations between $LC_{50}/LD_{50}$ values and total body fat contents for dieldrin in mosquito larvae, lindane (γ-HCH) in fish, and 2,3,7,8-TCDD in mammals. The higher the percent of total body fat in different species, the less toxic the OCs were. They concluded that storage of OCs in fat serves as a detoxification mechanism, removing them from sites of action. In light of this, increased lipid weight OC levels due to seasonal reductions in the amount of total body fats would imply increased risk of toxic effects in the organism.

When body fat is reduced, OCs may be redistributed from the adipose tissue to vital organs such as the central nervous system (Henriksen *et al.* 1996). Diseased and starved female Baltic grey seals have been shown to have higher lipid weight concentrations of polybrominated diphenyl ethers, polychlorinated camphenes (toxaphene), chlordanes, DDT, PCBs and methylsulfonated chlorobiphenyls than healthy females (Olsson *et al.* 1992a). Emaciated animals that have been found dead (e.g., Icelandic gyrfalcon, section 6.6.2.3.4) often contain high levels of contaminants because of low lipid levels, and in many cases it is believed that the contaminants may have contributed to the death of the animals.

There are no latitudinal patterns in the lipid content of dominant fish (cod, herring, and mackerel) in northern waters or in the lipid content of marine mammals (Wania 1995). However, latitudinal differences in the types of lipids in organisms could influence the bioaccumulation of OCs. Numerous latitudinal differences in lipid metabolism have

been reported. For example, it has been known for more than 20 years that poikilotherms increase the ratio of polyunsaturated to saturated fatty acids in membrane lipids in response to lowered ambient temperatures (Greene and Selivonchick 1987). Also, the lipid content of organs involved in osmoregulation has been reported to be lower at marine salinities (Henderson and Tocher 1987). However, such trends have not been related to levels of OCs.

#### 6.3.1.2. **Terrestrial food webs**

Arctic terrestrial food webs can be short, often consisting of plants or lichens at the primary producer level, a few herbivores, and one or two main predators. The air → plant → animal contaminant pathway is the major route of contamination into the food chain. Large Arctic herbivores include caribou and reindeer (*Rangifer tarandus*), muskoxen (*Ovibos moschatus*), ptarmigan (*Lagopus* spp.), and Arctic hare (*Lepus arcticus*). Also, waterfowl such as geese and swans are entirely herbivorous in both their southern and northern ranges. Numerous small mammals occur in the Arctic, including ground squirrels (*Spermophilus parryii*), voles (*Clethrionomys rutilus* and *Microtus* spp.), lemmings, mustelids, and shrews (Sage 1986). These animals are consumed by weasels, red fox (*Vulpes vulpes*), Arctic fox (*Alopex lagopus*), wolves (*Canis lupus*), wolverines (*Gulo gulo*), barren ground grizzly bears (*Ursus arctos*), and humans. The diversity and complexity of the food webs increases in subarctic habitats.

The lichen-to-caribou food chain is of interest because caribou are consumed by humans. Caribou's main winter food source, lichens, accumulate contaminants more readily than other plants because of their large surface area, longevity, and ability to bind heavy metals. They accumulate atmospheric contaminants in a non-selective manner, resulting in a contaminant load similar to atmospheric input (Thomas *et al*. 1992).

Many terrestrial shorebirds (Charadriidae and Scolopacidae) can be considered first- or second-level carnivores in terrestrial or freshwater food webs. They in turn are preyed upon by the mammalian predators listed above and by birds of prey, including owls, eagles, hawks, accipiters, and falcons.

#### 6.3.1.3. **Freshwater food webs**

Important freshwater systems include lakes, tundra ponds, and rivers. Arctic lake food webs usually have five trophic levels, with plants at the base, herbivores that include zooplankton, detrital feeders, such as benthic insect larvae and crustacea, carnivores feeding on benthic organisms, and piscivores. In North American Arctic lakes, Arctic char (*Salvelinus alpinus*), lake whitefish (*Coregonus clupeaformis*), and lake trout (*Salvelinus namaycush*) are the most common first-order carnivores. Several other species of whitefish (*Coregonus* sp.), several species of salmon (*Salmo* spp.), and grayling (*Thymallus arcticus*) replace char at lower latitudes or in more productive habitats. Many of the species that occur in the western Arctic of North America also occur in eastern Russia. In Europe, the most common carnivorous species are brown trout (*Salmo trutta*), European whitefish (*Coregonus lavaretus*), Arctic char, and Atlantic salmon (*Salmo salar*). Second-order carnivorous fish occur in some systems. Higher-level carnivores include inconnu (*Coregonus leucichthys*) in the western North American Arctic, and pike (*Esox lucius*) and burbot (*Lota lota*) on both continents. Terrestrial carnivores such as birds of prey and mustelids also feed on species in freshwater systems.

The trophic level of some species may be higher at later life stages. Many fish feed on phytoplankton and zooplankton at early life stages, then progress to feeding on invertebrates and mollusks, and later become piscivores. In the North American Arctic, individual Arctic char or lake trout, usually very large individuals, will eventually become cannibals, feeding on smaller individuals of their own species (Welch 1991).

Arctic ponds that freeze to the bottom in winter do not contain fish. However, the biological productivity in summer months can be high and the productivity does feed into several food chains. Numerous herbivorous and some carnivorous invertebrates live in such ponds. The third trophic level usually consists of migratory birds.

Riverine food webs are usually similar to lake food webs in habitats of the same dimensions, but detritivorous microorganisms and invertebrate species are more common in flowing water. In general, the same carnivorous fish species inhabit rivers as lakes.

Rivers are the migration routes for many species of anadromous fish such as Arctic char and coregonids, which are extensively fished during the fall migrations by local people. Also, seals and whales will move into inland lakes, rivers, and estuaries during the summer months.

#### 6.3.1.4. **Marine food webs**

The inter-relationships between water, land, and ice result in diverse habitats for marine Arctic flora and fauna. These habitats include open seas, channels, estuaries, shallow bays, deep fjords, lakes, rivers, fast ice, pack ice, ice edges, polynyas, and shore leads (chapter 4).

A generalized marine Arctic food web, or more accurately, a diagram of the flow of energy, nutrients, and organic matter, and thus OCs, which includes major species and taxa of interest, is shown in Figure 6·2 (next page). Different parts of this chain are more dominant in different marine areas and at different times of the year. The most commonly presented Arctic food chain, leading from primary producers to top level carnivores such as seabirds, polar bears, and humans, is most often discussed in relation to OCs.

In ice-free offshore waters, the phytoplankton-supported part of the food web dominates. The benthic component of the food web is most important in nearshore areas beyond the intertidal and ice scour zone (Clarke 1993). Estuaries such as Hudson Bay and the Mackenzie River Delta are moderately productive, and support coregonids and beluga. The marine subarctic consists of seas and basins in which Arctic waters mix with other Atlantic and Pacific waters or freshwaters. These areas are vertically unstable, and where nutrient upwelling from deep water occurs, these areas can be very productive. Some important fisheries, including Atlantic cod, salmon, redfish, and capelin, occur in these areas where water masses meet or upwelling occurs, such as in Lancaster Sound in the Canadian Arctic. Also, marine mammals and seabirds concentrate at polynyas, leads, and ice edges in these areas (Stirling and Cleator 1981, Bradstreet and Cross 1982).

Detrital food webs, which start with decomposing organic matter and then produce nutrients for primary production, are not easily demonstrated in food web diagrams, but are nevertheless important in the transfer of contaminants. Two examples are the importance of epontic ice algae in the marine food web and benthic food webs based on detritus.

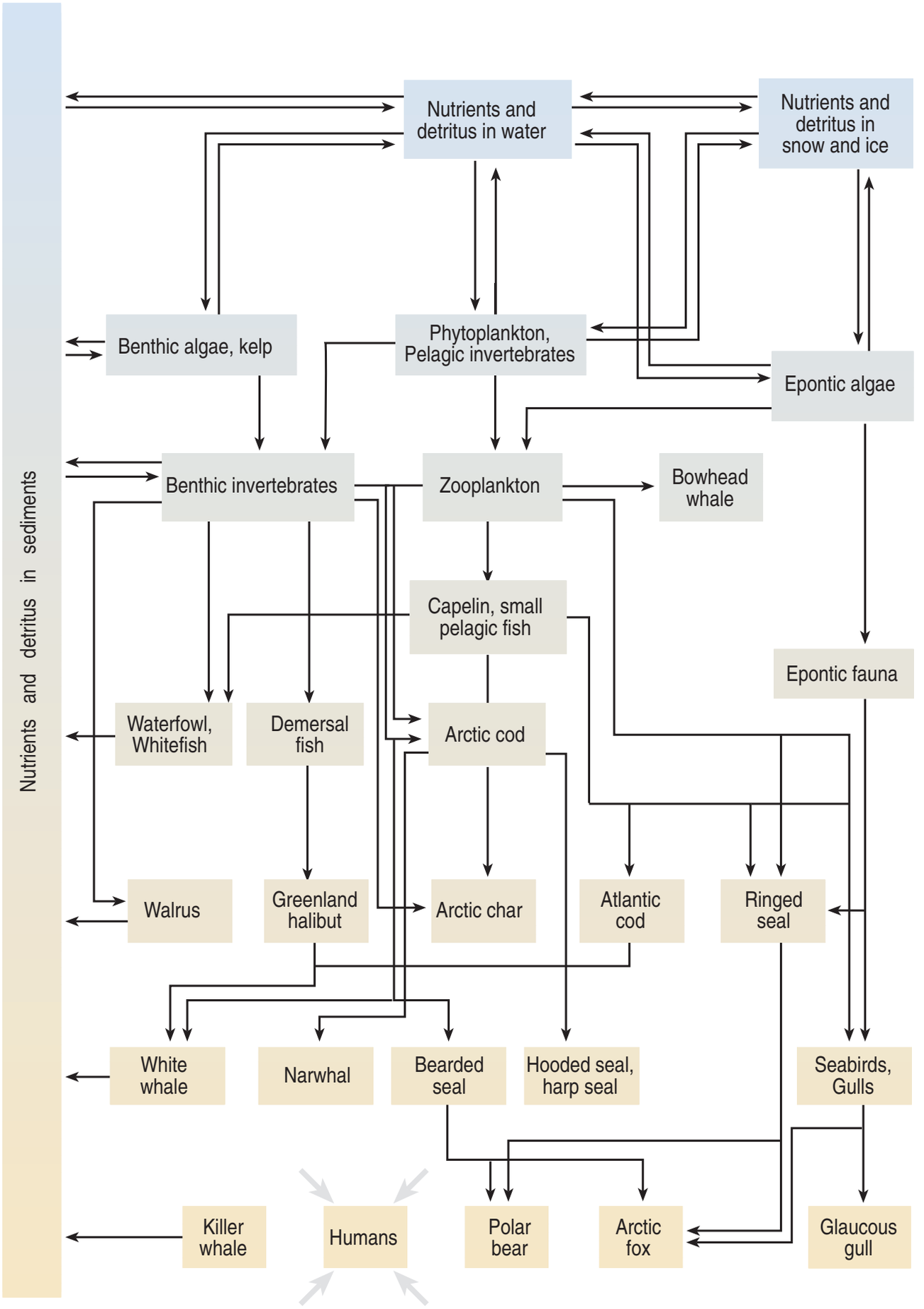



Figure 6·2. Generalized marine food web.

## 6.4. Bioaccumulation and biomagnification in terrestrial, freshwater, and marine environments – definitions and general principles

The majority of the POPs, particularly the organochlorines (OCs) discussed in this chapter, are semi-volatile, have low water solubilities and are highly lipophilic. These characteristics, combined with their chemical stability, lead to the establishment of a steady state between concentrations in water and air and in organic phases such as organic carbon and lipids (Mackay and Paterson 1981, 1982). Many POPs are therefore found strongly associated with organic matter, such as particles with high organic contents (humus, soot), after emission. A small fraction of the total is found dissolved in water or in the gas phase in air. Living organisms are exposed to POPs in these abiotic matrices, and the POPs enter the organisms by varying routes and accumulate in the body lipids (see for example: Thomann and Connolly 1984, Connolly and Pedersen 1988, Oliver and Niimi 1988, Thomann 1989, Bierman 1990, Opperhuizen and Sijm 1990, Thomann *et al*. 1992, Fox *et al*. 1994). Bioaccumulation is the term used to define the net accumulation of POPs from all exposure routes (Thomann 1989). Bioaccumulation is usually expressed as the concentration of a POP in an organism on a lipid weight basis divided by the concentration found in water (truly dissolved) or air (gas phase).

Biomagnification is the term used to define the increased accumulation of POPs with each trophic level in a food web and is expressed as the concentrations in the organism divided by the concentrations in its food, both on a lipid weight or organic carbon (sediments, soils) basis. In aquatic ecosystems, uptake through ingestion of contaminated food is the main route for bioaccumulation of POPs with log $K_{ow} > 5$, with only a small fraction being accumulated from gill respiration (Thomann 1989, Thomann *et al*. 1992).

The patterns of POPs in abiotic samples will resemble those from emission sources, although they will have been altered to some extent during long-range transport. Bioavailability refers to the extent to which pollutants associated with soils, plants, sediments, or suspended or dissolved organic carbon in the water column are available for uptake by biota (Dickson *et al*. 1994). Many factors such as organic carbon content of soils and sediments, pH, and kinetic limitations influence the amount of a contaminant that can be released from food particles in the gut or dissolved into sediment pore-waters, and therefore reduce the environmental bioavailability. Despite being tightly bound to particles, membranes, and fat globules, most organochlorine contaminants of concern in the Arctic have been shown in laboratory studies with invertebrates, fish, mammals, and birds, to be readily assimilated from the diet, and when present in the dissolved phase in water, to be rapidly accumulated from water.

The POP patterns may change for each trophic level in a food web as more easily metabolized POPs are eliminated and the more persistent POPs are biomagnified. Thus, predatory birds and mammals at high trophic levels will mainly be exposed to the most persistent POPs (e.g., in section 6.1 and Figure 6·1). This means that, for ecotoxicological risk assessment purposes, it is also important to analyze the levels of POPs at lower trophic levels, in order to estimate the intake and therefore exposure of organisms at higher trophic levels.

### 6.4.1. Terrestrial ecosystem bioaccumulation/ biomagnification

The presence of OCs in the terrestrial ecosystem is primarily the result of long-range transport of these contaminants in air. OCs bound to particles are deposited onto plants, snow, and soil, and OCs in the gas phase diffuse into the waxy layers of plants. Soil microorganisms and invertebrates accumulate OCs that are bound to organic matter via direct contact with the soil or from soil ingestion. Accumulation in plants is from dry and wet deposition of particles onto plant surfaces and diffusion from air into waxy layers. Plant uptake of OCs via the roots is very limited.

For animals, uptake from breathing air is considered to be negligible. Bioaccumulation is primarily from food ingestion. Herbivorous birds and mammals are exposed to OCs via ingestion of plants and soil while omnivores may ingest terrestrial invertebrates as well as smaller birds and mammals. Animals that eat perennial vegetation, for example lichens and twigs, will be exposed to more OCs than those eating annual herbs because of the accumulation of particles containing OCs on the plant surfaces. At the top of the food web are the predatory birds and mammals, which feed on herbivores and omnivores.

### 6.4.2. Freshwater ecosystem bioaccumulation/ biomagnification

There are a few cases where local sources of OCs have contaminated freshwater systems in the Arctic, but the presence of OCs in the freshwater ecosystem is primarily the result of long-range transport in air. OCs bound to particles are eventually deposited onto snow, soil, ice, and water surfaces. Particles on snow and soil particles are eventually washed into waterways by snowmelt, rain, and flooding. When the ice melts, the deposited particles enter the water. Particles deposited on water surfaces are mixed into the water mass. In these cases, the particles become suspended in the water column.

The suspended particles eventually sediment out in lake bottoms. OCs bound to the suspended particles diffuse between the particle's organic carbon fraction and the water, especially the dissolved carbon fraction. In sediments, OCs diffuse between the organic carbon of the sediment, the pore water between sediment particles, and the overlying water layer.

Aquatic microorganisms, plankton, algae, and plants are exposed to OCs via contact with the dissolved fraction in water and possible direct contact with suspended particles. Uptake occurs by diffusion into the lipid-rich outer membranes and waxy layers and from food ingestion for microorganisms and zooplankton.

Invertebrates bioaccumulate OCs dissolved in the water phase when these come in contact with the gills as well as from ingestion of microorganisms, plankton, suspended particles, and sediment. For fish, bioaccumulation will mainly be from food ingestion, whether this is sediment, plankton, or other fish, but some accumulation will come from gill respiration, depending on how lipophilic the OCs are. The more lipophilic the OC, the less important gill uptake becomes. Accumulation in mammals and birds will be entirely from food ingestion.

### 6.4.3. Marine ecosystem bioaccumulation/ biomagnification

The presence of OCs in the marine ecosystem is the result of long-range transport in air, diffuse pollution from land-based and water-based sources in the Arctic, the riverine input of

contaminated sediments via water flow or ice, as well as from the mixing of oceanic waters. The input of OCs will mainly be in the form of particles that are deposited, washed, or transported in ice into the marine ecosystem. In the case of the sea ice edge, the particles will be released to the water as the ice melts. The presence of TBT in the marine ecosystem is the result of diffusion from ship bottoms, nets, and other devices in seawater that have been treated. TBT will also mainly be bound to particles.

In open waters, the suspended particles eventually sediment out to the sea bottom. POPs bound to the suspended particles diffuse between the particle's organic carbon fraction and the water, especially the dissolved carbon fraction. In sediments, POPs diffuse between the organic carbon of the sediment, the pore water between sediment particles, and the overlying water layer.

Aquatic microorganisms, plankton, algae, and plants are exposed to POPs via contact with the dissolved fraction in water and possible direct contact with suspended particles. Uptake occurs to the lipid-rich outer membranes and waxy layers. Microorganisms and zooplankton are also exposed via food uptake. Suspended particulate matter concentrations in ice-covered regions of the Arctic Ocean are very low (Gordon and Cranford 1985), but increases occur during late summer due to production by under-ice epontic algae and phytoplankton in the upper water column (Hargrave *et al*. 1989b). Rates of inorganic and organic particulate matter sedimentation also increase by an order of magnitude during August (Hargrave *et al*. 1989b, Hargrave 1994) and scavenging of particle-reactive POPs from the water column should be greatest at this time of year. Particle-adsorbed POPs are then available for uptake by grazing organisms or sedimentation and subsequent incorporation in the benthic food web.

Invertebrates bioaccumulate OCs and TBT dissolved in the water phase when these come in contact with the gills, as well as from ingestion of microorganisms, plankton, suspended particles, and sediment. For fish, bioaccumulation will mainly be from food ingestion, whether this is sediment, plankton, or other fish, but some accumulation will be from gill respiration, depending on how lipophilic the OCs are. The more lipophilic the OC, the less important gill uptake is. OC accumulation in birds and mammals occurs entirely from food ingestion.

Particles as well as nutrients are also released at the ice edge as the ice melts and retreats during the summer. The OCs bound to the particles diffuse between the particle's organic carbon fraction and the water layer they are trapped in, especially the dissolved carbon fraction. The stable layer of nutrient-rich, low salinity water that forms leads to a bloom of epontic (ice-edge) algae. These will be exposed to POPs via contact with the dissolved fraction of OCs in the water layer, and direct contact with suspended particles. Zooplankton will be exposed to OCs in a similar manner although food intake will also play a role. Ice-associated amphipods will bioaccumulate OCs dissolved in the water phase via the gills and from ingesting epontic plankton and particles. These amphipods in turn are important food for fish, seabirds, and seals.

## 6.5. Toxicology

### 6.5.1. Toxicokinetics

The majority of the substances dealt with in this chapter are lipophilic, stable, and persistent. They are taken up by aquatic living organisms via diffusion over the gills and from food in the gastrointestinal tract. POPs, particularly OCs, cross the gill/gut membrane and enter the blood where they are quickly distributed to high lipid tissues such as the liver and adipose tissue. Metabolism and elimination are often slow, leading to a net increase of these substances in the organism over time.

There are species differences in the tissue distribution of OCs, partly due to differences in lipid distribution. For example, high concentrations of orally administered 2,3,3',4,4'-PeCB (CB105) were found in the liver and brain of cod, while rainbow trout accumulation was in the extrahepatic fat depots (Bernhoft *et al*. 1994). Lipid dynamics can also affect the distribution of OCs. Female kittiwakes (*Rissa tridactyla*) showed a redistribution of PCBs from the liver and body fat to the brain during the period of pre-breeding to late chick rearing. This was in part due to the mobilization of lipids from the liver and body fat during reproduction and subsequent loss in body mass, which in turn led to higher lipid weight PCB concentrations in the remaining lipids (Henriksen *et al*. 1996). These examples imply that different tissues in different species will be the targets for possible effects from OCs, and this in turn is affected by lipid distribution and dynamics.

Metabolism of xenobiotics occurs mainly in the liver via a two-phase process. In phase I, xenobiotics are converted by oxidation reactions to metabolites that can undergo phase II reactions. In phase II, the product is conjugated with glucuronic acid or glutathione, for example, to produce water-soluble compounds that can be excreted in urine or bile. These processes are catalyzed by liver enzymes such as the cytochrome P450 containing monooxygenases (Nebert and Gonzalez 1987). Substances that are resistant to metabolism will be selectively accumulated in living organisms. In addition to detoxification, the enzymatic processes can also create reactive intermediates that may be mutagenic and/or carcinogenic, or metabolites that are lipophilic and have retained toxicity, or that have the ability to bind selectively to proteins and accumulate in the organism.

Many OCs form metabolites that are biologically active. DDT is metabolized in living organisms to DDD and further to DDE, both of which are lipophilic and toxic, and accumulate in biota (WHO 1989a). In some cases, a methylsulfone ($MeSO_2$) group is added during metabolism and a number of $MeSO_2$-DDE and $MeSO_2$-PCB congeners have been identified in animals (Jensen and Jansson 1976, Lund *et al*. 1988, Haraguchi *et al*. 1990, 1992, Bergman *et al*. 1992b, 1994b, Brandt *et al*. 1992, Letcher *et al*. 1994, 1995). Some congeners of PCB may also form hydroxylated metabolites (Jansson *et al*. 1975). This type of metabolite has been found to selectively bind to transthyretin, one of the major transport proteins for retinol and thyroid hormones in the blood (Brouwer *et al*. 1988, 1990, Bergman *et al*. 1994a). Aldrin is metabolized in living organisms to dieldrin by the cytochrome P450-dependent monooxygenase, aldrin epoxidase (WHO 1989c). γ-chlordane is metabolized to some extent to oxychlordane (WHO 1984a). Hexachlorobenzene is metabolized to some extent, mainly by the liver, and may form, among other metabolites, pentachlorophenol, tetrachlorohydroquinone, pentachlorothiophenol and lower chlorinated benzenes (Debets and Strik 1979, Renner 1988).

The major excretion route of OCs and their metabolites is via the feces. Some of this is passive diffusion over the gut membrane and some from bile excretion of metabolites. In invertebrates and fish, excretion also occurs by diffusion over the gill membranes. Female fish and birds excrete lipophilic OCs via their eggs, and female mammals via placental transfer to the fetus and in breast milk. A particular charac-

teristic of the Arctic is that most marine mammals have very high fat contents in breast milk in order to facilitate fast growth in the young during the short growing season. For example, polar bear milk has a fat content of 20-46% (Derocher *et al*. 1993, Oehme *et al*. 1995a, Polischuk *et al*. 1995, Bernhoft *et al*. 1996) and different seal species have milk fat contents of 30-60% (Bacon *et al*. 1992, Pomeroy and Green 1993, Addison and Brodie 1977, 1987) as compared to fat contents of 1-4% in human and cow's milk (Becher *et al*. 1995, Norén 1988, Rappe *et al*. 1987). Therefore, excretion of OCs via milk is more important than via placental transfer for adult females in marine mammal species. This in turn enhances OC exposure of young, particularly for polar bears, Arctic foxes, whales, and seals. Young harp and hooded seals, for example, have as high levels of some OCs as their mothers at the end of the lactation period (Espeland *et al*. 1996). Young polar bears (1-2 years) have similar PCB levels to adult females with high PCB levels (Bernhoft *et al*. 1996), and polar bear cubs-of-the-year have higher concentrations of many OCs than their mothers (Polischuk *et al*. 1995). This is of concern as young animals may be more sensitive to the effects of OCs than adults.

The net result of uptake, distribution, metabolism, and excretion will determine the OC levels found in an organism. This is in turn affected by other factors. Studies carried out to determine the uptake, distribution, metabolism, and excretion of OCs are usually done with one substance at a time. Wildlife and humans, however, are exposed to complex mixtures of OCs. Very little is known about how different OCs affect each other's toxicokinetics. OCs that induce the hepatic cytochrome P450 system will affect the metabolism of other xenobiotics, for example. This may lead to an increase in xenobiotic metabolism, thus increasing excretion. For example, studies on Baltic seals show that high body burdens of DDT and PCB are associated with lower relative amounts of the mono-*ortho* CB, 2,3',4,4',5-PeCB (CB 118). At PCB concentrations (sum of CBs 28, 52, 101, 118, 138, 153, 181) of 50 μg/g lw or higher, CB 118 could not be found (Haraguchi *et al*. 1992, Olsson *et al*. 1992b). PCB levels in Arctic ringed seal are much lower, xenobiotic metabolism does not seem to be induced, and concomitantly, CB 118 is present in higher relative amounts (Norstrom and Muir 1994).

Lipid weight concentrations of PCBs, chlordanes, and chlorobenzenes have been found to increase in female polar bears during their fasting period, but DDT and HCH do not. This implies that DDT and HCH are metabolized during fasting (Polischuk *et al*. 1995) which could be a result of liver enzyme induction. It has also been shown that mink exposed to both DDT and PCB only biomagnify PCB whereas mink exposed either to DDT compounds or PCBs biomagnify the two groups of compounds at a similar rate (Kihlström *et al*. 1976).

An increase in xenobiotic metabolism may also lead to an increase in the formation of reactive intermediates, with increased toxicity and tissue damage (Boon *et al*. 1992). There are indications, for instance, that PCB exposure may influence the magnitude of carcinogenicity of PAHs in fish (Bailey *et al*. 1989) and that exposure to PCBs also increases the uptake of PAHs in English sole (Stein *et al*. 1984).

Xenobiotic metabolism is also subject to biological variation. In a study of salmon (*Salmo salar*), the cytochrome P450 enzyme system was followed for a year and showed cyclical variations in enzyme activity. The basal enzyme activity measured as ethoxyresorufin-O-deethylase (EROD) levels was higher in both males and females during the winter months and then dropped during the summer, most particularly during the period of sexual maturation (Larsen *et al*. 1992). Significant differences were seen in EROD levels between males and females at sexual maturation, with females having lower or non-detectable activity just prior to ovulation.

Thus, it is very difficult to evaluate the toxicokinetics of environmental exposures to mixtures of OCs. The interactions that have been seen indicate that the relative amounts and the composition of various contaminants in animals may partly be the result of selective effects on the organism's uptake, metabolism, and excretion of OCs, and not solely a result of the specific pollution burdens of contaminants in the area.

### 6.5.2. Types of effects

In most laboratory experiments studying the toxicological effects of POPs, animals are exposed to single substances or to technical products, often at acutely toxic doses. In a few studies, combinations of a few substances have been used. It should be remembered that wildlife are exposed to complex mixtures of POPs, most often at low doses. There are often considerable species differences in sensitivity to specific POPs as well as differences in response. It is, therefore, often difficult to generalize results found in one species to other species. The following is a short, general summary of results from laboratory studies as well as results from field studies where POPs have been quantified and concentrations have been correlated with biological and toxicological effects.

A wide range of effects are seen after exposure to POPs. Some of these types of effects are currently being used as biological markers for POP exposure. These include, among other things, effects on reproduction, development, cytochrome P450-dependent enzymes, porphyrins, the immune system, the adrenals, the thyroid gland, thyroid hormone levels, and vitamin A levels. Almost all POPs considered in this report also cause visible changes in the liver, including hypertrophy, lesions, and in some cases, tumors.

POPs can cause short-term acute effects when administered in high doses as well as long-term chronic effects at lower doses. In the Arctic, the major concern is long-term chronic exposures as organisms are exposed to low levels over their entire lifetimes. In this context, the major effects of concern are those that may affect reproduction and survival at the individual and population level. However, effects at the individual, population, or ecosystem level come at a late stage of exposure (Figure 6·56, section 6.8). It would be more useful to have earlier warning of exposure to POPs and, therefore, biological marker systems based on subtle, low dose effects are being developed. Most biological markers measure effects at the molecular, cellular, or organismal level, however, it is still not established what these changes may mean at the individual or population level.

#### 6.5.2.1. Reproduction and development

OCs have a number of effects on the ability of organisms to reproduce and develop normally (for reviews see Peterson *et al*. 1993, Bosveld and van den Berg 1994, Barron *et al*. 1995, Brouwer *et al*. 1995). Exposure to some OCs may cause embryo- and fetotoxicity, decreased offspring survival, abnormalities in the estrus cycle and sex hormone levels, reduced sperm production, reduced litter sizes, and even total reproductive failure in mammals. In birds, some OCs cause decreased egg production, retarded egg production, increased embryo mortality, eggshell thinning, embryonic deformities, growth retardation, and reduced egg hatchability, as well as detrimental effects on parental behavior. In fish, some OCs

cause decreased egg and larval survival, reduced sexual maturation, and reduced gonad size. Other effects of OCs on organisms may include structural malformations, neurotoxic effects, and neurological and behavioral changes in offspring. Behavioral changes also occur in adult animals, including changes in mating behavior.

Some POPs act as hormones or interfere with endocrine systems and are therefore called endocrine or hormone disrupters. The reproductive effects of embryonic or fetal exposure to these may only become obvious at later developmental stages or at sexual maturity. The estrogenic and antiestrogenic effects of POPs are the best studied. Endocrine disruption is also implicated in thyroid and immune system effects, which are discussed below. Environmental estrogens can mimic estrogen by binding to the estrogen receptor and turning it on. Antiestrogens bind to the receptor and block the normal binding of estradiol, thus inhibiting estrogen activity. Normally, estradiol stimulates proliferation and growth in the reproductive tract and mammary glands of mammals. It is necessary for reproduction and maintaining a pregnancy. In female fish, estradiol normally stimulates the liver to produce vitellogenin, a precursor to yolk. Vitellogenin is not present in males unless they have been treated with estrogens (Ng and Idler 1983). POPs may also function as androgens or antiandrogens. *p,p'*-DDE is antiandrogenic, binding to the androgen receptor and blocking the binding of endogenous androgens (Kelce *et al*. 1995). Estrogens and androgens are important in the normal sexual differentiation of developing organisms.

Biomarkers have been developed for testing the estrogenicity of POPs. Estrogenic effects can be assayed by measuring proliferation of the reproductive tract. This can be done by measuring uterine weight in immature or ovariectomized rodents (Bulger and Kupfer 1983, Galey *et al*. 1993) or increases in oviduct weight in birds (Eroschenko and Palmiter 1980). One *in vitro* bioassay (E-screen) uses a human breast tumor cell line (MCF7) that requires estrogen to proliferate (Soto *et al*. 1992, Sonnenschein *et al*. 1995). Cultured rainbow trout (*Oncorhynchus mykiss*) hepatocytes have been used to measure vitellogenin production after estrogen and phytoestrogen exposure (Pelissero *et al*. 1993). The estrogen receptor of the spotted seatrout (*Cynoscion nebulosus*) has been used to measure the affinity of pesticides for the estrogen receptor (Thomas and Smith 1993). The presence of vitellogenin in males and non-reproductive females in the wild is being used as a biomarker for environmental exposure to estrogenic substances.

### 6.5.2.2. Cytochrome P450 system

The most well developed of the biological markers is the study of cytochrome P450-dependent liver enzymes (see, for example, Förlin *et al*. 1994). Exposure to OCs and some PAHs induces liver cytochrome P450-dependent enzymes known as mixed function oxidases (MFO), which metabolize xenobiotics and endogenous substances (Nebert and Gonzalez 1987). Exposure to high concentrations of MFO-inducing OCs can affect the metabolism of endogenous substrates, such as steroid hormones, leading to disturbances in critical biological functions (Kupfer and Bulger 1976).

There are several gene families of cytochrome P450 in vertebrates (Nelson *et al*. 1993) and those that are most relevant for the metabolism of OCs are the cytochrome P450 1A and 2B gene families. Cytochrome P450 1A forms are induced by planar aromatic hydrocarbons (such as 3-methylcholanthrene), PCDD/Fs, and nPCBs. This induction is often measured as increases in several enzymes including ethoxyresorufin-O-deethylase (EROD) and aryl hydrocarbon hydroxylase (AHH). Induction of cytochrome P450 1A is mediated by the aryl hydrocarbon (Ah) receptor that has been found in the cell membranes of all vertebrates studied so far.

Cytochrome P450 2B forms are induced by another class of substances, typified by phenobarbital (PB) and measured as aminopyrine N-demethylase (APND) activity, aldrin epoxidase (AE), and pentoxyresorufin-O-dealkylase (PROD), for example. Substances that induce cytochrome P450 2B are DDT, chlordane, and di- to tetra-*ortho* PCBs and 3-$MeSO_2$-PCB. Mono-*ortho* PCB congeners and technical PCBs are mixed type inducers inducing both cytochromes P450 1A and 2B.

There are a number of critical species differences in the cytochrome P450 system, particularly with reference to the Arctic situation. The cytochrome P450 system of terrestrial mammals, particularly laboratory rodents, has been studied rather extensively (see for example Nebert *et al*. 1991). Mammals generally have two forms of cytochrome P450 1A known as 1A1 and 1A2 as well as functioning cytochrome P450 2B forms, and they thus have higher capacity for metabolizing both groups of OCs.

The cytochrome P450 system has been studied very little in wildlife, but some information is available for fish, birds, and a few mammals. Fish seem to lack cytochrome P450 2B and have only one version of cytochrome P450 1A (Nebert *et al*. 1989, Stegeman 1989, Stegeman and Hahn 1994, Goksøyr 1995b), and thus have a low OC metabolizing capacity.

Cytochromes P450 1A1 and 1A2 are found in birds (Livingstone and Stegeman 1989) and although there is only weak immunological cross-reactivity with mammalian cytochrome P450 2B, studies imply that birds do have functional cytochrome P450 2B (Ronis and Walker 1989). PCB metabolism in birds is both similar in activity and pattern to that in terrestrial mammals, and there is no evidence that this capability is substantially different in piscivorous or terrestrial bird species.

The cytochrome P450 system has been characterized in harbour (*Phoca vitulina*), grey (*Halichoerus grypus*), harp (*Phoca groenlandica*), and hooded (*Cystophora cristata*) seal (Engelhardt 1982, Addison and Brodie 1984, Addison *et al*. 1986, Goksøyr *et al*. 1992, Goksøyr 1995a) and in harbour porpoise (*Phocoena phocoena*), bottlenose dolphin (*Tursiops truncatus*), white-sided dolphin (*Lagenorhyncus acutus*), striped dolphin (*Stenella coeruleoalba*), beluga or white whale (*Delphinapterus leucas*), short finned pilot whale, (*Globicephala macrorhynchus*), killer whale (*Orcinus orca*), and minke whale (*Balaenoptera acutorostrata*) (Geraci and St. Aubin 1982, Goksøyr *et al*. 1985, 1986, 1988, 1989, Watanabe *et al*. 1989, White *et al*. 1994). Seals and cetaceans, like other mammals, seem to have functional cytochromes P450 1A1 and 1A2 and thus a higher capacity to metabolize planar compounds. They differ from terrestrial mammals in having weak cytochrome P450 2B activity and, thus, a reduced ability to metabolize xenobiotics that are substrates for these particular MFOs (Boon *et al*. 1992, Goksøyr *et al*. 1992, Goksøyr 1995a). Seals seem to have more cytochrome P450 2B than whales (Goksøyr 1995a).

The cytochrome P450 enzyme system has also been characterized in the polar bear. Polar bear liver possesses several enzymes which immunochemically cross-react with those in rat, including cytochrome P450 1A1, cytochrome P450 2B1, cytochrome P450 3A1, and epoxide hydrolase (Bandiera *et al*. 1995). Polar bear have a high metabolic capacity, particularly for PCB and DDT (Norstrom and Muir 1994).

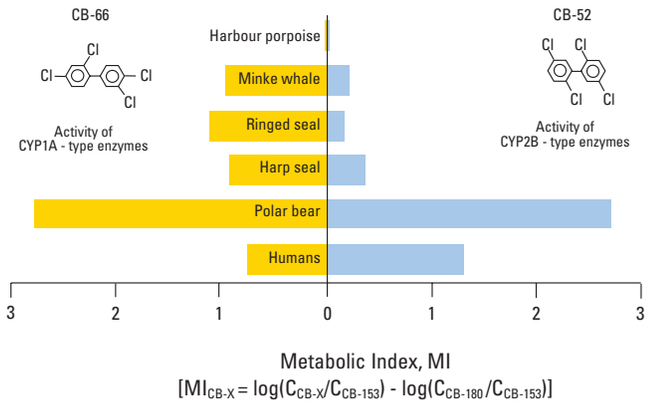



Figure 6·3. Cytochrome P450 1A (CYP1A) and cytochrome P450 2B (CYP2B) type enzyme activities in some Arctic marine mammals and humans using the metabolic index (MI) suggested by Tanabe *et al.* 1994a. The indices are based on PCB data from the Norwegian College of Veterinary Medicine/National Veterinary Institute (Skaare 1996).

Figure 6·3 compares the metabolic index as a measure of cytochrome P450 1A and 2B enzyme activities for different PCB structures in polar bear, harp seal, ringed seal, minke whale, and harbour porpoise. The metabolic index for humans is included for comparison. It can be seen that polar bear have a very high capacity for metabolizing both PCB structures, while harp seal, ringed seal, and minke whale have relatively low capacity for metabolizing non-planar PCB structures. Harbour porpoise have low metabolizing capacity for both types of structures.

Organisms lacking functional cytochromes P450 1A or 2B will not be able to eliminate the OCs metabolized by these, leading to their bioaccumulation. This is particularly the case for fish, making them carriers of many OCs in food webs. Those organisms that cannot eliminate OCs may accumulate concentrations high enough to cause effects. The presence of functional cytochrome P450 enzymes means that OCs may be metabolized and eliminated, metabolized to lipophilic and toxic metabolites, and/or that OC exposure may lead to cytochrome P450 enzyme induction, increasing the amounts of metabolic enzymes present. The species differences described above will lead to species-specific responses.

### 6.5.2.3. **Porphyria**

Longer-term exposure to high concentrations of many OCs leads to porphyria. Porphyrias are disorders in heme biosynthesis that lead to tissue dysfunction. Heme is synthesized in an eight-step process with a specific enzyme responsible for the conversion of the various heme-precursors known as porphyrins at each step. OC-related porphyria is associated with the inhibition of uroporphyrinogen decarboxylase, an enzyme that converts uroporphyrinogen III to coproporphyrinogen III in the fifth step of heme synthesis. This porphyria is also associated with the induction of the enzyme delta-amino-levulinic acid synthetase, which leads to an increased rate of porphyrin production at the first step of the process (ATSDR 1994). Thus, production of porphyrins will be increased leading to increases in the production of heme precursors in the following steps. Inhibition of uroporphyrinogen decarboxylase at step five will lead to an accumulation of these precursors in the liver, body tissues, and feces. The analysis of porphyrin levels and composition are thus useful biomarkers for exposure to some OCs. For example, there is an apparent elevated level of total heme in highly contaminated beluga from the Gulf of St. Lawrence (Payne 1994). Accumulation of highly carboxylated porphyrins is seen in herring gulls from the Great Lakes with high levels of DDE and PCDD/F (Fox *et al.* 1988).

### 6.5.2.4. **Immune system**

Many OCs are known to disrupt humoral- and cell-mediated immunity as well as non-specific responses, leading to immunosuppression. Humoral-mediated immunity involves the body's ability to recognize foreign substances (helper T-cells) and mount a response by stimulating the production of antibodies (B-cells). Cell-mediated immunity is involved in delayed hypersensitivity reactions (e.g., skin reactions to allergens) and the production of cytotoxic T-cells against tumors and viruses. Natural killer cells are involved in the elimination of pathogens. Most OCs cause multiple effects on the immune system.

Some OCs (PCDD/Fs, some PCBs) as well as TBT have direct effects on the thymus, causing atrophy. The most insidious effect of OCs on the immune system is to decrease an organism's resistance to infection or cancer. Immunosuppressive effects of OCs can be measured as reduced antibody production when exposed to a foreign antigen, changes in T-cell populations, suppression of the anti-sheep red blood cell (SRBC) plaque-forming responses due to suppression of T-cell responses, decreased delayed-type hypersensitivity, decreased resistance to virus infections, and decreased natural killer cell activity (Tryphonas 1994, Wong *et al.* 1992). All of these have been used as biomarkers for immunosuppressive effects in laboratory as well as wild animals.

Immunosuppressive effects may be one of the most sensitive and environmentally relevant effects of OCs (Vos and Luster 1989). Immunosuppression has been measured in harbour seals fed Baltic fish under semi-field experiments and was found to correlate with levels of PCDD/F and planar PCBs expressed as TEQs, for example (De Swart *et al.* 1995, Ross *et al.* 1995, 1996). OC-induced impaired immune function is suspected to have played a role in the morbillivirus-induced mass mortalities of harbour seals, grey seals, and striped dolphin populations in Europe during 1987-1991 (Hall *et al.* 1992, Aguilar and Borrell 1994, Ross *et al.* 1995, 1996). Immunosuppression is also suspected to be the cause of an increasing prevalence of moderate to severe intestinal ulcers in Baltic grey seals (Bergman *et al.* 1996).

### 6.5.2.5. **Adrenal effects**

Certain PCBs and DDTs have been shown to cause severe damage to the adrenal glands including hyperplasia and necrosis. Under experimental conditions, elevated blood glucocorticoid levels, as seen in hyperadrenocorticism, as well as ultrastructural signs of hyperactivity of the zona fasciculata have been found in mammals after exposure (Wassermann and Wassermann 1972, Wassermann *et al.* 1973a, 1979). A disease complex has been found in Baltic grey and harbour seals that have high levels of OCs, particularly PCBs. This includes reduced reproduction as well as adrenocortical hyperplasia with symptoms similar to Cushing's syndrome (Bergman and Olsson 1985). Adrenal effects have mainly been studied as pathological and histological changes in the adrenal glands of laboratory animals and wildlife. Other, less invasive methods include measuring plasma corticosterone levels and urinary excretion of cortisol.

### 6.5.2.6. **Thyroid and retinol effects**

Thyroid hormones control metabolism and growth and are essential for normal reproduction. They are also important for the development of normal brain functions during fetal

development (Morse *et al.* 1993, Morse 1995). Some effects on the thyroid seem to be related to the ability of some OCs, particularly hydroxy-PCB metabolites, to attach to the binding sites on the transthyretin-retinol-binding protein complex (TTR-RBP) in plasma, thereby disrupting the normal transport of the thyroid hormones triiodothyronine (T3) and thyroxine (T4) as well as vitamin A (retinol) to their target tissues. Disruption of this transport leads to lowered plasma levels of T3 and T4, which in turn may initiate increased release of thyroid stimulating hormone (TSH) to stimulate the thyroid gland to secrete more T3 and T4. This disruption of the feedback system for thyroid hormones may lead to thyroid hyperplasia (goiter), hypothyroidism, and disruptions of metabolism. This is indicated in herring gulls from the Great Lakes with high DDT, PCB, and mirex levels, which suffer from goiter and decreased T3 and T4 levels (Jeffries 1975). Another effect may also be related to the ability of some OCs, such as PCBs, to induce the production of liver enzymes involved in the breakdown of thyroid hormones. This also results in reduced amounts of thyroid hormones circulating in the plasma.

Imbalance in vitamin A (retinol and its esters) status can cause immunosuppression, susceptibility to cancers, and skin lesions, as well as disruption of reproduction, growth, and development. Some OCs (particularly PCDDs and PCBs) affect vitamin A metabolism and transport in several ways. Hepatic stores may be depleted (Håkansson *et al.* 1990), possibly due to enhanced retinol metabolism (Adams *et al.* 1990). Useful biomarkers for thyroid and retinol effects include measuring plasma levels of free and bound T3 and T4, as well as vitamin A levels.

#### 6.5.2.7. **Mutagenic and carcinogenic effects**

Current research supports a two-stage cancer model characterized by a primary mutagenic event (initiation) followed by a long latency period and second event (promotion) that leads to tumor growth. Most of the OCs dealt with in this assessment are not mutagenic. However, most of them are strong tumor promoters. Several OCs are associated with increased tumor prevalence found in highly exposed wildlife from areas outside of the Arctic. Observation of tumor prevalence is being carried out on some Arctic species.

#### 6.5.2.8. **Effects of mixtures**

Laboratory studies show that some OCs cause additive effects, others antagonize, and others cause synergistic responses together with other OCs. For example, PCDD/F, nPCB, and some mono-*ortho* PCBs are now considered to exert their toxic effects via the same mechanism, which involves binding to the aryl hydrocarbon (Ah) receptor, and the effects seem to be additive. This has facilitated the development of toxic equivalency factors (TEFs) for the different PCDD/F and dioxin-like PCB congeners where their individual toxicity is weighted against the most toxic congener, 2,3,7,8-TCDD (Tables 6·4 and 6·5) (NATO/CCMS 1988, Ahlborg *et al.* 1994). Using the TEF values and the individual congener concentrations, the total TCDD-equivalents (TEQ) can be calculated for a mixture of PCDD/Fs, nPCBs, and mono-*ortho* PCBs.

In another example, mink exposed experimentally to PCB (Clophen A50, Aroclor 1254), fractions of the technical product containing only non-*ortho* CBs, mono-*ortho* CBs, 2-4 *ortho* CBs, or bi- and tricyclic contaminants (PCDFs) or combinations of these had reduced reproduction. However, the group exposed to a combination of 2-4 *ortho* CBs, mono-*ortho* CBs, and the bi- and tricyclic contaminant fraction showed similar reproductive success as the controls. This indicates antagonistic effects between the CB fractions and the PCDF contaminants of the PCB (Kihlström *et al.* 1992).

Mixtures of two pesticides, such as endosulfan, dieldrin, chlordane, or toxaphene were found to be up to 1000 times more estrogenic than any individual substance, indicating synergistic effects (Arnold *et al.* 1996). However, these results could not be repeated by other laboratories (Ashby 1997, Ramamoorthy *et al.* 1997). The results have since been retracted (McLachlan 1997). A mixture of two hydroxylated PCB congeners has been found to cause synergistic estrogenic effects (Crews *et al.* 1995).

Table 6·4. TCDD toxic equivalency factors (TEFs) for some specific PCB congeners according to the WHO International Programme on Chemical Safety (IPCS) model (Ahlborg *et al.* 1994).

| Congener | CB (IUPAC number) | WHO-IPCS TEFs (1994) |
|---|---|---|
| 3,3'4,4' | 77 | 0.0005 |
| 3,3',4,4',5 | 126 | 0.1 |
| 3,3',4,4',5,5' | 169 | 0.01 |
| 2,3,3',4,4' | 105 | 0.0001 |
| 2,3',4,4',5 | 118 | 0.0001 |
| 2,3,4,4',5 | 114 | 0.0005 |
| 2',3,4,4',5 | 123 | 0.0001 |
| 2,3,3',4,4',5 | 156 | 0.0005 |
| 2,3,3',4,4',5' | 157 | 0.0005 |
| 2,3',4,4',5,5' | 167 | 0.00001 |
| 2,3,3',4,4',5,5' | 189 | 0.0001 |
| 2,2',3,3',4,4',5 | 170 | 0.0001 |
| 2,2',3,4,4',5,5' | 180 | 0.00001 |

Table 6·5. TCDD toxic equivalency factors (TEFs) for specific PCDD/PCDF congeners according to the international toxic equivalency factor (I-TEF) model (NATO/CCMS 1988).

| Congener | I-TEF |
|---|---|
| 2,3,7,8-TeCDD | 1.0 |
| 1,2,3,7,8-PeCDD | 0.5 |
| 1,2,3,4,7,8-HxCDD | 0.1 |
| 1,2,3,6,7,8-HxCDD | 0.1 |
| 1,2,3,7,8,9-HxCDD | 0.1 |
| 1,2,3,4,6,7,8-HpCDD | 0.01 |
| OCDD | 0.001 |
| 2,3,7,8-TeCDF | 0.1 |
| 1,2,3,7,8-PeCDF | 0.05 |
| 2,3,4,7,8-PeCDF | 0.5 |
| 1,2,3,4,7,8-HxCDF | 0.1 |
| 1,2,3,6,7,8-HxCDF | 0.1 |
| 1,2,3,7,8,9-HxCDF | 0.1 |
| 2,3,4,6,7,8-HxCDF | 0.1 |
| 1,2,3,4,6,7,8-HpCDF | 0.01 |
| 1,2,3,4,7,8,9-HpCDF | 0.01 |
| OCDF | 0.001 |

### 6.5.3. **Effects of specific POPs**

The following descriptions of the toxicology of different POPs are short reviews and are not meant to be comprehensive. They mainly cover chronic effects and effects that are relevant to the Arctic discussion. Results for controlled studies in laboratory animal species are discussed first. Where done, controlled studies using wild animal species under laboratory conditions are then presented. If field studies outside of the Arctic have been performed, these are then presented. Last come studies correlating specific effects with contaminant concentrations found from field studies of wild species. This last type of study has inherent problems as it is never possible to state that the contaminant measured is the cause of the effect, as there may be other contaminants not measured that covary with the one measured. Field studies and correlation studies of Arctic species are discussed under section 6.8.

#### 6.5.3.1. Halogenated industrial chemicals and by-products

##### 6.5.3.1.1. PCDDs, PCDFs, and PCBs

The 2,3,7,8-polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), as well as polychlorinated biphenyls (PCBs) substituted in the 3,3'4,4'-positions with no (nPCBs) or one *ortho* chlorine (mono-*ortho* PCBs) are among the most toxic OCs. The most toxic and best studied of these planar compounds is 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). These substances exert their toxic effects via a common mechanism that requires binding to the aryl hydrocarbon (Ah) receptor. They produce essentially the same spectrum of toxic effects in treated animals as TCDD, differing only in their potencies. All are less potent than TCDD. The non-*ortho* PCBs are more toxic than the mono-*ortho* congeners.

Although PCDD/F levels in the Arctic are low, PCB levels are higher and the effects of the dioxin-like PCBs may be of concern. Therefore, the toxic effects of PCDD/Fs are of interest in the context of the non-*ortho* and mono-*ortho* PCBs and these substance groups will be discussed together here.

TCDD exposure causes a wide spectrum of biochemical and pathological effects in laboratory animals (for reviews see Kimbrough 1974, Moore *et al*. 1979, McConnell 1980, Ahlborg *et al*. 1988, WHO 1989b, and Vanden Heuvel and Lucier 1993). Of the 209 PCBs, only 11 are considered to have dioxin-like properties. Other PCBs do not bind to the Ah receptor and exert their toxic effects by other mechanisms. Chronic exposure to commercial PCB elicits a broad spectrum of toxic responses including various indicators of liver toxicity, porphyria, weight loss, thymic atrophy, immunosuppression, reproductive and developmental toxicity, cancer, genotoxic responses, neurotoxicity, and endocrine disruption (reviewed in Safe 1994). The liver seems to be a major target organ for PCB toxicity.

• *Reproductive effects*

TCDD and other PCDD/Fs produce a range of reproductive and developmental effects (for a review see Peterson *et al*. 1993). TCDD is fetotoxic in several laboratory animals including mice, guinea pigs, hamsters, rabbits, and rats. TCDD affects reproduction in rats causing reduced number of pups, reduced survival rates for fetuses and pups, and decreased growth in surviving pups and their offspring. Reproductive effects are seen in both males and females. Prenatal exposure to TCDD and several individual PCDFs causes cleft palate in mice at doses that do not cause other maternal or fetal toxic effects (Hassoun *et al*. 1984, Weber *et al*. 1984, Birnbaum *et al*. 1987).

Rats exposed prenatally to TCDD (Mably *et al*. 1991, 1992a, 1992b, 1992c, Peterson *et al*. 1992) had reduced testosterone levels, reduced sperm production, and demasculinized and feminized sexual behaviors. Some effects were present at one-time maternal doses as low as 64 pg TCDD/g body weight. Reduced fertility and reproductive capacity are also seen in rhesus monkeys treated with TCDD (see Couture *et al*. 1990 for review).

TCDD and PCBs have both been shown to decrease estrogen hormone receptor levels in rats (Korach *et al*. 1987, Romkes *et al*. 1987, Umbreit and Gallo 1988). Mink treated with technical PCB had decreased progesterone levels (Byrne *et al*. 1975). PCB exposure in laboratory mammals causes a wide range of reproductive effects (Drill *et al*. 1982), including embryo- and fetotoxicity and decreased offspring survival in rodents, as well as abnormalities in the estrus cycle and sex hormone levels of treated monkeys (for reviews see Morissey and Schwetz 1989, Delzell *et al*. 1994a). Exposure to organochlorines, particularly technical PCB, causes reproductive failure in mink (Platonow and Karstad 1973, Aulerich and Ringer 1977, Jensen *et al*. 1977, Bleavins *et al*. 1980, Ringer 1981, Eisler 1986). The non-*ortho* PCB 3,3',4,4',5,5'-hexachlorobiphenyl (CB 169), given in low chronic doses, severely impairs reproduction in the mink (Aulerich *et al*. 1985). PCB fed to mink resulted in reduced litter sizes when they were exposed to technical PCB (Clophen A50, Aroclor 1254) or a combination of non-*ortho* and mono-*ortho* PCB congeners (Kihlström *et al*. 1992).

Numerous animal and human studies demonstrate that PCB exposure leads to neurobehavioral effects, particularly in offspring, and that these effects are not dependent on binding to the Ah receptor (for reviews see Tilson *et al*. 1990, Seegal and Shain 1992). Holene *et al*. (1995) studied pre- and postnatal effects on offspring of pregnant female rats treated with subtoxic concentrations of CB 118 or 3,3',4,4',5,-PeCB (CB 126). Significant behavioral alterations, including hyperactivity and impaired visual discrimination learning, were seen for both congeners with CB 126 being the most potent. In another study, male rats were treated with 2,2',4,4',5,5'-HxCB (CB 153) or CB 126 during lactation to study behavioral effects of exposure from mother's milk on the offspring (Holene *et al*. 1996). Significant increases in hyperactivity were seen after exposure to both congeners. In addition, the offspring exposed to CB 153 showed other changes in behavior. In a recent study (Holene *et al*. unpubl. results), female rats were exposed to CB 153 through mothers milk. The females showed a significant sex-specific behavioral response, being less sensitive since only deficient acquisition of time discrimination was seen.

Chronic exposure to TCDD leads to decreased egg production and decreased egg hatchability in ring-necked pheasants (*Phasanius colchicus*) (Nosek *et al*. 1992). Increased embryotoxicity is seen in TCDD-treated eggs of chickens (Brunström and Andersson 1988) and ring-necked pheasant (Nosek *et al*. 1993) and cardiovascular malformations are seen in treated chick embryos (Cheung *et al*. 1981a, 1981b). Chickens (white leghorn strain) fed Aroclor 1232, 1242, 1248 or 1254 had decreased egg production. Aroclor 1221 and 1268 had no effect (Lillie *et al*. 1974). Embryonic mortality was also seen when chickens are treated with Aroclor 1248 (Peakall 1975, 1986). Hatchability was reduced in ring doves (*Streptopelia risoria*) fed Aroclor 1254 (Peakall and Peakall 1973). Ring doves treated with the nPCB 3,3',4,4'-TeCB (CB 77) before mating showed retarded egg laying and high embryo mortality (Spear *et al*. 1989). Herring gulls (*Larus argentatus*), bald eagles (*Haliaeetus leucocephalus*) and double-crested cormorants (*Phalacrocorax auritus*) fed Great Lakes fish in the laboratory have reduced reproductive capacity (Ludwig *et al*. 1993).

Eye and beak deformities are seen in chick embryos treated with specific planar PCBs (Brunström and Andersson 1988, Brunström 1990). Large differences in sensitivity to the embryotoxic effects of CB 77 are also seen between different bird species (Brunström and Reutergårdh 1986, Brunström and Lund 1988, Brunström 1988). All three nPCBs (Brunström 1989, 1992a) as well as the mono-*ortho* PCBs are embryotoxic in chick embryos (Brunström 1991).

Two hydroxylated PCBs have been shown to be estrogenic in turtles (Crews *et al*. 1995). Early life stages of fish are more sensitive than adults to the effects of PCBs and PCDD/Fs. In fish, TCDD caused decreased egg hatchability, increased sac fry mortality, and edema in lake, brook (*Salvelinus fontinalis*), and rainbow trout (Spitsbergen *et al*. 1991, Walker *et al*. 1991, Walker and Peterson 1991, 1992, 1994) and re-

duced number of eggs and lethal malformations in young zebrafish (*Brachydanio rerio*) (Wannemacher *et al*. 1992). The same effects are seen in lake trout eggs which received their TCDD exposure via maternal transfer and these effects were dose-dependent (Walker *et al*. 1994).

Exposure to TCDD in water leads to dose-related reductions in egg development, reduced growth in fry, and increased fry mortality in pike (*Esox lucius*) (Helder 1980) and rainbow trout (*Salmo gairdneri*) (Helder 1981). Similar exposure in the Japanese medaka (*Oryzias latipes*) causes reduced egg development, decreased embryo survival, increased vascular hemorrhage, and edema (Wisk and Cooper 1990a, 1990b). Lake trout are the most sensitive species studied so far in this respect with a lowest-adverse-effect-level (LOAEL) of 40 pg TCDD/g egg and an $LD_{50}$ of 65 pg TCDD/g egg (Spitsbergen *et al*. 1991, Walker *et al*. 1991). Several other PCDD and PCDF congeners cause similar types of dose-related egg mortality in fish as TCDD (Wisk and Cooper 1990b, Walker and Peterson 1991).

CB 77 and CB 126 cause similar types of dose-related egg mortality in fish as TCDD (Wisk and Cooper 1990b, Walker and Peterson 1991). Adult female white perch (*Morone americana*) exposed to CB 77 showed reduced sexual maturation, reduced gonad size, and decreased larval survival (Monosson *et al*. 1994).

Mink fed Great Lakes fish containing high concentrations of OCs such as PCBs, PCDD/Fs, and DDT showed reproductive failure (Aulerich *et al*. 1973, Aulerich and Ringer 1977). Captive harbour seals fed fish containing high PCB levels had reduced pup production compared to seals fed less contaminated fish in a semi-field study in the Netherlands (Reijnders 1986).

It has been suggested that PCB was the cause of the decline of the European otter (*Lutra lutra*) population after the 1950s, including the otter population living north of the Arctic Circle in Sweden (Sandegren *et al*. 1980, Mason and Macdonald 1986, Olsson and Sandegren 1991a, 1991b). Reductions in wild mink and otter populations in North America may also be correlated with high levels of organochlorines, particularly PCB (Wren 1991). In support of this, a recent study has shown that the northern Swedish otter populations as well as the Swedish mink population have recovered since the 1980s and that these improvements are significantly correlated with decreasing PCB concentrations in the otter and in the food of both otter and mink (Olsson *et al*. 1996a).

High concentrations of OCs, especially PCB, are correlated with reduced fertility in Baltic Sea grey, harbour, and ringed seals (Hook and Johnels 1972, Olsson *et al*. 1975, 1992a, Helle *et al*. 1976a, 1976b, Olsson 1978, Helle 1980, 1981, 1986, Bergman *et al*. 1981, Almkvist 1982) and harbour seals from the Dutch part of the Wadden Sea (Reijnders 1978, 1979, 1980, 1986). An increase in aborted fetuses is associated with high PCB and DDT concentrations in California sea lions (*Zalophus californianus*) (De Long *et al*. 1973, Gilmartin *et al*. 1976).

Recent studies show that PCB, PCDD/F, and DDT levels have declined in the Baltic Sea, with concomitant increases in grey seal populations in the northern parts of the Baltic and harbour seal populations in the southern parts of the Baltic (Bergman *et al*. 1996). The ringed seal population in the northern parts of the Baltic still exhibits a fairly high incidence of uterine occlusions even in young animals (Helle 1995) and the population trend is still uncertain (Härkönen *et al*. 1996). Recent findings based on autopsies of Baltic grey seals have also revealed increased pregnancy rates and population censuses have shown population increases after the sudden drop in the environmental PCB concentrations in the 1970s. However, the same data also disclosed an increased prevalence of intestinal ulcers when comparing data from 1977-1986 with data from 1987-1996 (Bergman *et al*. 1996).

The beluga population in the St. Lawrence River estuary has low proportions of calves and juveniles and this is thought to be the result of high organochlorine concentrations, particularly PCB (Martineau *et al*. 1987).

There are similarities in toxic effects seen in colonial fish-eating birds in the Great Lakes area of North America as those seen in chickens accidentally poisoned with 1,2,3,7,8,9-HxCDD (reviewed in Gilbertson *et al*. 1991). These effects are correlated with the concentrations of PCDDs/PCDFs and planar PCBs found in the different bird species studied (reviewed in Giesy *et al*. 1994a, 1994b). The effects in birds include reduced egg hatching, embryotoxicity, deformities, and impaired parental behavior (Hoffman *et al*. 1987, Kubiak *et al*. 1989, Tillitt *et al*. 1989, 1991, 1992, 1993, Yamashita *et al*. 1993).

Numerous piscivorous bird species, such as bald eagle, herring gull, double-crested cormorant, black-crowned night heron (*Nycticorax nycticorax*), Caspian tern (*Hydroprogne caspia*), Forster's tern (*Sterna forsteri*), osprey (*Pandion haliaetus*), ring-billed gull (*Larus delawarensis*), and common tern (*Sterna hirundo*), from the Great Lakes region have shown decreased reproductive performance and population declines which may be associated with high concentrations of PCBs, particularly the non-*ortho* and mono-*ortho* congeners (reviewed in Giesy *et al*. 1994a, 1994b, Delzell *et al*. 1994a, Bosveld and van den Berg 1994, Barron *et al*. 1995).

Significant negative correlations have been found between PCDD/F concentrations and testosterone levels in male and female white sucker (*Catostomus commersoni*) (van den Heuvel *et al*. 1994). Reduced reproduction has been seen in chinook salmon (*Oncorhynchus tshawytscha*) and lake trout from Lake Michigan (Mac 1988). A significant negative correlation was found between egg hatching success in chinook salmon and PCB concentrations in the eggs (Ankley *et al*. 1991). Reduced hatching and an increase in hatchling deformities were found to correlate well with concentrations of CB 105 in the common snapping turtle (*Chelydra serpentina serpentina*) (Bishop *et al*. 1991).

• *Cytochrome P450-dependent monooxygenases*

TCDD induces cytochrome P450 1A1, increasing EROD and AHH activity in a range of laboratory animals (rodents, cattle). AHH and EROD induction are also seen in laboratory animals after exposure to other 2,3,7,8-chlorinated PCDD/F congeners and planar PCBs (for reviews, see Ahlborg *et al*. 1988, 1992, WHO 1989b, Safe 1990, 1994).

Aroclor 1242 fed to male and female mink induced cytochrome P450, but ferrets fed the same diet showed no effects. AHH and ethoxycoumarin-O-deethylase (ECOD) activities were induced in both species. Aroclor 1016 had no effects (Shull *et al*. 1982). EROD, AHH, and APND activities were measured in female mink treated with PCB before mating and in their kits after birth (Brunström 1992b). Technical PCB (Clophen A50 and Aroclor 1254) and fractions containing non- and mono-*ortho* PCB induced EROD and AHH activity, whereas APND was induced by the technical PCB and fractions containing mono- and di-*ortho* PCB. The response was stronger in the kits born after fetal exposure than in the adult females.

Female mink treated with CB 77 had severe anorexia and intestinal hemorrhage (Gillette *et al*. 1987a, 1987b) and significant increases in cytochrome P450 content, but no changes in ECOD, AHH, APND or AE activities. No changes were seen after treatment with 2,2',4,4'-TeCB (CB 47).

All 2,3,7,8-substituted PCDDs/PCDFs (tetra- to heptachlorinated) and non- and mono-*ortho* PCBs that have been tested in bird embryos have led to liver enzyme induction measured as EROD (Brunström and Andersson 1988, Nikolaidis *et al*. 1988a, Brunström 1989, 1990, 1991, 1992a, Bosveld *et al*. 1992).

Pigeons (*Columba livia*) injected with Aroclor 1254 showed significant increases in cytochrome P450 proteins, AE, EROD and several other MFOs (Borlakoglu *et al*. 1991). Japanese quail (*Coturnix coturnix japonica*) exposed chronically to CB 105, CB 126, or CB 153 had increased EROD and APND activity, however the response was strongest for CB 105. For kestrels (*Falco sparverius*), chronic doses of CB 126 led to increased EROD and AE activity, CB 105 led to increased APND activity, and CB 153 led to increased APND and AE activity (Elliott *et al*. 1990, 1991). Eider ducklings (*Somateria mollisima*) given CB 77 had increased EROD and PROD activity and cytochrome P450 1A levels. Treatment with Clophen A50 only increased cytochrome P450 1A levels (Rozemeijer *et al*. 1992, Murk *et al*. 1994).

TCDD causes liver enzyme induction in experimentally treated fish (Janz and Metcalfe 1991, van der Weiden *et al*. 1992, 1993, 1994a, 1994b, Hektoen *et al*. 1994) as well as in cultured rainbow trout hepatocytes (Pesonen *et al*. 1992), and in a trout hepatoma cell line (Lorenzon and Okey 1990). Several PCDDs and PCDFs cause EROD induction in rainbow trout (Muir *et al*. 1990d), scup (*Stenotomus chrysops*) (Gooch *et al*. 1989, Smolowitz *et al*. 1991), and carp (van der Weiden *et al*. 1994a).

Liver enzyme induction and increased EROD activity are also seen after treatment with individual nPCBs and mono-*ortho* PCBs in rainbow trout (Skaare *et al*. 1991), in winter flounder (*Pleuronectes americanus*) (Monosson and Stegeman 1991), in adult and larval fathead minnow (*Pimephales promelas*) (Lindstrom-Seppa *et al*. 1994), in scup (Gooch *et al*. 1989, Smolowitz *et al*. 1991), in a fish hepatoma cell line (Hahn *et al*. 1993), and in carp (*Cyprinus carpio*) (van der Weiden *et al*. 1994a). Rainbow trout (*Oncorhynchus mykiss*) treated with CB 105 showed increased EROD activity and increased cytochrome P450 1A1 levels, whereas immature cod (*Gadus morhua*) showed no increased activity in either enzyme or cytochrome P450 1A1 levels (Bernhoft *et al*. 1994).

A number of wild animal species have shown associations between EROD and/or AHH activity and concentrations of dioxin-like substances, including seals (Addison *et al*. 1988), various birds (Hoffman *et al*. 1987, Bellward *et al*. 1990, Yamashita *et al*. 1992, Bosveld *et al*. 1993, van den Berg *et al*. 1994), and fish (Monosson and Stegeman 1991, Servos *et al*. 1994, van den Heuvel *et al*. 1994).

A significant correlation was found between EROD activity and PCB concentrations in the liver of short-finned pilot whale from the western North Pacific (Tanabe and Tatsukawa 1991). Correlations have also been found between EROD, PROD and PCB concentrations in northern fur seal (*Callorhinus ursinus*) from northern Japan (Tanabe *et al*. 1994a, 1994b).

Levels of cytochrome P450 1A1 correlate with concentrations of PCBs in shag, cormorant, razorbill, guillemot, puffin, and Manx shearwater (Ronis *et al*. 1989). EROD and AHH activities as well as cytochrome P450 1A and 2B levels significantly correlate with total PCB concentrations in black-crowned night herons (Rattner *et al*. 1993).

Laboratory studies in rats show that methylsulfone metabolites, particularly the 3-Me$SO_2$ forms of several PCB congeners, also induce cytochromes P450 2B1 and 2B2, and lead to increased APND, ECOD, and AHH activity (Kato *et al*. 1993a, 1993b).

• *Porphyria*

TCDD is a potent inducer of porphyria in laboratory rodents (for review see WHO 1989b) and a number of PCBs also cause porphyria in experimental animals (Strik *et al*. 1980). PCBs induce accumulation of porphyrins in Japanese quail (Vos *et al*. 1971, Miranda *et al*. 1983) and CB 105 is more potent than CB 126 or CB 153 (Elliott *et al*. 1990). Female kestrels fed chronically with the same three PCB congeners showed no porphyrin accumulation (Elliott *et al*. 1991).

Porphyrin levels were measured in livers of herring gulls from different sites on the Great Lakes and on the Atlantic coast and related to concentrations of organochlorines. Between-site porphyrin levels were found to correlate with concentrations of PCDDs and DDE (Fox *et al*. 1988).

• *Immunosuppression*

PCDD/Fs have been shown to adversely affect the immune system in laboratory animals (Vos and Luster 1989). Exposure to high levels of TCDD leads to thymic atrophy, and exposure to lower levels suppresses the immune system. For reviews, see Holsapple *et al*. (1991a, 1991b). Immunosuppression is also seen after exposure to several other PCDDs and PCDFs (Luster *et al*. 1979, Kerkvliet *et al*. 1985, Kerkvliet and Brauner 1987, Harper *et al*. 1993). Effects on immune function occur at exposure levels that do not produce signs of toxicity. Changes occur in both cell-mediated and humoral immunity in both laboratory rodents and chickens, and in non-human primates exposed to TCDD. The fetus and lactating newborn animals are particularly sensitive to the immunosuppressive effects of TCDD. Antibody production is affected and host defense mechanisms are suppressed in treated animals, increasing illness and mortality rates when they are exposed to infectious agents.

PCBs also adversely affect the immune system in laboratory animals (Wassermann *et al*. 1973b, 1979, Vos and Luster 1989). In some cases, this immunosuppression has increased the animal's susceptibility to infections (Vos *et al*. 1988). Immunosuppression is also seen in mice after exposure to CB 126 and CB 169 (Harper *et al*. 1993). In birds, TCDD, CB 77, CB 126, CB 169, CB 105, and CB 118 all impair lymphoid tissue development in the bursa of Fabricius and in the thymus of chicken embryos (Nikolaidis *et al*. 1988a, 1988b, Andersson *et al*. 1991).

Significant impairment of natural killer (NK) cell activity, *in vitro* T-lymphocyte function, antigen-specific *in vitro* lymphocyte proliferative responses, and *in vivo* delayed-type hypersensitivity (DTH) responses were found in captive harbour seals fed highly PCB-contaminated Baltic Sea herring compared to seals fed relatively uncontaminated Atlantic Ocean herring (De Swart *et al*. 1994, 1995, Ross *et al*. 1995, 1996). Immunosuppression may have increased mortality of harbour seals, grey seals, striped dolphins, and bottlenose dolphins in the phocine distemper (morbillivirus) epizootics that occurred in the North Sea, Mediterranean Sea, and Gulf of Mexico in the 1987-1991 period (Hall *et al*. 1992, Aguilar and Borrell 1994, Lahvis *et al*. 1995). Signs of immunosuppression are also seen in heavily contaminated beluga whales from the St. Lawrence estuary (De Guise *et al*. 1995). No correlation has been found between blood ΣPCB levels and serum IgG levels in grey seal from Norway and Canada having low PCB levels (Sørmo, Jensen, Skaare and Larsen 1995 unpubl. results, Sørmo 1996).

• *Skeletal changes and adrenal hyperplasia*
Certain PCB congeners are taken up and retained in the adrenocortical tissue of laboratory rodents (Brandt 1977, Brandt *et al.* 1978). $MeSO_2$-PCB metabolites are retained in the lung, adrenals, and fetuses of mice (Lund *et al.* 1984, Brandt *et al.* 1985, Darnerud *et al.* 1986). Primarily 3- and 4-$MeSO_2$-2,2',4',5,5'-PeCB, metabolites of 2,2',4,5,5'-PeCB (CB 101), are found in mink exposed to technical PCB or fractions containing 2-4-*ortho* PCB (Bergman *et al.* 1992b). These are the same as those previously found in Baltic grey seals (Haraguchi *et al.* 1992). Mink treated with Clophen A50 or Aroclor 1254 had increased urinary excretion of cortisol (Madej *et al.* 1992).

Baltic grey and ringed seals suffer from a disease complex caused by hyperadrenocortism (Bergman and Olsson 1985, 1989). The pathological changes include skull-bone lesions, adrenal hyperplasia, glomerulopathy, hypotrichosis, regional alopecia, and uterine occlusion. The prevalence of skull-bone lesions in grey and harbour seals in the Baltic Sea coincides with high PCB and DDT concentrations and is suggested to be the result of PCB toxicity (Bergman *et al.* 1992a, Mortensen *et al.* 1992).

Several $MeSO_2$-PCB metabolites accumulate in the adrenal tissue of Baltic grey seals (Jensen and Jansson 1976, Haraguchi *et al.* 1990, 1992). More than twenty-two $MeSO_2$-PCB metabolites have been identified in Canadian polar bears (*Ursus maritimus*) (Bergman *et al.* 1994b, Letcher *et al.* 1994, 1995). $MeSO_2$-PCB metabolites have also been identified in otter and wild mink from northern Sweden, beluga from the St. Lawrence estuary, as well as Arctic ringed seal (Bergman *et al.* 1994b, Letcher 1996).

• *Thyroid and retinol effects*
Rats exposed to TCDD have reduced liver vitamin A levels (Thunberg *et al.* 1980). Laboratory and field studies have shown that PCB exposure leads to drastic reductions in plasma concentrations of vitamin A (retinol) (Innami *et al.* 1974) and thyroid hormones (Brouwer *et al.* 1989, Durham and Brouwer 1989). This is thought to be due to metabolism of some PCBs to hydroxylated metabolites which in turn bind effectively to transthyretin, the major transport protein for retinol and the thyroid hormones, thyroxine (T4) and triiodothyronine (T3) (Brouwer and van den Berg 1986, Brouwer *et al.* 1988, 1990). 4-OH-3,3',4,4'-TeCB, a metabolite of CB 77 binds three times more effectively to TTR, and 4-OH-2',3,3',4',5-PeCB (CB 105) binds six times more effectively to TTR than thyroxine (Bergman *et al.* 1994a). TCDD and nPCBs also induce the enzymes responsible for hepatic and brain thyroid hormone metabolism, leading to hypothyroidism in fetal and neonatal rats (Morse *et al.* 1993, Morse 1995).

Treatment with CB77 causes thyroid hypertrophy, thyroid hyperplasia, reduced T4 and T3 levels, and increased TSH in marmoset monkeys (van den Berg *et al.* 1988). PCB exposure generally increased blood thyroxine concentrations in mink except during estrus and the reproductive season (Byrne *et al.* 1975). Exposure to non- and some mono-*ortho* PCBs causes vitamin A reduction in mink (Håkansson *et al.* 1992).

In a semi-field study, harbour seal (*Phoca vitulina*) were fed fish containing low or high levels of PCB. Those fed the more highly contaminated fish had lower levels of plasma retinol, total and free T4, and total T3 than those fed the cleaner fish (Brouwer *et al.* 1989).

Treatment of different bird species with technical PCB mixtures often causes an increase in thyroid gland weights (reviewed in Peakall 1986) as well as reduction of thyroid hormone levels (Jeffries and French 1971, Grässle and Biessmann 1982). Ring doves exposed to CB 77 before mating were studied for effects on serum retinol and retinoids in the eggs (Spear *et al.* 1989). Exposed females that laid viable eggs had higher serum retinol levels than those that laid eggs that failed to hatch. Eggs from exposed females had reduced levels of yolk retinol and retinyl palmitate compared to controls.

Herring gull eggs collected from several breeding colonies on the Great Lakes were analyzed for yolk retinoids (Spear *et al.* 1990). Correlations were found between the molar ratios of retinol to retinyl palmitate and (1) 2,3,7,8-TCDD concentration, (2) TCDD-equivalents calculated from PCDD/Fs, and (3) the sum of PCDD and PCDF concentrations. Significant correlations were found between reductions in free plasma thyroxine levels and concentrations of PCDD/Fs expressed as TCDD-equivalents in cormorants (van den Berg *et al.* 1994). Eider ducklings exposed to CB 77 and Clophen A50 had decreased plasma T3 and retinol levels, and CB 77 also decreased levels of plasma T4 and hepatic retinyl palmitate (Murk *et al.* 1994). Correlations have been found between PCB levels and reduced thyroid hormone levels in common tern (Murk *et al.* 1993) and cormorants (Craane *et al.* 1991).

Blood samples were collected from 17 grey seal pups at Froan, Norway (64°10'N, 09°20'E) (Jenssen *et al.* 1995a). PCB (sum of 22 congeners) and DDT concentrations were determined as well as plasma concentrations of thyroid hormones. The two pups with the highest PCB concentrations also had the lowest thyroxine concentrations, but no significant correlation was found between PCB and thyroxine concentrations for the entire data set. In a larger study, blood samples from grey seal pups at Froan were analyzed for PCBs, retinol, free and total thyroxin (T4), and free and total triiodothyronine (T3) (Jenssen *et al.* 1995b). A significant negative correlation was found between blood PCB concentrations and plasma retinol concentrations. A borderline significant negative correlation was found between PCB concentrations and the ratio of total T4 to free T4. This ratio would be expected to be reduced if hydroxylated PCBs bind to transthyretin and hinder T4 from binding. The mean PCB levels in blood were low, 1100 ng/g lw, and are similar to or lower than the lipid weight levels found in Arctic seals.

• *Cancer*
TCDD is a potent carcinogen in laboratory animals (Kociba *et al.* 1978, NIH 1982). It is a powerful tumor promoter in both cell culture and animal studies, especially in females (Pitot *et al.* 1980, 1987, Graham *et al.* 1988). For reviews on the carcinogenicity of TCDD see WHO (1989b), Ahlborg (1993), Lucier *et al.* (1993) and Huff *et al.* (1994). Several other PCDD/F congeners, as well as planar PCBs, have been shown to be potent tumor promoters (NIH 1980a, 1980b, Silberhorn *et al.* 1990, Sargent *et al.* 1991, Wærn *et al.* 1991, Hemming *et al.* 1993, 1994). PCB mixtures and congeners are not mutagenic when tested using the Ames test and no DNA adducts are formed (reviewed in Safe 1994). PCB mixtures are cancer promoters, however.

#### 6.5.3.1.2. **Polybrominated diphenyl ethers (PBDEs)**

Very little is known about the toxicity of PBDEs as only limited studies have been carried out so far.

• *Cytochrome P450-dependent monooxygenases*
The commercial product Bromkal 70-5DE, containing 2,2',4,4'-TeBDE, 2,2',4,4',5-PeBDE, and an unidentified

PeBDE, has weak dioxin-like activity expressed as EROD in rat hepatoma cells (Hanberg *et al*. 1991). Bromkal 70 has also been shown to induce cytochrome P450 levels in rats, leading to increased EROD activity, and the PBDEs in Bromkal 70 were concluded to be mixed-type inducers (von Meyerinck *et al*. 1990). EROD activity is weakly induced in rainbow trout embryos microinjected with Bromkal 70-5DE (Norrgren *et al*. 1993, Holm *et al*. in press) and in three-spined stickleback (*Gasterosteus aculeatus*) fed with Bromkal 70-5DE (Holm *et al*. 1993). For a review see also WHO (1994).

• *Thyroid effects*
Rats fed PeBDE showed thyroid hyperplasia (WHO 1994).

### 6.5.3.2. **Persistent organochlorine pesticides**

Most of the cyclic, aromatic, chlorinated pesticides are neurotoxic. They alter sodium and potassium ion flux across nerve cell membranes and interfere with nerve impulse transmission. In general, they are more toxic to insects than to other animals. All pesticides discussed here produce adverse effects on the liver after chronic exposure, often including hypertrophy and various changes in histopathology.

#### 6.5.3.2.1. **Aldrin and dieldrin**

Aldrin is oxidized to dieldrin in plants and animals and is rarely found in samples from living organisms (WHO 1989c). Chronic exposure leads to signs of liver toxicity. Dieldrin is more toxic to fish than aldrin.

• *Reproductive effects*
Dieldrin administration produces reduced fertility in male and female rats fed 15 ng/g body weight for 300 days (Thomas and Colborn 1992). High doses cause reduced litter size in mice and increased pup mortality in mice, rats, and dogs (reviewed in WHO 1989c). Different bird species dosed with dieldrin generally show no effects on reproduction unless the adults have been treated with sufficiently high doses to cause toxic effects such as reduced food intake (WHO 1989c).

Rainbow trout eggs exposed to dieldrin in water showed increased mortality only for the early fry stage ($LC_{50}$ of 3000 ng/L) (Van Leeuwen 1986). Rainbow trout embryos, alevins, and fry were exposed to a range of dieldrin concentrations (12-52 000 ng/L) (Chadwick and Shumway 1970). There was no effect on egg hatchability but newly hatched alevins exposed to higher concentrations had lower weights. Survival was reduced for alevins and trout fry at water concentrations above 390 ng/L. Flounder exposed to dieldrin showed reduced fertilization of eggs (Smith and Cole 1973).

Dieldrin has been shown to be estrogenic when measured using the *in vitro* E-screen test (Soto *et al*. 1994).

• *Cytochrome P450-dependent monooxygenases*
Both aldrin and dieldrin treatment lead to increased APND and AE activity and increased cytochrome P450 in rodents, dogs, and rhesus monkeys (Zavon and Stemmer 1975, Campbell *et al*. 1983). Dieldrin also induces monooxygenases that hydroxylate testosterone (Haake *et al*. 1987).

• *Immunosuppression*
Dieldrin has been shown to adversely affect the immune system of laboratory animals (Fournier *et al*. 1988). These effects include impaired macrophage antigen processing, reduced tumor cell killing ability, suppression of humoral immunity, and reduced host resistance to infections (Loose *et al*. 1981, Loose 1982, Wong *et al*. 1992).

• *Cancer*
Aldrin and dieldrin are not mutagenic in a variety of test systems. Dieldrin treatment causes increases in benign and malignant liver tumors in mice but not other rodents (reviewed in WHO 1989c).

#### 6.5.3.2.2. **Chlordane**

• *Reproductive effects*
Rats treated with chlordane showed reduced rates of mating, reduction in viable litters, and increased offspring mortality (Ambrose *et al*. 1953). Treated mice fed chlordane at 100 μg/g for six generations showed decreased offspring survival in the first two generations and no offspring in the third generation (Keplinger *et al*. 1968). Treatment with 25 μg/g once a week for three weeks in mice led to a reduction in the number of females that became pregnant (Welch *et al*. 1971). Chlordane has a damaging effect on spermatogenesis and causes degeneration of testicular tissue in mice (Balash *et al*. 1987). Chlordane caused decreased progesterone binding in the rabbit uterus and the shell gland in ducks (Lundholm 1988).

• *Cytochrome P450-dependent monooxygenases*
Chlordane induces cytochrome P450 levels and enzyme activities (WHO 1984a). It also induces monooxygenases that hydroxylate testosterone.

• *Immunosuppression*
Pregnant female mice treated with chlordane had offspring with decreased cell-mediated immune competence (Spyker-Cranmer *et al*. 1982).

• *Cancer*
Chlordane does not appear to be mutagenic. However, it does display characteristics that are common for other substances that act as tumor promoters. Studies in mice indicate that chlordane treatment leads to increases in liver tumors (WHO 1984a).

#### 6.5.3.2.3. **DDT**

DDT products contain primarily *p,p'*-DDT and some *o,p'*-DDT. These DDTs are metabolized to the comparable DDDs and DDEs in many organisms and it is primarily these metabolites that are found at higher trophic levels.

• *Reproductive and developmental effects*
No reproductive failure was seen in mink treated with DDT (Kihlström *et al*. 1976). Chicken eggs injected with DDT showed no change in hatchability, but chick survival was reduced (Dunachie and Fletcher 1969).

DDT has long been known to affect bird reproduction (Ratcliffe 1967, Longcore *et al*. 1971, Postupalsky 1971, Wiemayer *et al*. 1972, 1975, 1978, Cooke 1973, Peakall *et al*. 1973, 1975, Newton and Bogan 1974, Longcore and Stendell 1977). *p,p'*-DDE has been shown to cause significant egg-shell thinning in treated birds and predatory species have been found to be the most sensitive (Bitman *et al*. 1969, Heath *et al*. 1969, Longcore *et al*. 1971, Peakall *et al*. 1973, Longcore and Stendell 1977, Vangilder and Peterle 1980). For example, American kestrels were fed different doses of DDE and DDE concentrations in the eggs were found to correlate well with dietary intake. These concentrations

were found to correlate significantly with the degree of eggshell thinning (Lincer 1975). The same relationship was found when kestrel eggs from the field were studied.

Eggshell thinning above 18% is associated with population decreases (Hickey and Andersson 1968, Peakall and Kiff 1988). Suggested mechanisms for *p,p'*-DDE's effects on eggshell thinning are that it inhibits calcium ion ATP-ase and possibly also carbonic anhydrase (Lundholm 1987).

*o,p'*-DDT is strongly estrogenic and its metabolites *o,p'*-DDE and *o,p'*-DDD are also estrogenic (Kupfer and Bulger 1980). *o,p'*-DDT causes uterine proliferation in rats and binds to rat and human estrogen receptors (Kupfer and Bulger 1980, Mason and Schulte 1980). *o,p'*-DDE also strongly inhibits the binding of progesterone to its receptor (Lundholm 1988).

*p,p'*-DDE has been shown to be antiandrogenic, with the ability to block androgen-stimulated gene transcription and androgen effects in the rat (Kelce *et al*. 1995). It is not estrogenic, however.

Reduced survival after hatching was seen in Japanese quail treated *in ovo* with 6.25-10 μg *o,p'*-DDT/g egg (Bryan *et al*. 1989). Japanese quail treated *in ovo* and subsequently mated also had reduced egg hatchability and both males and females showed significant reductions in normal sexual behavior. Male California (*Larus californicus*) and western gull (*Larus occidentalis*) eggs injected with 2 μg/g *o,p'*-DDT showed feminized embryos (Fry and Toone 1981, Fry *et al*. 1987). Female California and western gull eggs injected with 20 μg/g *o,p'*-DDT developed both left and right oviducts (development of a right oviduct is abnormal).

Japanese quail embryos treated with *p,p'*-DDT had reduced survival (1.75-5 μg/g egg) and when subsequently mated, also had reduced egg hatchability (1.75 μg/g egg) (Bryan *et al*. 1989). *p,p'*-DDE fed to adult birds also causes a wide range of reproductive behavioral disturbances (reviewed in WHO 1989a). Male California and western gull eggs injected with 20 μg/g *p,p'*-DDE were feminized (Fry and Toone 1981).

Killifish (*Fundulus heteroclitus*) exposed to DDT in water showed reduced fertilization and delayed development of fertilized eggs (Crawford and Guarino 1976). Flounder (*Pseudopleuronectes americanus*) embryos exposed to DDT in water showed abnormal gastrulation and high incidences of vertebral deformities (Smith and Cole 1973). Water concentrations greater than 0.5 μg DDT/L are toxic to early life stages of coho salmon (*Oncorhynchus kisutch*) (Halter and Johnson 1974).

Atlantic salmon (*Salmo salar*) eggs exposed to different doses of DDT in water resulted in fry with impaired balance and retarded behavioral development at the highest exposures (50 and 100 μg/L) (Dill and Saunders 1974). Other behavioral changes have also been observed in adult fish, including reduced righting ability and changes in temperature selection and exploratory behavior (reviewed in WHO 1989a).

A significant negative correlation was found between serum testosterone levels and DDE-concentrations in blubber from Dall's porpoises (*Phocoenoides dalli*) in the northwest Pacific Ocean (Subramanian *et al*. 1987). The authors hypothesize that this may be due to induction of the cytochrome P450 system leading to an increased metabolism of testosterone.

Significant correlations between DDE concentrations and eggshell thinning have been seen in a number of wild bird species (see for example Ratcliffe 1967, Koeman and van Genderen 1972, Cooke 1973, Koeman *et al*. 1973, Parslow *et al*. 1973, Weseloh *et al*. 1983, Elliott *et al*. 1988).

DDT exposure, particularly to *p,p'*-DDT, has been linked to reduced hatchability, reduced offspring survival, male demasculinization, and female superfeminization in American alligators (*Alligator mississippiensis*) from Lake Apopka, Florida (Guillette *et al*. 1994).

• *Immunosuppression*

DDT has been shown to adversely affect the immune system in laboratory animals (Banerjee *et al*. 1986, Banerjee 1987). DDT causes increased susceptibility to parasitic and viral infections, decreased delayed-type hypersensitivity, and decreased antibody response in mice, rats, rabbits, and chickens (Wong *et al*. 1992).

• *Adrenal effects*

DDT is taken up and retained in the adrenocortical tissue of birds and laboratory rodents (Backström *et al*. 1965, Brandt *et al*. 1978). *o,p'*-DDD binds covalently in human adrenal cortex but not in mice. 3-$MeSO_2$-DDE is a lipophilic, persistent metabolite of DDT and has a high binding affinity for, and high toxicity in, the adrenal cortex of mice (Lund *et al*. 1988, Jönsson *et al*. 1991, 1992, Brandt *et al*. 1992). 3-$MeSO_2$-DDE is metabolized to a reactive and cytotoxic intermediate that binds covalently to the zona fasciculata causing necrosis and decreased plasma corticosterone levels.

$MeSO_2$-DDE has been found to be highly corticolytic although there are substantial species differences. *In vivo* and *in vitro* experiments show that *p,p'*- and *o,p'*-DDD are metabolically activated to intermediates that bind covalently to the adrenal cortex of mink (Jönsson *et al*. 1993) where they cause necrosis, but $MeSO_2$-DDE is not activated (Lund *et al*. 1988, Brandt *et al*. 1992). Jönsson *et al*. (1993) also showed that *o,p'*- and *p,p'*-DDD were metabolically activated and covalently bound by otter adrenal as well in an *in vitro* system. $MeSO_2$-DDE did not bind covalently in either species and the authors concluded that this metabolite does not seem to be an adrenal toxicant in mink or otter. Previously, it has been shown that DDD, particularly *o,p'*-DDD, is toxic to the adrenal cortex in humans and other species (Nelson and Woodard 1948, Bergenstal *et al*. 1960, Eriksson *et al*. 1987) and it has been used as a corticolytic agent in human and veterinarian medicine.

Birds treated with *p,p'*-DDE show increased adrenal weights compared to controls (reviewed in WHO 1989a). In vivo and *in vitro* experiments show that *o,p'*-DDD and 3-$MeSO_2$-DDE are metabolically activated to intermediates that bind covalently to the adrenal interrenal cells of chickens (Jönsson *et al*. 1994) where they cause cellular degeneration. Experiments also showed that *o,p'*-DDD and 3-$MeSO_2$-DDE are metabolically activated and bind covalently to adrenal tissue in eider and domestic duck embryos.

3-$MeSO_2$-DDE is found in Baltic grey seals and is suspected to be one possible cause of adrenal cortex hyperplasia seen in these animals (Jensen and Jansson 1976, Bergman and Olsson 1985). Lund (1994) has found that *o,p'*-DDD, and to a lesser extent even 3-$MeSO_2$-DDE, bind covalently to adrenal cortical tissue from grey seal. Two $MeSO_2$-DDE congeners have been identified in tissues from Canadian polar bears and ringed seal, beluga from the St. Lawrence estuary, and otter and wild mink from northern Sweden (Bergman *et al*. 1994b, Letcher *et al*. 1994, 1995, Letcher 1996).

• *Thyroid effects*

Birds treated with *p,p'*-DDE show increased thyroid weights compared to controls (reviewed in WHO 1989a).

6.5.3.2.4. **Hexachlorobenzene** (**HCB**)

HCB may form metabolites such as pentachlorophenol (PCP), tetrahydroquinone, pentachlorothiophenol, and lower chlorinated benzenes (Debets and Strik 1979, Renner 1988). Pentachlorophenol has been detected in green sunfish (*Lepomis cyanellus)* (Sanborn *et al*. 1977), rainbow trout (*Oncorhynchus mykiss*), rats, mice, and guinea pigs (Koss and Koransky 1978, Koss *et al*. 1978) after HCB treatment.

• *Reproductive and developmental effects*

HCB has been shown to affect male reproduction when given at high doses (30 000-221 000 ng/g body weight/d). The effects include reduced serum testosterone levels in mice (Elissalde and Clark 1979), histologic changes in testes causing retarded sexual maturation in pigs (den Tonkelaar *et al*. 1978), and reduced mating index in rats (Simon *et al*. 1979).

*In utero* exposure to HCB leads to increases in fetal malformations and cleft palate in mice (Courtney *et al*. 1976). Fetotoxicity has been seen in treated rats and mink (Khera 1974, Grant *et al*. 1977, Kitchin *et al*. 1982, Rush *et al*. 1983). Grant *et al*. (1977) also observed reduced fertility index in rats. Reduced litter sizes, increased number of stillbirths, and reduced offspring survival are seen in HCB-treated rats and cats (Mendoza *et al*. 1977, 1978, 1979, Hansen *et al*. 1979, Kitchin *et al*. 1982). The no-effect level for rats was 20 000 ng/g food (approximately 1000-1200 ng/g body weight/d) (Grant *et al*. 1977).

Rush *et al*. (1983) observed a significant decrease in kit survival for female mink fed 1000 and 5000 ng/g HCB diets for 47 weeks. Female ferrets (*Mustela putorius furo*) fed 125 000 ng/g HCB failed to reach estrus (Bleavins *et al*. 1984). Female mink fed 25 000 ng/g produced fewer kits and had a higher incidence of stillbirths. A diet of 1000 ng/g led to reduced kit birthweights in both mink and ferret. Offspring mortality was higher in treated mink and ferrets.

Japanese quail exposed to HCB had decreased fertility, egg production, and hatchability after 90-day exposure to 20 000 ng/g food (Vos *et al*. 1971). Herring gull embryos injected with HCB showed reduced embryonic weights at a dose of 1500 ng/g body weight and an $LD_{50}$ of 4300 ng/g body weight (Boersma *et al*. 1986).

Offspring of rats exposed to 2500 or 25 000 ng/g body weight/d showed increased hyperactivity (Goldey and Taylor 1992). The startle response to noise was reduced at age 23 days, but significantly increased at 90 days in the high dose group.

• *Cytochrome P450-dependent monooxygenases*

HCB induces cytochrome P450 1A (increased AHH activity) and 2B in laboratory animals (Delzell *et al*. 1994b). HCB treatment leads to increased cytochrome P450 and EROD activity in mink exposed *in utero* (Rush *et al*. 1983). HCB-treated hens had increased APND and cytochrome P450 (Kan *et al*. 1979). HCB also leads to increases in cytochrome P450 and some enzymes in Japanese quail (Carpenter *et al*. 1985a, 1985b). HCB does not induce cytochrome P450 1A or EROD in rainbow trout (Tyle *et al*. 1991).

• *Porphyria*

HCB is a potent inducer of porphyria in laboratory rodents (reviewed in Delzell *et al*. 1994b). In rats, females are more sensitive to HCB's porphyrogenic effects than males (Grant *et al*. 1975, Kuiper-Goodman *et al*. 1977, Rizzardini and Smith 1982, Smith *et al*. 1985). Chronic exposure to HCB (500 000 ng/g body weight/d for 5-10 days) leads to increased liver porphyrin levels in Japanese quail (Miranda *et al*. 1983, Carpenter *et al*. 1985a, 1985b, 1985c, Buhler and Carpenter 1986, Lambrecht *et al*. 1988).

Liver porphyrin concentrations were significantly correlated with concentrations of HCB, PCB, and DDE in herring gulls (*Larus argentatus*) from Saginaw Bay on Lake Huron (Fox *et al*. 1988).

• *Immunosuppression*

HCB has been shown to adversely affect the immune system in laboratory animals (Vos 1986, Barnett *et al*. 1987, Vos and Luster 1989, Van Loveren *et al*. 1990). Mice exposed to HCB were more susceptible to infection (Loose 1982), and had reduced resistance to tumor cells (Loose *et al*. 1981), depressed delayed-type hypersensitivity response to contact allergen (Barnett *et al*. 1987), and depressed humoral immunity, cell-mediated immunity and host resistance (Vos 1986, Carthew *et al*. 1990).

Mink and ferrets treated with HCB had depressed T-cell function and other immunosuppressive effects (Bleavins *et al*. 1983).

• *Thyroid effects*

Subchronic exposure to HCB affects the thyroid in various strains of rats, leading to decreased serum levels of total and free thyroxine (T4) and triiodothyronine (T3). This, in turn, has led to changes in the thyroid gland including increased thyroid weight, increases in circulating levels of thyroid-stimulating hormone and increased iodine uptake (Rozman *et al*. 1986, Kleiman de Pisarev *et al*. 1989, 1990, Van Raaij *et al*. 1991b, 1993a, 1993b, Foster *et al*. 1993, Den Besten *et al*. 1993, Sopena de Krakoff *et al*. 1994). These effects were seen in rats dietarily exposed to as little as 9500 ng/g body weight/d during a 13-week period (Den Besten *et al*. 1993).

Possible mechanisms include increased metabolism of thyroid hormones by HCB-induced enzymes, increased deiodination of thyroxine (Kleiman de Pisarev 1989, Van Raaij *et al*. 1993b), and competition for T4 binding sites on thyroid hormone transport proteins in plasma (Van Raaij *et al*. 1991a, 1993a). This may be due to the formation of pentachlorophenol as a metabolite of HCB. Pentachlorophenol has been shown to bind to the thyroid hormone transport protein, transthyretin, two times better than T4, for example (van den Berg 1990).

• *Cancer*

HCB has not been found to be mutagenic (Haworth *et al*. 1983, Górski *et al*. 1986, Siekel *et al*. 1991, Rumsby *et al*. 1992) and does not bind strongly to DNA (Stewart and Smith 1987). HCB does cause cancers of the liver (hepatomas, hepatocellular carcinomas), thyroid (alveolar adenomas), kidney (renal cell adenomas), parathyroid (adenomas), and adrenal glands (cortical adenomas, phaeochromocytomas) in mice, hamsters, and rats (reviewed in Delzell *et al*. 1994b). Results from several studies indicate that HCB is a tumor promoter (Shirai *et al*. 1978, Pereira *et al*. 1982, Herren-Freund and Pereira 1986, Smith *et al*. 1989, 1993a, Stewart *et al*. 1989).

6.5.3.2.5. **α-, β-, and γ-hexachlorocyclohexanes** (**HCH**s)

Technical HCH is a mixture of α-, β-, γ- and δ- HCH.

• *Reproductive effects*

No information on the effects of α-HCH on reproduction, embryotoxicity, or teratogenicity in laboratory animals is available.

β-HCH is estrogenic in rodents and leads to decreased testes weights, atrophied testes, and atrophied ovaries (Van Velsen *et al.* 1986). A two-generation study in rats found that litter size was reduced in the highest dose group and the offspring had reduced fertility and higher mortality (Van Velsen 1986).

β-HCH has been shown to be estrogenic in both male and female guppies (*Poecilia reticulata*), causing excess production of vitellogenin (Wester *et al.* 1985, Wester 1991). The effect was seen at water concentrations of 100 μg/L or higher. Medaka (*Oryzias latipes*) eggs and young fish were exposed to β-HCH for one and three months (Wester and Canton 1986, Wester 1991). Intersexuality and hermaphroditism were seen in males and production of vitellogenin was seen in both sexes, both evidence of estrogenic activity.

Lindane (γ–HCH) may have both estrogenic and antiestrogenic effects. Female rats treated with lindane showed disrupted estrous cycling, reduced pituitary and uterine weights, and delays in vaginal opening (Chadwick *et al.* 1988). Fertilization rate was reduced in female rats treated with 10 000 ng/g compared to rats fed only 5000 ng/g (Trifonova *et al.* 1970). Lindane given to mice during early pregnancy reduced implantation and when given at later stages of pregnancy, caused resorption of fetuses, high offspring mortality, and reduced offspring body weights (Sircar and Lahiri 1989). Other studies in several species of laboratory animals indicate no direct effects of lindane on reproduction in terms of litter sizes, offspring survival, or defects in offspring (WHO 1991).

Lindane fed to chickens and Japanese quail does not affect eggshell thickness, but does reduce egg production at high doses (100 000-200 000 ng/g food) (Whitehead *et al.* 1972a, 1972b, 1974). Ducks (*Anas platyrhynchus domesticus*) treated with lindane stopped laying eggs immediately after treatment, and had reduced clutch sizes when laying restarted (Chakravarty *et al.* 1986). No effects are seen on spawning, egg hatchability, or fry survival in fish treated with lindane (Macek *et al.* 1969).

• *Cytochrome P450-dependent monooxygenases*
α–HCH-treated laboratory animals have increased APND activity (Schulte-Hermann *et al.* 1974, Seifart and Buchar 1978, den Tonkelaar *et al.* 1981, Kuiper *et al.* 1985). Rats treated with β-HCH had increased APND activity and cytochrome P450 levels (Van Velsen 1986, Van Velsen *et al.* 1986). Lindane increases APND, ECOD, AE, EROD, and cytochrome P450 levels (Van Velsen *et al.* 1984, Videla *et al.* 1990).

• *Immunosuppression*
β-HCH has been shown to adversely affect the immune system in laboratory animals (Cornacoff *et al.* 1988). Dose-dependent reductions in immune response have been seen in rabbits treated with lindane (Desi 1976, Desi *et al.* 1978).

• *Adrenal effects*
Rats treated with β-HCH had increased adrenal weights (Van Velsen 1986).

• *Thyroid effects*
Medaka treated with β-HCH developed thyroid hypertrophy (Wester and Canton 1986).

• *Cancer*
α-HCH does not appear to be mutagenic (WHO 1992), but seems to be a promoter. High doses in rats and mice cause liver tumors. β-HCH does not appear to be mutagenic (WHO 1992). In some experiments with mice, increases in liver tumors have been seen. No increased tumor production has been seen in treated rats. β-HCH has been classified as a tumor promoter. Lindane is not mutagenic (WHO 1991). High doses cause liver tumors in mice and lindane is considered to be a tumor promoter.

#### 6.5.3.2.6. **Mirex**

Chronic exposure leads to body weight loss, various signs of liver toxicity, and induced monooxygenases. Reduced survival of fish has been seen in ponds treated with mirex (Bookhout and Costlow 1975).

• *Reproductive effects*
Chronic studies of mirex on mice and rats have shown decreased litter sizes, decreased or absent reproduction, and reduced viability of the young (reviewed in WHO 1984c). Mirex treatment in birds did not reduce egg production or embryo survival in chickens, mallard ducks, or bobwhite quail (*Colinus virginianus*) (Heath and Spann 1973, Davison and Cox 1974). No effects were seen on eggshell thickness. High doses (600 000 ng/g) caused a significant reduction in egg hatchability and chick survival in chickens (Naber and Ware 1965). Mallards fed mirex for three generations had reduced duckling survival in the third generation (Hyde *et al.* 1973).

• *Cytochrome P450-dependent monooxygenases*
Rats, rabbits, and mice treated with mirex show an increased microsomal cytochrome P450 content (Fabacher and Hodgson 1976, Iverson 1976, Villeneuve *et al.* 1977, 1979, Kaminsky *et al.* 1978, Warren *et al.* 1978, Chambers and Trevathan 1983). Mirex causes an induction pattern similar to phenobarbital (cytochrome P450 2B) (Madhukar and Matsumura 1979). No induction of APND or cytochrome P450 was seen in treated chickens or Japanese quail (Davison and Cox 1974, Davison *et al.* 1976).

• *Immunosuppression*
Mirex treatment leads to decreased thymus and spleen weights, reduced antibody response, and reduced antibody levels in chickens and mice (Wong *et al.* 1992).

• *Cancer*
Mirex does not appear to be mutagenic when tested in the Ames test (Hallett *et al.* 1978). It does induce cancer in rats and mice (IARC 1979).

#### 6.5.3.2.7. **Toxaphene (campheclor, polychlorinated camphenes, polychlorinated bornanes)**

Toxaphene is acutely toxic to fish, with lethal water concentrations in the range of 5-100 μg/L, and saltwater fish are more sensitive than freshwater fish (Saleh 1991). Laboratory studies have shown that exposure to toxaphene affects behavior and growth in fish (Mayer *et al.* 1975, 1977, Pollock and Kilgore 1978). Fish injected with environmentally relevant doses of technical toxaphene and released into the wild had reduced long-term survival (Delorme *et al.* 1993).

• *Reproductive effects*
Oral dosing in pregnant mice and rats resulted in maternal toxicity and fetotoxicity (Chernoff and Carver 1976) with a LOAEL for these effects of less than 15 000 ng/g body weight/d. No reproductive effects are seen in multigenerational studies using laboratory rodents or chickens (WHO

1984d, Chu *et al*. 1988). No effects are seen on egg hatchability or eggshell thickness. Brook trout (*Salvelinus fontinalis*) treated with toxaphene have decreased ova viability (Mayer *et al*. 1977). Brook trout fry show high mortality after exposure to toxaphene (Mayer *et al*. 1975). Toxaphene has been shown to be estrogenic in the *in vitro* E-screen test (Soto *et al*. 1994).

• *Cytochrome P450-dependent enzymes*
Rats fed more than 2400 ng/g body weight/d for 1-6 months had increased liver enzyme induction (Peakall 1976b).

• *Skeletal effects*
Bones of fish are altered after exposure to toxaphene (Mayer *et al*. 1977, 1978, Hamilton *et al*. 1981). There are increased calcium levels, but decreased levels of collagen and hydroxyproline, leading to brittleness in the backbone. Reduced hydroxyproline is also seen. Channel catfish fry (*Ictalurus punctatus*) with body concentrations of 3400 ng/g ww or higher had decreased growth, and body concentrations of 600 ng/g ww adversely affected bone development (Stickel and Hickey 1977).

• *Immunosuppression*
Offspring of mice fed toxaphene during mating, pregnancy, and lactation showed immunosuppression including depressed antibody formation (Allen *et al*. 1983).

• *Adrenal effects*
Toxaphene is taken up and retained in the adrenocortical tissue of rats (Mohammed *et al*. 1985). Adrenal hypertrophy was seen in bobwhite quail fed toxaphene (Hurst *et al*. 1974).

• *Thyroid effects*
Increased thyroid growth was seen in bobwhite quail fed toxaphene (Hurst *et al*. 1974). Thyroid effects were seen in rats and dogs fed different doses of toxaphene for 13 weeks. The no-adverse-effect-levels (NOAELs) found in this study for thyroid effects were 4000 ng/g feed or 350 ng/g body weight/d for rats and 200 ng/g body weight/d for dogs (Chu *et al*. 1986).

• *Cancer*
Toxaphene is mutagenic in the Ames test (Hooper *et al*. 1979, Mortelmans *et al*. 1986) and a potent carcinogen in rats and mice (reviewed in Saleh 1991).

#### 6.5.3.3. **Less persistent organochlorine pesticides**
#### 6.5.3.3.1. **Endosulfan**

• *Reproductive effects*
Male rats treated with endosulfan had reduced testes weights (FAO/WHO 1968, WHO 1984b), lowered plasma gonadotrophin and testosterone levels, as well as lowered testicular testosterone levels (Singh and Pandey 1990). Endosulfan has been shown to be estrogenic in the *in vitro* E-screen test (Soto *et al*. 1994). Endosulfan causes fetal resorption and skeletal and internal abnormalities in fetuses of treated pregnant rats at levels that also cause maternal toxicity (Naqvi and Vaishnavi 1993).

Female vitellogenic catfish (*Clarias batrachus*) treated with endosulfan had reduced plasma vitellogenin levels (Chakravorty *et al*. 1992).

• *Cytochrome P450-dependent monooxygenases*
Rats treated with endosulfan had increased MFO activity (den Tonkelaar and van Esch 1974) and increased APND activity (Naqvi and Vaishnavi 1993). Endosulfan is an effective inducer of cytochrome P450 1A activity in fish (Goksøyr 1995b).

• *Immune effects*
Endosulfan treatment leads to depressed humoral and cellular immune responses at subtoxic doses (Naqvi and Vaishnavi 1993).

• *Cancer*
Endosulfan does not appear to be genotoxic. Data are inadequate about the possible carcinogenicity of endosulfan (Naqvi and Vaishnavi 1993).

#### 6.5.3.4. **Other pesticides**
#### 6.5.3.4.1. **Tributyltin (TBT)**

• *Reproductive effects*
TBT is arguably the most toxic substance deliberately introduced to natural waters (Goldberg 1986). Chronic effects are observed at exposure levels of 1 μg/L or less for oysters, mussels and crustaceans (Rexrode 1987), while the most sensitive species (dogwhelk snails, e.g., *Nucella*) show sublethal effects at concentrations of only a few ng/L or less (Bryan and Gibbs 1991, Gibbs 1993, Stewart and Thompson 1994). At these ultra-low concentrations, which challenge present analytical detection limits for TBT in seawater, dogwhelk snails exhibit imposex (development of male characteristics leading to sterilization of females). Although imposex has been observed most frequently along shores adjacent to obvious sources such as marinas or harbors and has been associated with both pleasure boats and commercial shipping, the problem has been defined as long-term and global rather than short-term and regional (Ellis and Pattisina 1990, Bryan and Gibbs 1991, Ten Hallers-Tjabbes *et al*. 1994).

TBT is moderately lipophilic and may therefore bioaccumulate in the marine environment (bioaccumulation factors are of the order of $10^3$-$10^4$). However, it is also metabolized by vertebrates and invertebrates, reducing the body burden of TBT and its level of bioaccumulation. While TBT is clearly causing harm to components of the marine ecosystem, studies of its hazard to humans suggest that it is not neurotoxic, nor is it considered mutagenic, teratogenic, or carcinogenic. In view of its effects on sensitive marine species, particularly the mollusks, TBT falls into the category of 'endocrine disrupters' (Colborn *et al*. 1993). The main argument supporting its continued use is that the effective antifouling properties reduce fuel consumption. It has been estimated by the European Chemical Industry Council (unpubl. manuscript) that world fleet consumption of oil is reduced by $7 \times 10^6$ tonnes/y with consequent reduction in $CO_2$ ($22 \times 10^6$ tonnes/y) and sulfur ($0.6 \times 10^6$ tonnes/y) emissions to the atmosphere.

• *Immunosuppression*
TBT induces thymic atrophy (Wong *et al*. 1992).

## 6.6. Regional and circumpolar levels and trends in abiotic and biotic media

The state of knowledge of contaminants in Arctic abiotic and biotic media has advanced considerably since the publication of the first major reviews such as those by Andersson *et al*. (1988), Thomas *et al*. (1992), Lockhart *et al*. (1992), and Muir *et al*. (1992b). The most significant gains are new knowledge of spatial trends of organochlorines (OCs) in air,

snow, and sediments, as well as in terrestrial and marine biota where no information was previously available. There are also some new data on temporal trends in biota (section 6.7).

Most data discussed in this chapter are from recent publications, from unpublished data and short summaries from the scientists in charge of projects on Arctic contaminants, or from AMAP-associated databases. Summary data are usually presented in the same form selected by the scientist responsible for the data, and in some cases, data are summarized further. Results from older studies are often mentioned, however, raw data were not available in most cases.

Most interpretation in this chapter, including comparisons and conclusions about spatial and temporal trends, were made by scientists in charge of projects (most listed as co-authors), contributors, and by the three editors. In some cases interpretation was not possible due to limited sample numbers. These data were primarily entered as summary data in the Annex Tables, and interpretations were kept to a minimum. It should also be noted that sample sizes for means given in these tables vary greatly. Readers should be aware of the risk of spurious conclusions drawn from small data sets.

It should be noted that results from different studies are not always comparable for a number of reasons. One reason is that the same compounds were not measured. For example, PCB (sum of PCBs) could refer to the sum of 7-100 PCB congeners. For this reason, results are usually subscripted by explanations of the compounds presented. It is suggested that serious comparisons be made only in references to the raw, not summarized, data, obtained from the scientists or agencies in charge of the data.

Another reason for limited comparisons is that the concentrations of OCs in biotic and abiotic samples are expressed in several ways in this report. Due to lack of supporting information (e.g., percent lipid), it was not always possible to convert a value in one basis to its corresponding value in another basis. In biological samples, concentrations can be expressed on a wet weight (ww) basis, giving the concentration of OC in the total weight of tissue analyzed; on a lipid weight (lw) basis, giving the concentration in the lipids in that particular tissue; or as a body burden, giving the contaminant concentration in the whole organism. OC levels in abiotic samples are often expressed on a dry weight (dw) basis, and some biotic samples, most often plants, were also reported on a dry weight basis. All lipid weight concentrations are calculated from the wet weight concentrations unless otherwise indicated.

In the case of abiotic samples, results for sediments are usually expressed on a dry weight basis because organic carbon, an important covariate, is not always available for all samples. For water samples, a distinction needs to be made between whole water (containing dissolved and particulate organic carbon) and filtered or centrifuged samples, because very hydrophobic compounds could be primarily associated with particles. Concentrations of OCs in air samples, expressed as $pg/m^3$, are presented for gas phase and particle phase separately or are summed to give a total air concentration. For most semi-volatile OCs, total air concentrations are similar to gas phase concentrations, except at very low temperatures (e.g., – 30 to – 40°C which occur in some regions during January-February).

When comparing the OC concentrations in different tissues and organs, in different species, or when describing spatial and time trends or biomagnification, the comparisons are best made on a lipid weight basis (section 6.3.1.1). However, this has not always been done for the following reasons: 1) When discussing intake of OCs from food or prey, for example, from reindeer muscle or liver, whole eggs, or small mammalian or avian prey, OC levels on a wet weight or whole weight basis, whichever represents the consumed food, may be the most informative statistics since they represent the total mass of OCs consumed. 2) OC levels on a wet weight basis may be an adequate measure of the OC level on a lipid weight basis. For example, in marine mammals, OC level in blubber is the accepted measure, since blubber is mostly lipid. 3) Whether lipid-normalized OC concentrations are the only valuable measure is still open to some debate. Lipid levels in some tissues fluctuate greatly, for example, lipid levels in liver in relation to feeding and starvation cycles. Two similarly contaminated animals could appear to have very different lipid weight levels of OCs in liver tissue simply because they were measured at different times of the year, yet each animal may experience the same stresses due to contaminant load over an extended period of time. Modeling approaches have been used to describe and predict the movement of OCs between tissues and the risk at various stages (Kingsley and Hickie 1994), however, such approaches are usually not possible. As an intermediate approach, Hebert and Keenleyside (1995) recommend calculation of lipid-adjusted means using analysis of covariance rather than normalizing individual values. 4) The measure of percent lipid can be unreliable due to variation with extraction methods and due to weighing errors. 5) Lipid content was not determined. Even today, lipid amount is not always determined when doing chemical analyses of OCs, therefore, some results can only be given here on a wet weight basis, as is evident in values in most of the Annex Tables in this chapter. This is a major shortcoming in many of the available data presented.

Nevertheless, wet weight comparisons can be made with the same tissues with individuals of the same species in similar condition. Since OCs occur in lipids, tissues with high fat content will have higher OC wet weight concentrations than lean tissues. For example, blubber will have higher wet weight concentrations than leaner muscle from the same animal. Organisms with a large amount of lipid in a particular tissue, for example, fish with fatty livers (e.g., cod) or with fatty muscle (e.g., salmonids) will often have higher wet weight concentrations of OCs than leaner organisms.

An organism's size, sex, age, fitness, reproductive phase, nutritional status, health status, individual food preferences or feeding habits, and many environmental factors such as seasonal or climatic characteristics can also influence OC levels in biota (sections 6.3 and 6.4). In statistical analyses, these factors are called 'covariates' because they vary along with the dependent variables of interest, OC levels in this case. Methods of working with covariates vary, and opinions about valid methods differ among scientists. Linear and non-linear models which were relatively difficult to work with as few as fifteen years ago, for example, mixed models which combine analysis of variance, covariance, and regression models, are now available as computer software and relatively easy to apply and interpret, with appropriate advice from statisticians who are aware of the pitfalls.

The correlation between OC levels and one or more of these factors is often the main objective of a study, for example the relationships between OC levels and organisms' size and reproductive status. In most cases, a covariate is a factor that accounts for much of the unexplained variation in the data, but is not one of the main factors of interest in the study. An example of this is the use of stable isotopes of nitrogen to quantify an organism's food preferences and trophic status, and this measurement is then related to OC levels. In other studies, particularly in wide-ranging surveys, the effects of some factors are 'adjusted for' by the use of appropriate linear modeling methods before comparisons

are made (e.g., the polar bear study, section 6.6.4.8). Linear coefficients calculated to describe the effects of these covariates are of interest in themselves. In such cases, the unexplained error is high, but results have wide applicability. In other studies, efforts have been made to keep the values of some of these covariates as consistent as possible in order to determine the effects of other factors of interest. Specifically, in studies in Sweden, efforts were made to collect fish annually of similar age, sex, stage of gonad development, and at the same stations at the same time of the year in order to determine subtle temporal trends (section 6.7.3.2).

If important covariates are ignored, spurious temporal and spatial trends may appear to exist. More seriously, if important covariates are not considered, ecologically important temporal trends may not be noticed, and the unexplained variability in the data can be extremely high. For example, Bignert *et al*. (1993, 1994) demonstrated that temporal trends could not be distinguished if they pooled tissue samples from Baltic guillemots or from herring. These studies showed that interpretations of temporal and spatial trends based on a single year's sampling or from scattered sampling over time should be made with caution.

Studies of spatial trends require a standardized sampling design to minimize the effect of covariates on concentrations. Sampling should also preferably be done during the same time period so as to minimize the influence of temporal trends. In only a few cases were samples taken in such a way that spatial trends over major parts of the Arctic were possible to study with some confidence (polar bears, caribou). In other cases, results are available for the same species from various parts of the Arctic from independent studies. Sampling was done in different areas for different reasons, sometimes in different years, and because of this, this type of spatial comparison is more tenuous and conclusions about spatial trends must be made with caution.

Environmental measurements, such as air and water temperature, wind speed, basic water chemistry (pH, suspended solids, dissolved organic carbon), particle size, extent of ice cover, snow depth, and water flow rate, are just some of the possible ancillary data needed for interpreting OC data in abiotic samples. The development of a mass balance budget for OCs in the Arctic Ocean (section 6.6.4.3) illustrates the need for detailed environmental measurements (e.g., flow rates of north-flowing rivers) along with sound physical/chemical property information on the chemicals of interest. There are large uncertainties in physical properties, for example, in H and $VP_L$ for OCs at Arctic temperatures.

All data reported in this chapter are relatively recent, and most results satisfy basic standards of quality assurance set by AMAP. Virtually all of these recent data have been generated by laboratories which have participated in national and international interlaboratory comparisons on OCs in biological tissues (e.g., cod liver oil) or in standard solutions. Thus, it is believed that uncertainties associated with analytical measurements reported in this chapter are relatively low, and attempts have been made to identify results that appear suspect. The major uncertainty in the OC data sets probably arises from contamination during sample collection and storage prior to analysis. This is especially the case with abiotic samples such as air, suspended solids, and water. Several contributors of abiotic data have stressed that contamination from lubricating oils, possible insecticide use on ships, and evaporation of OCs from construction materials may have contributed to unusually elevated levels of PCBs and DDT. Air drying of samples in the laboratory has been noted as another source of contamination of plants, sediments, and soils. These issues are briefly addressed where relevant to the interpretation of individual data sets. However, the reader should be aware that inadvertent contamination is a major problem, especially in dealing with very low levels of POPs, which is generally not addressed through interlaboratory-type studies.

## 6.6.1. Air and precipitation

### 6.6.1.1. Air

Measurements of persistent organic pollutants, including herbicides, pesticides, synthetic industrial compounds, and polycyclic aromatic hydrocarbons (PAHs), have been made on a weekly basis in the Canadian, Norwegian, and Russian Arctic since 1992 (Barrie *et al*. 1997, Oehme *et al*. 1995b, 1995d, Rovinsky *et al*. 1995). Limited numbers of air samples have also been collected in the past six years in the Bering/Chukchi Seas (Iwata *et al*. 1993, Jantunen and Bidleman 1995, Chernyak *et al*. 1996) and during the USA/Canada transpolar cruise (Barrie *et al*. 1997). Air samples have also been collected in Iceland on Heimaey Island (63°24'N, 20°17'W) from January to December 1995 (Icelandic Meteorological Office 1996 unpubl. results). Results for PAHs in air are discussed in chapter 10.

The Canadian program involved high-volume air samplers placed at Alert, NWT (82°30'N, 62°20'W); Tagish, Yukon (60°20'N, 134°15'W); Cape Dorset, Baffin Island (64°20'N, 76°00'W), and, in collaboration with Russian scientists, at the mouth of the Lena River on Dunai Island (74°00'N, 125°00'E) in Russia. Samples were collected every six days ($11\text{-}13 \times 10^3$ m$^3$) using glassfiber filters and polyurethane foam. A total of 18 PAHs, 29 OCs, and 90 PCB congeners were determined in samples taken at Alert, Tagish, and Dunai. Results for Cape Dorset are not yet available. The Norwegian program involved collection of 1000-1100 m$^3$ air over a two- or three-day period once a week at Ny-Ålesund, Svalbard (78°55'N, 11°56'E) during 1993 to 1995. Air samples were also collected at Ny-Ålesund and Svanvik (N. Norway) from March to May 1992 (Oehme *et al*. 1995b). Samples were analyzed for ten PCB congeners plus HCH, HCB, and chlordane-related compounds. Results for the four Canadian locations are available for 1993 and 1994. Additional samples from 1995 from all sites have been collected and analyzed but final results are not yet available. The Icelandic program, on Heimaey Island, also used a high-volume air sampler with collection on polyurethane foam over a period of approximately 14 days. This site is located 4.5 km from a small town of 4500 inhabitants and is 118 m above sea level.

One of the predominant OCs in Arctic air is HCH (Oehme *et al*. 1995b, 1995d, Barrie *et al*. 1997). At Ny-Ålesund, ΣHCH concentrations during 1993 ranged from 10 to 240 pg/m$^3$ and from 21-148 pg/m$^3$ in 1994 (Table 6·6, Figure 6·4; next page). During 1993, a similar range of concentrations was found at Tagish (50-200 pg/m$^3$), while lower concentrations were generally observed at Alert (15-145 pg/m$^3$) and Dunai Island (1-100 pg/m$^3$). At Heimaey Island (Iceland), ΣHCH concentrations (in 1995) were lower than at other locations (8-68 pg/m$^3$). At all locations, α-HCH was the major individual OC present, with concentrations that were two to more than ten times higher than other OCs. Lindane (γ-HCH), ΣPCBs, and toxaphene are also prominent in Arctic air samples.

A summer minimum concentration of HCH isomers was observed at Alert and at Heimaey Island, but it was less distinct at the other three sites (Figure 6·4). At Alert, this minimum in HCH concentration corresponds to a summer maximum in precipitation. Alert sees air that has spent much time over the ice-covered Arctic Ocean, in an environment that has a strong summer maximum in precipitation. Seasonal

Table 6·6 Mean and range of concentrations (pg/m$^3$) of major OCs in Arctic air (1992-1994).

| Location | Svanvik, Norway | Ny-Ålesund Norway | Ny-Ålesund Norway | Ny-Ålesund Norway | Alert Canada | Alert Canada | Tagish Canada | Dunai Island Russia | Heimaey Island Iceland | Bering-Chukchi Seas | Resolute Canada |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Mar-May 1992 | Mar-May 1992 | 1993 | 1994 | Mar-May 1992 | 1993 | 1993 | 1993 | 1995 | 1993 | May-Sept. 1992 |
| α-HCH | 97 (39-205) | 144 (68-338) | 78 (6.8-203) | 61 (16-112) | 57 (1.1-116) | 61 (13-116) | 79 (24-160) | 40 (0.63-77) | 19 (7.8-46) | 91 (60-114) | 114 |
| γ-HCH | 46 (11-194) | 32 (13-99) | 14 (3.3-38) | 16 (5.3-62) | 11 (0.03-37) | 10 (1.9-29) | 11 (3.3-25) | 9.8 (<0.05-23) | 15 (<0.1-50) | 23 (12-37) | 9.8 |
| HCB | 108 (55-188) | 205 (121-373) | 92 (23-312) | 115 (76-541) | 56 (1.8-109) | 58 (12-108) | 42 (10-14) | 55 (19-173) | 8.0 (3.1-15.7) | –[a] | – |
| Dieldrin | – | – | – | – | 1.1 (0.12-2.8) | 1.2 (0.06-3.2) | 0.91 (<0.05-2.2) | 1.1 (<0.05-2.0) | <0.1 | – | – |
| Endosulfan | – | – | – | – | 4.2 (0.05-11) | 3.6 (<0.05-9.4) | 5.3 (0.08-15) | 3.0 (0.05-7.2) | – | – | – |
| Toxaphene[b] | – | – | – | – | 19 (3-73) | 11 (<3-43) | 14 (3-41) | 14 (3-32) | – | 4.9 | 7 |
| ΣCHL[c] | 3 (1.6-5.4) | 4 (1.7-15) | 2.6 (1.2-4.9) | 3 (1.5-7.3) | 4.8 (0.50-18) | 2.5 (0.34-10) | 2.9 (1.1-6.6) | 2.3 (0.11-6.0) | 3.3 (1.3-9.8) | – | 3 |
| ΣDDT | – | – | 1.2 (0.47-2.6) | 5.4 (1.5-15) | 4.3 (0.32-16) | 0.82 (0.12-2.2) | 1.4 (0.26-3.2) | 0.93 (0.24-2.0) | –[d] (<1.0-7.1) | – | – |
| ΣPCB (10 congen.) | 98[e] (2.8-33) | 13 (7.0-25) | 13 (2.9-121) | 57[f] (7-990) | – | 5.8 (0.18-25.9) | 3.7 (0.92-8.3) | 8.1 (6.5-23) | 17 (<1.0-45) | – | – |
| ΣPCB (total) | – | – | – | – | – | 32 (3.1-102) | 22 (14-41) | 34 (5.4-95) | – | – | – |
| Reference | Oehme *et al.* 1995d | Oehme *et al.* 1995d | Oehme *et al.* 1995b | Hagen unpubl. data | Fellin *et al.* 1996 | Barrie *et al.* unpubl. data | Barrie *et al.* unpubl. data | Barrie *et al.* unpubl. data | Thorlacius 1996 | Jantunen and Bidleman 1995 | Bidleman *et al.* 1995b |

a. not determined or results not available; b. Toxaphene determined by GC-ECD with hi-resolution NIMS confirmation. GC-ECD results multiplied by 2.7 to yield results equivalent to GC-NIMS based on analysis of 15 samples from Tagish by both methods; c. Sum of *cis*- and *trans*-chlordane + *cis*- and *trans*-nonachlor; d. Mean not calculated; 14 of 22 samples were < detection limit of ≈1 pg/m$^3$; e. Sum of seven congeners (CB 28, 31 and 52 not determined); f. Elevated PCB results in 1994 at Ny-Ålesund may be due to local contamination.

variations of HCH observed at Tagish (Yukon) do not show the summer dip in concentrations. This is likely due to the strong influence of flow off the North Pacific Ocean over the Rocky Mountains into this site. The seasonal variation of α- and γ-HCH at Dunai (Russia) peaked in the spring and decreased in summer similar to that seen at Alert. However, the spring decrease occurred in early May while at Alert it was in June. The Dunai site, at 74°N, is farther south than Alert (82.5°N) and, like Alert, it sees Arctic air much of the winter, however, spring melt comes earlier than at Alert.

The occurrence of an early HCH peak may reflect volatilization of cold-condensed HCHs from the snow pack during spring warm-up. It also may be due to use of technical HCH and lindane during spring months in temperate regions. The γ-/α-HCH ratios at Alert increased from 0.1 to 0.45 between March and June 1992 indicating fresh sources of lindane (Fellin *et al*. 1996). At Ny-Ålesund, little seasonal variation in ΣHCH was observed, but concentrations were influenced by long-range transport episodes where concentrations increased by 50-100% from one sampling time to the next (Oehme *et al*. 1995d). Highest γ-/α-HCH ratios at Ny-Ålesund were observed episodically in May and July.

At the Icelandic site, a springtime maximum of HCH was observed in 1995 followed by low concentrations during July and August (Figure 6·4). HCH profiles in Icelandic air (Figure 6·4) during 1995 differed from the other sites by having much higher γ-/α-HCH ratios (average 0.9, range 0.3-2.78), indicating greater proportions of the insecticide lindane, which was widely used in Icelandic sheep-farming until 1990, but only sparsely, if at all, after that. As its half-life is a maximum of three years, these practices cannot explain the high ratio and a source outside of Iceland is suspected (Icelandic Meteorological Office, pers. comm. 1996). Schreitmüller and Ballschmiter (1995) found a wide range of HCH ratios in the North Atlantic (average 0.86, range 0.2 to 3.6) and attributed high ratios to air masses from Western Europe where lindane is still used.

The seasonal variation of PCBs at five Arctic locations is shown in Figure 6·5. PCB results for Ny-Ålesund and Heimaey Island consisted of ten congeners (CB 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180), whereas 90 congeners were determined (about 50 were routinely detected) at Alert, Tagish, and Dunai Island. The ΣPCB results shown in Figure 6·5 are all based on the sum of the above ten congeners in the gas and particle phase, in order to allow comparison among sites. These ten congeners represent 6-25% of ΣPCB in air at Alert (Table 6·6).

Highest PCB concentrations for all four sampling sites in 1993 were found at Ny-Ålesund (Table 6·6). PCB concentrations (based on ten congeners) at Ny-Ålesund were two times higher than at Alert and 1.5 times higher than at Dunai during 1993. The PCB profile at Ny-Ålesund was dominated by long-range transport events. Increases in concentrations of more volatile PCBs (e.g., trichlorobiphenyl, CB 28/31; and tetrachlorobiphenyl, CB 52) were associated with long-range transport back trajectories from western Europe and central Russia, while lowest concentrations were found in air passing over the Arctic Ocean. At Alert, the PCB homologue pattern varied seasonally. The di-, tri-, and tetra-chlorinated PCBs peaked in the coldest months while the penta-, hexa-, and hepta- PCBs peaked later in the summer. Results for PCBs at Ny-Ålesund in 1994 may be compromised by unidentified contamination that resulted in some elevated PCB concentrations (J.-E. Haugen pers. comm. 1996). Mean ΣPCB concentrations are higher in 1994 than in 1992-1993, but lowest concentrations are similar each year. Similar contamination problems, possibly associated with contaminated oils in vacuum pumps used for air sampling, were encountered at Alert in 1992 (L. Barrie pers. comm. 1996).

The PCB profile at Alert in 1993 shows a peak in July corresponding to higher air temperatures. Most of the mass is in the gas phase (80-90%), but the particulate fraction is still important because it is more readily scavenged from the atmosphere than the gaseous fraction. PCB profiles at Dunai Island and at Tagish did not show as much seasonal variation as Alert or Ny-Ålesund. Some of the highest concentrations at Tagish

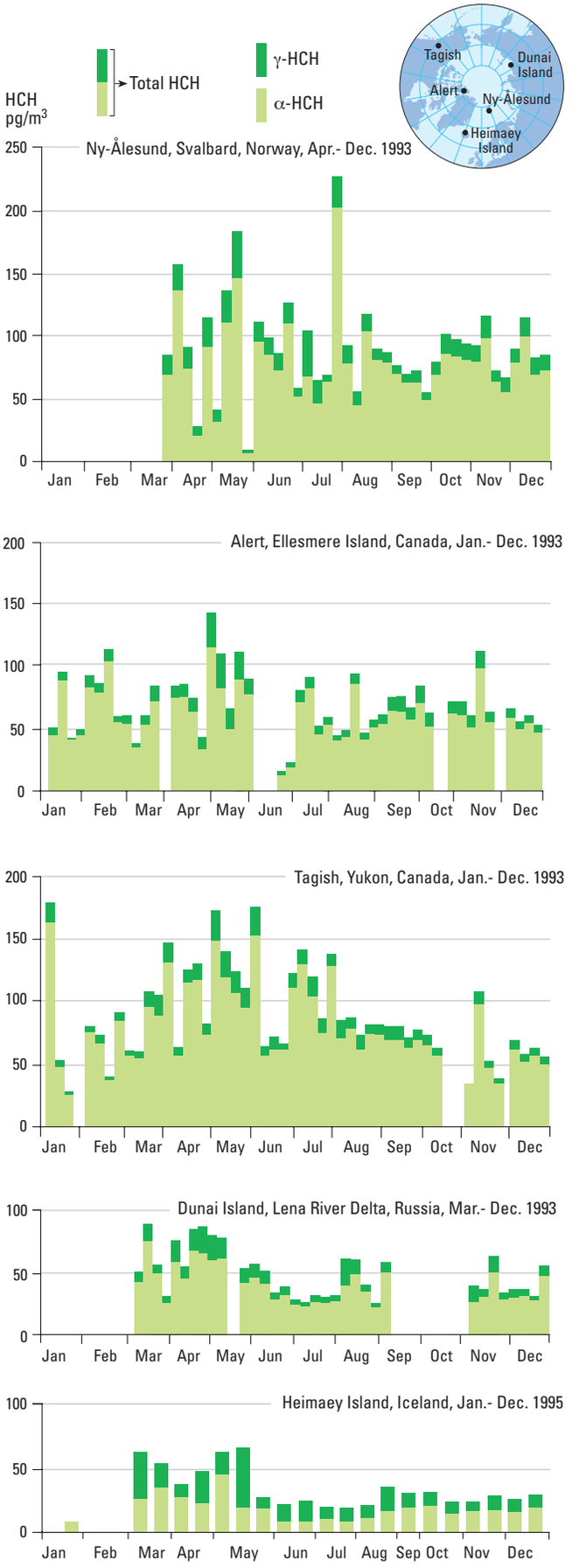



Figure 6·4. Concentrations of ΣHCH in Arctic air.

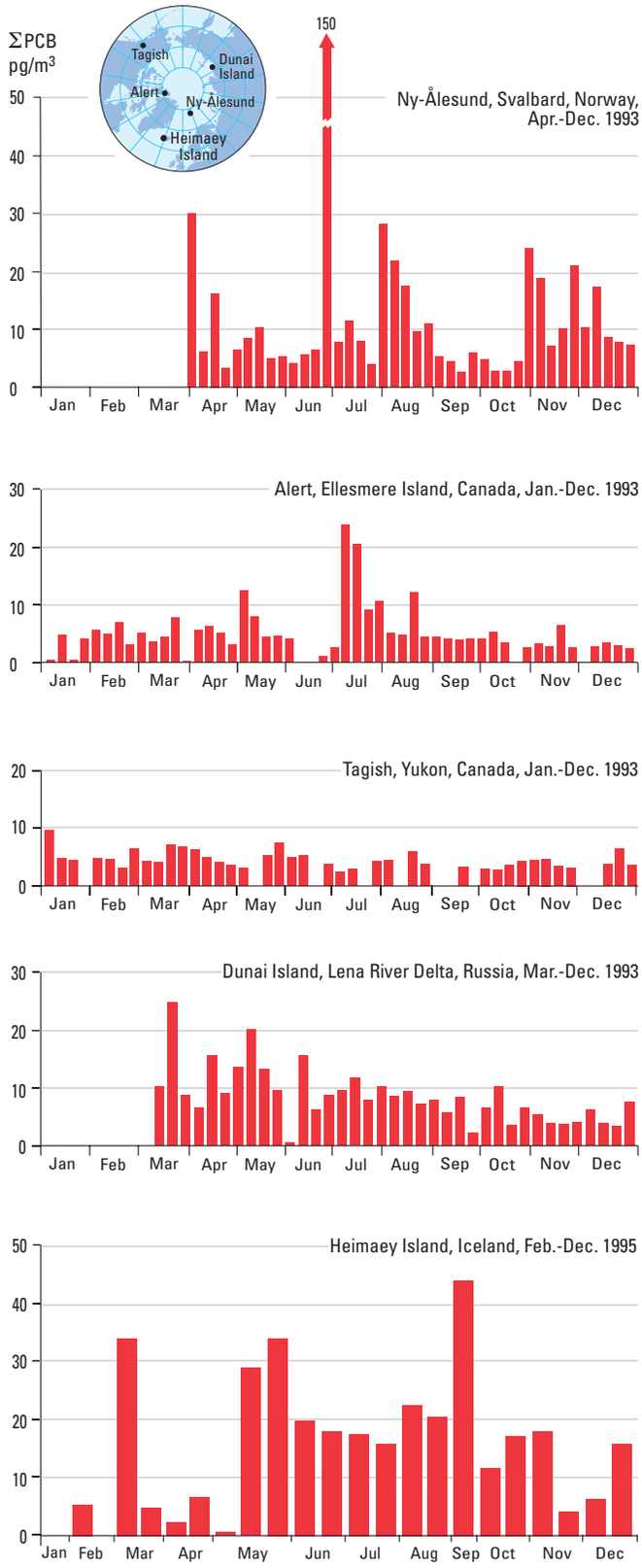



Figure 6·5. Concentrations of ΣPCB in Arctic air (sum of CBs 28, 31, 52, 101, 105, 118, 138, 153, 156, 180).

and Dunai were found during the winter months. Maximum ΣPCB levels at Tagish were 2.5 times lower than at Alert.

HCB is found at the highest concentrations of any individual OC in Arctic air with mean levels ranging from 8 pg/m$^3$ at Heimaey Island to 205 pg/m$^3$ at Ny-Ålesund. HCB results for the two Canadian and one Russian location are about two-fold lower than in the Norwegian Arctic (Table 6·6). The regional variation of HCB concentrations, particularly the low levels at the North Atlantic (Heimaey Island) site, is higher than for HCH, chlordanes, or DDT-related compounds, and warrants further investigation.

Chlordane-related compounds were also relatively prominent in air samples collected at all five Arctic sites, especially


Figure 6·6. Source regions for HCH, chlordane, toxaphene, and PCBs in Arctic air based on 5-day back-trajectories for elevated air concentrations at Tagish, Alert, and Ny-Ålesund (Oehme *et al.* 1996; Barrie *et al.* unpubl. data).

those in Canada. ΣCHL (sum of *cis*- and *trans*-chlordane, and *cis*- and *trans*-nonachlor) at Ny-Ålesund averaged 2.6 pg/m$^3$ in 1993, and 3.0 pg/m$^3$ in 1994, similar to levels at Tagish and Alert (Table 6·6). Elevated ΣCHL concentrations at Svalbard were found to be mainly of North American origin (Oehme *et al.* 1995b, 1995d). The major pathways identified by back trajectory analysis for Tagish, Alert, and Ny-Ålesund are shown in Figure 6·6. Episodic increases at Ny-Ålesund, especially of *trans*-chlordane, were associated with air mass back trajectories that passed over North America. At Alert, *trans*-chlordane levels were also elevated in June (Fellin *et al.* 1996) consistent with higher temperatures in source regions of North America. Patton *et al.* (1991) found that *trans*-/*cis*-chlordane ratios were useful for inferring air mass history because of the gradual conversion of *trans*- to *cis*-isomer during transport in the atmosphere. At Tagish, elevated concentrations of *cis*- and *trans*-chlordane, oxychlordane, heptachlor epoxide, along with dieldrin, α-HCH, γ-HCH, and *p,p'*-DDE were observed during the sample week of the 7-14th January 1993. Air mass back trajectories over this period revealed the movement of air from as far away as India, where DDT and HCH are still used for mosquito control in both agricultural and urban areas. Indeed, higher ratios of *p,p'*-DDE/(*p,p'*-DDT + *p,p'*-DDE) than those predicted by temperature for this period are probably due to the conversion of applied *p,p'*-DDT to *p,p'*-DDE. This photo-degradation process occurs post application in agricultural areas and/or during long-range transport.

Toxaphene is also an important OC contaminant in Arctic air. Toxaphene concentrations are usually lower than HCH, HCB, or PCBs but greater than ΣCHL or ΣDDT. Year-round monitoring of toxaphene has been carried out at Alert, Tagish, and Dunai from 1992-94, with analysis by GC-ECD and confirmation by GC-NIMS. Toxaphene has also been determined (by GC-NIMS) from summer 1992-1993 air sampling at Resolute Bay (Bidleman 1996) and on the BERPAC-93 cruise in the Bering and Chukchi Seas (Bidleman *et al.* 1995b). A comparison of 12 air sample extracts from Tagish by both GC-ECD and GC-NIMS found that the NIMS method yielded average values that were 2.7 times higher than ECD results (D. Muir unpubl. data). In contrast, toxaphene measured at the Ice Island by GC-NIMS was about a factor of 2 lower than ECD values (Patton *et al.* 1989). Results for toxaphene in Table 6·6 for Alert, Tagish, and Dunai are based on measured GC-ECD values adjusted assuming an NIMS/ECD factor of 2.7 and are regarded as best estimates of toxaphene at these sites. The discrepancy between the Resolute-BERPAC results, which were determined by GC-NIMS, and the adjusted Alert-Tagish values is a factor of 2-3.5. The lack of NIMS data for all of the sites limits our ability to accurately predict current atmospheric loadings of toxaphene to the Arctic Ocean and is a matter that needs to be resolved. Toxaphene concentrations at Alert were found to be much higher during May-September than during the cold period from October to April (1992-1993) (Fellin *et al.* 1996). The toxaphene profile in Arctic air consists mainly of hepta- and octachlorobornanes and differs markedly from the chlorobornane pattern in technical toxaphene (Bidleman *et al.* 1993).

Back trajectories for samples at Tagish in 1993 showed that air masses originating over North America were associated with high toxaphene concentrations (Figure 6·6). Lowest toxaphene concentrations at Tagish were found in air masses passing over the North Pacific Ocean. Nearly 80% of the toxaphene applications in the USA were in the southern states (Voldner and Schroeder 1989), and it is likely that volatilization of residues from agricultural soils in this region are contributing to the background concentrations in North America, including the Arctic. Despite a decade-old ban on toxaphene use in the USA, relatively high concentrations of toxaphene still occur in the southern USA. The mean concentration of toxaphene in Columbia, South Carolina, USA, during August 1994-January 1995 was 180 pg/m$^3$ (Bidleman unpubl. data). Based on limited information, Siberia does not appear to be a significant source of toxaphene. Average concentrations of toxaphene measured by GC-NIMS at Lake Baikal in June 1991 were 16 pg/m$^3$ (McConnell *et al.* 1996), similar to average concentrations at the three Canadian Arctic sites (Table 6·6).

### 6.6.1.2. **Precipitation**

Snow is an important component of annual precipitation in the Arctic and is highly efficient at scavenging organic contaminants from the atmosphere (see chapter 3). As a result, much of the emphasis on monitoring of persistent OCs in precipitation in the Arctic has been on snow chemistry. In Canada, snow collection and analysis of OCs were conducted at about 20 locations in the NWT and three locations in the Yukon (Swyripa and Strachan 1994, Palmer 1993). Results for OCs in snow were also available from the Taimyr Peninsula in the Russian Arctic. OCs were also determined in wet precipitation collected during 1995 in Iceland (Icelandic Meteorological Office, unpubl. data 1996), and in wet precipitation collected in August 1994 on the Taimyr Peninsula (Melnikov *et al.* 1995, 1996a, 1996b). Several current-use pesticides were determined in precipitation collected in northern Norway (Lode *et al.* 1995).

There are major uncertainties associated with determination of semi-volatile contaminants in snow. These compounds are not permanently encapsulated in snow crystals and may exchange with the atmosphere as snow morphometry changes. Thus, fresh snow generally shows much higher concentrations of contaminants than does aged snow (Gregor *et al.* 1995). Studies of OCs in snow at Amituk Lake (Cornwallis Island) showed that there were losses of most compounds from shallow snowpack, especially for di- and trichloroben-

zenes, heptachlor epoxide, α-chlordane, ΣPCB, and ΣDDT prior to melt (Semkin 1996). Hoff *et al.* (1995) have calculated that, based on theoretical considerations, less than 1.5% of the snowpack burden initially found at about –30°C remains at 0°C because of partitioning into the vapor phase as the temperature of the snowpack increases. This calculation assumes that there are no particles in snow for these chemicals to bind to although this still may be valid for the more volatile compounds such as HCB and HCH. Semkin (1996) has demonstrated further that the loss from the snowpack can be controlled by its thickness as only the top layer (perhaps of the order of 20-30 cm) is in contact with the atmosphere.

Table 6·7. Estimated seasonal deposition of selected contaminants to the Canadian Arctic, based on samples collected from snow collectors at various locations (μg/m²/season) (Barrie *et al.* 1997).

| Station | ΣHCH | ΣCHL | ΣDDT | HCB | ΣPCB |
|---|---|---|---|---|---|
| Alert, 1993/94 | 0.50 | 0.01 | 0.05 | 0.11[a] | 0.37 |
| Alert, 1992/93 | 0.88 | 0.03 | 0.05 | 0.06 | 0.19 |
| Eureka, 1991/92 | 0.78 | <0.01 | 0.01 | 0.01 | 0.26 |
| Mould Bay, 1993/94 | 0.25 | <0.01 | 0.03 | 0.02 | 0.23 |
| Cape Dorset, 1993/94 | 0.22 | 0.01 | 0.06 | 0.01 | 0.51 |
| Dawson City, 1993/94 | 0.19 | 0.03 | 0.04 | 0.01 | 0.41 |
| Whitehorse, 1993/94 | 0.12 | 0.02 | 0.02 | 0.01 | 0.20 |
| Whitehorse, 1992/93 | 0.07 | 0.04 | 0.05 | <0.01 | 0.43 |
| Tagish, 1993/94 | 0.20 | 0.03 | 0.04 | 0.01 | 0.35 |
| Tagish, 1992/93 | 0.28 | 0.09 | 0.01 | 0.01 | 0.24 |

a. Value reported to be suspect.

Deposition rates of major OC groups in snow at samplers in the Canadian Arctic are presented in Table 6·7. There is surprising uniformity in the deposition at these sites. Flux estimates for PCBs from a snow core on the Agassiz Ice Cap (406 ± 187 ng/m²) (Gregor *et al.* 1995, Franz *et al.* 1997) are well within the range of measured fluxes for these collectors. These fluxes are also in good agreement with fluxes of PCBs to sediments (Muir *et al.* 1996b). However, it is difficult to determine the significance of contaminant flux in snow in the context of delivery to the aquatic environment because of the many routes of OC loss during snow melt. The actual delivery of contaminants to the aquatic system is perhaps better estimated using the snowpack concentrations in samples collected toward the end of the accumulation season.

A wide range of fluxes of OCs to snow was observed in the Yukon Territory. The presence of mountains close to the North Pacific Ocean results in much higher deposition rates in the mountains of northern British Columbia, western Yukon, and neighboring Alaska. Stations in the Yukon River valley had wintertime deposition of approximately 600 ng PCB/m², but in the mountains, such as at White Pass, annual deposition is of the order of 1800 ng/m² (Palmer 1996). The increased snowpack in the mountains, consistent with the observations above, also affects the concentration of highly volatile compounds such as HCH. Analytical results indicated that shallow snowpacks in most of the Yukon had a mean ΣHCH concentration of 243 pg/L compared to the mean concentration at the high elevation, high snowfall site at White Pass which was 1348 pg/L. This difference is presumably due to less volatilization from the deep snowpack than from the shallow snowpack (Palmer 1996) as observed at Amituk Lake by Semkin (1996). A statistically significant difference such as this was not seen for PCBs, presumably due to their lower volatility.

During 1995, rainfall was collected in an open collector (area 1 m²) at Heimaey Island, Iceland (Icelandic Meteorological Office unpubl. data 1996) (Figure 6·7). The rainwater was adsorbed on polyurethane foam over a period of about 14 days. The foams, plus cotton swabs of the collector surface, were analyzed for PCBs, chlordane, DDT, and HCH compounds. ΣHCH dominated in rainfall at Heimaey Island (mean 31 ng/m²) compared to other OCs. γ-/α-HCH ratios during 1995 averaged 1.0 (range 0-9) indicating fresh sources of lindane at this location, as was observed for air samples from the same site. ΣPCB (sum of ten congeners) were the next most prominent OCs (18 ng/m²). On an annual basis, ΣPCB fluxes (436 ng/m²) were very similar to those for snow in the Canadian Arctic (Table 6·7).

A study of lindane and current-use pesticides in wet precipitation in Norway (Lode *et al.* 1995) included samples

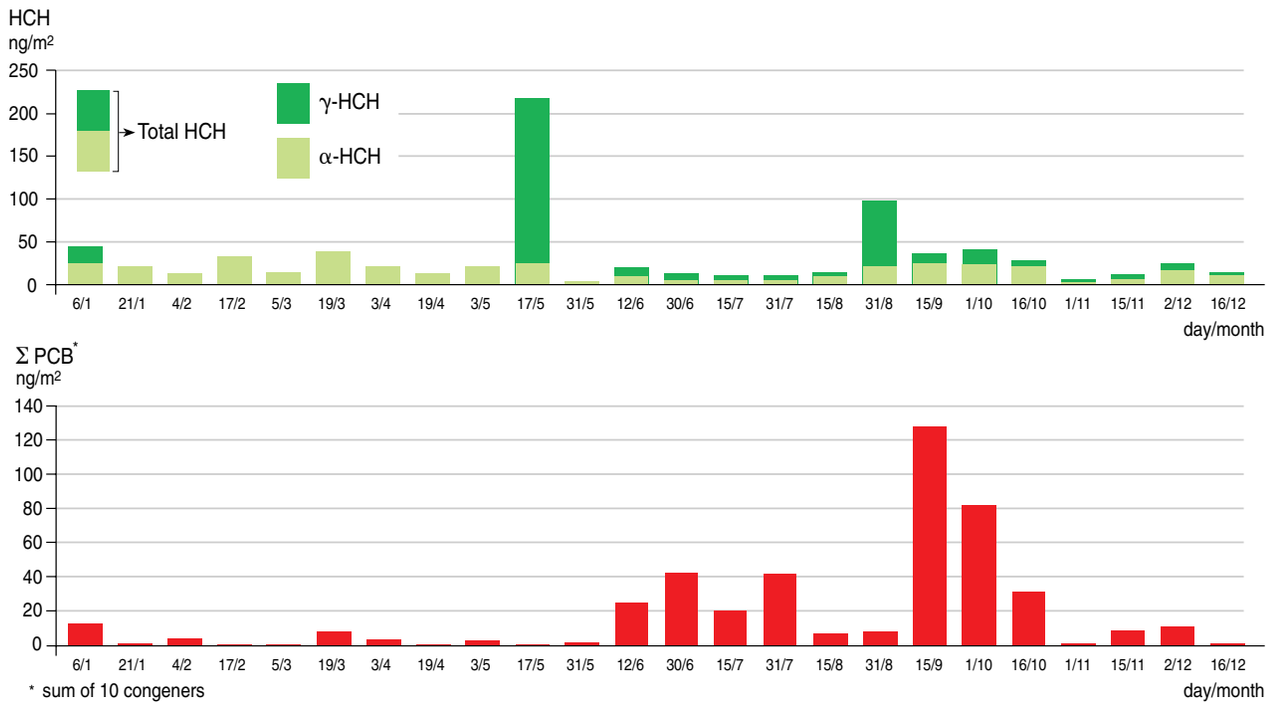



Figure 6·7. Flux of HCH and ∑PCB (sum of ten congeners) in precipitation at Heimaey Island, Iceland.

Table 6·8. Concentrations (ng/L, mean±SD) of major OC groups in snow and precipitation in samples collected during 1994 and 1995 from the Taimyr Peninsula, and from rainfall events in the Barents and Laptev Seas (Melnikov *et al*. 1995, 1996a, 1996b).

| Area | N | Latitude | Longitude | ΣHCH[a] | ΣCBz[b] | ΣCHL[c] | ΣDDT[d] | ΣPCB[e] |
|---|---|---|---|---|---|---|---|---|
| *Snow (May 1995)* | | | | | | | | |
| Taimyr Peninsula | 9 | 72-74°N | 98-106°E | 5.61±2.97 | 0.76 | 0.02±0.03 | 2.06±1.28 | 5.3±3.7 |
| *Precipitation (August 1994)* | | | | | | | | |
| Taimyr Peninsula | 7 | 74°33'N | 98°37'E | 0.88±0.42 | 0.46±0.26 | 0.13±0.20 | 3.2±1.5 | 11.9±6.1 |
| Laptev Sea | 3 | 74°00'N | 127-130°E | <0.1 | 0.07±0.12 | 0.10±0.17 | 1.1±0.77 | 1.1±0.65 |
| Barents Sea | 1 | 70°00'N | 53°30'E | 0.67 | 0.06 | <0.05 | 0.34 | 4.3 |

a. Sum of α- and γ-HCH.
b. Sum of HCB and pentachlorobenzene.
c. Sum of *cis*- and *trans*-chlordane and *cis*- and *trans*-nonachlor.
d. Sum of *p,p'*-DDE, DDD, DDT, and *o,p'*-DDT.
e. PCBs quantified using seven congeners (CBs 28, 52, 101, 118, 138, 153, 180).

from Tromsø. Lindane was not detected (< 1 ng/L) in samples from Tromsø, although it was readily detected in precipitation at two southern sites in Norway (Lista and Ås, near Oslo).

Snowcover on the Taimyr Peninsula in Russia was sampled and analyzed for POPs during May 1995 by the Russian Federal Service for Hydrometeorology and Environmental Monitoring (Roshydromet) (Melnikov *et al*. 1996a, 1996b). Precipitation samples (24 h) were collected from the same area and the Laptev Sea during August 1994. Concentrations of selected OCs in snow and precipitation are presented in Table 6·8. Additional data from this monitoring program, concerning OCs in snow deposited on sea ice, are discussed in section 6.6.4.1.1. The information necessary to determine the contaminant mass in the snowpack or fluxes in rainfall was not provided with these data and, thus, reporting is limited to concentrations. The concentrations of ΣHCH and ΣDDT found in the Russian snow and wet precipitation survey tend to be an order of magnitude higher than those reported for Canada (Barrie *et al*. 1997). For example, Swyripa and Strachan (1994) found ΣDDT concentrations ranging from <0.02 to 0.42 ng/L in snow at 20 sites across the NWT during 1991/92. However, PCB concentrations in snow from the Taimyr Peninsula were similar to levels found in the NWT by Swyripa and Strachan (1994) (0.9-13 ng/L).

### 6.6.1.3. Summary and conclusions – air and precipitation

A large amount of data is now available on levels of POPs, particularly OC compounds, in Arctic air and snow. The most frequently detected are the persistent OC pesticides, hexachlorocyclohexanes (α- and γ-HCH), toxaphene, chlordane-related compounds, and industrial products (PCBs and chlorobenzenes). DDT-related compounds were present at levels very near detection limits at both the Canadian and Norwegian sampling sites.

Although previous air measurements have been made for limited time periods during summer, results from year-round monitoring at two sites in northern Canada, Svalbard, and eastern Russia are now available which give far more information on temporal and spatial trends in air.

Spatial coverage for monitoring of airborne contaminants is circumpolar, which is not the case for precipitation. Most of the measurements of OCs in snow are from the Canadian Arctic and Russia; none are available from Greenland, Norway, or Alaska. Apart from precipitation monitoring in Iceland and on Taimyr Peninsula in Russia there has been little monitoring of rainfall for OC contaminants in the Arctic. This may not be significant for areas which receive most precipitation in the form of snowfall, but is a major gap for areas such as northern Norway/Kola Peninsula and the Bering Sea region of Russia and Alaska where a significant portion of precipitation is in the form of rain.

Concentrations of ΣDDT and ΣPCB in snow from the Taimyr Peninsula and Laptev Sea in 1995 were about ten times higher than observed in the Canadian Arctic in 1993. PCB concentrations averaged 10 ng/L in precipitation at the Taimyr Peninsula site which was about ten times higher than found in recent measurements in the Great Lakes region of Canada (Hoff *et al*. in press, Franz *et al*. 1998) or in samples collected in the Laptev Sea. The high levels are of particular concern given that only seven PCB congeners were analyzed (Melnikov *et al*. 1996a, 1996b); total PCB levels would be higher. The analytical laboratory had participated successfully in interlaboratory comparisons. However, there is clearly a need to confirm the high levels which could also be influenced by contamination of collection equipment by, for example, PCBs in electrical equipment near the sampler. This is difficult to study in an interlaboratory context.

Elevated levels of OC pesticides in Arctic air (especially lindane and chlordane) are correlated with long-range transport episodes from use areas in the mid-latitudes of North America and Europe (Figure 6·6). Higher concentrations of PCBs are also related to transport of air masses from industrialized areas of western Europe and eastern North America in the mid-latitudes. The results for air demonstrate that current and past use of OCs in the mid-latitudes of the northern hemisphere is the most likely source of OC contaminants to the Arctic environment. These OCs are resistant to environmental degradation and have high enough volatilities to continue to recycle in the environment. A considerable fraction of past production of PCBs and OC pesticides is still cycling in the abiotic and biotic environments.

The movement of fresh sources of lindane and chlordane from source regions was particularly evident from changes in ratios of γ-/α-HCH and *trans*-/*cis*-chlordane. Highest *trans*-/*cis*-CHL and γ-/α-HCH ratios were observed in May-June 1992 at Alert and in May-June 1993 at Ny-Ålesund (Svalbard).

Best correlations of OC concentrations with air mass movements were obtained with the Norwegian data, which were based on 48-hour samples. One week samples collected at two Canadian and the one Russian (Dunai) site are less suitable for back trajectory work because temporal resolution is lost. However, the larger size of the latter samples enables slightly better detection limits for most OCs.

Between long-range transport episodes of elevated levels, a significant 'background' concentration is observed of all OCs. This is due to volatilization of OCs from snow, plant, and soil surfaces, as well as to air-sea exchange.

What happens to OCs in snowfall after deposition, when snow surface area decreases as a result of snowpack metamorphosis processes, remains an open question. There are no detailed studies of the time variation of concentrations in

snow on the ground within hours to days following deposition. However, seasonal decreases in concentrations of OCs in snow over time, by a factor of two to ten times, have been documented.

Concentrations of OCs in Arctic air from northern Canada and Norway are generally one order of magnitude lower than in air from southernmost locations in the same countries. For example, Oehme *et al.* (1995d) found γ-HCH (lindane) concentrations ranging from 10-300 pg/m$^3$ at Lista (S. Norway) compared to levels of 2-38 pg/m$^3$ at Ny-Ålesund (Svalbard). PCB levels at Lista were also about ten times higher than at Svalbard or Svanvik (N. Norway) (Oehme *et al.* 1995b).

Air concentrations of most OCs at Alert in the Canadian High Arctic show a strong seasonal summer minimum in concentration corresponding to a summer maximum in precipitation. Less of a trend is observed at Ny-Ålesund or at Tagish in the southern Yukon. The magnitude of the summer minimum is roughly proportional to the substance's solubility in precipitation. At Tagish, HCH (Figure 6·4) and endosulfan do not show the summer dip in concentration. This is likely due to the strong inputs of air to the Arctic from the North Pacific Ocean over the Rocky Mountains into this site. In contrast, air arriving at Alert has spent much time over the ice-covered Arctic Ocean where there is a strong summer maximum in precipitation.

At most locations γ-/α-HCH ratios increased between March and June indicating fresh sources of lindane from temperate regions of the northern hemisphere. HCH profiles in Icelandic air during 1995 differed from the other sites by having much higher γ-/α-HCH ratios, indicating proximity to fresh sources of lindane.

The lack of NIMS data for toxaphene for all of the sites limits our ability to accurately predict current atmospheric loadings of toxaphene to the Arctic Ocean and is a matter that needs to be resolved.

There is a need to standardize the number of individual OC components measured in future air and precipitation monitoring. For example, the PCB results from Svalbard and Iceland were based on ten congeners which represented only 10-30% of ΣPCB (measured at Alert, Tagish, and Dunai). The analysis of total PCBs in snow and rainfall from the Russian Arctic was based on seven PCB congeners (these seven were also part of the suite used for the Svalbard and Iceland analyses).

## 6.6.2. Terrestrial environment

Information on levels of OCs in soils, plants, and terrestrial species in Arctic and subarctic regions was very limited prior to 1991, and data on temporal trends were virtually nonexistent (Wong 1985, Thomas *et al.* 1992). Long-lived ungulates such as *Alces alces* (moose in America, elk in Europe) and caribou and reindeer (*Rangifer tarandus*) accumulate significant levels of cadmium in their organs, but information on levels of persistent OCs in these important species, as well as in waterfowl, were too limited to derive any conclusions about spatial or temporal trends of contaminants.

Although the levels of OCs detected in terrestrial mammals (Thomas *et al.* 1992) were substantially lower than in marine mammals and birds, the importance of species such as caribou and reindeer as food species in communities across the north has made the determination of baseline residue data particularly important. Both caribou and reindeer are a staple source of food for people and other predators in the north, and their potential level of contamination is therefore of great concern. *Rangifer* has a winter diet consisting primarily of lichens, which are noted for their ability to accumulate nutrients and contaminants from the air. Other terrestrial animals consumed by humans include waterfowl, game birds, and shorebirds and their eggs. Other terrestrial species such as mink and wolves are top trophic species that may serve as sensitive indicator species to assess trends in terrestrial environmental contamination and ecosystem health.

### 6.6.2.1. Soils and plants

• *Soils*

Data on OCs in soils are available from Canada and Russia. The Canadian study involved collection of 3500 soil and 1600 vascular plant samples in the immediate vicinity of recently active (main) military radar (or DEW line) sites (section 6.2.4.1) as well as at 'intermediate' sites, i.e., those abandoned during the 1960s (Reimer *et al.* 1991, 1993a, 1993b, 1993c, 1994, Dodd and Reimer 1992, Dushenko and Reimer 1994, Grundy *et al.* 1994). 'Site background' (within several kilometers of the site) and 'remote' (20 km or more from the site) locations were also sampled. PCBs and lead were the primary contaminants found along the DEW Line. The average (Aroclor) PCB concentrations for 525 associated plants and soils collected at various background locations and different radar sites ranged from about 1 ng/g to 10 000 ng/g dw (Figure 6·8a; next page). The PCB results indicate that past activities at these sites, such as spills, solid waste disposal, and down-the-drain discharge to sewage outfalls, have had a significant influence on concentrations relative to local background and more remote Arctic locations. Lower concentrations of PCBs in plants relative to underlying soils indicate that not all of the PCBs occurring in the soils are directly bioavailable to plants and/or a portion of the PCBs bioaccumulated is lost during leaf/plant senescence. Significantly higher concentrations of PCBs are also found in soils (maximum 35 ng/g dw) and plants (maximum 66 ng/g dw) from site background areas than more remote Arctic locations (0.90 and 1.71 ng/g dw) indicating that the sites can serve as local aerial sources of PCBs to the surrounding ecosystem. This has been confirmed by the analysis of congener signatures in both soils (Bright *et al.* 1995a) and plants. The ratio of PCB concentrations in soils to plants also decreases when moving away from sites toward background and remote locations (Figure 6·8a). This suggests a decline in the importance of PCBs redistributed to the surrounding tundra from local sources (e.g., spills) relative to long-range atmospheric transport. Concentrations of PCBs in plants are found to be significantly correlated with soil levels ($r^2 = 0.98$, $p < 0.001$) from background and radar stations (Figure 6·8b).

Soil samples were collected from nine locations in the eastern and western Russian Arctic during August 1994 and 1995 and analyzed by the Russian 'Regional Center Monitoring of the Arctic' (Annex Table 6·A3, Melnikov *et al.* 1996a, 1996b). The samples were collected some distance from population centers. PCBs and HCH isomers were the most prominent OCs detected. Highest ΣPCB (7.9 ng/g dw, seven congeners) concentrations were found in peaty soils from the Pechora River mouth area. Lowest PCB and sum of pentachlorobenzene and HCB levels were found in samples from the eastern Russian mainland, eastern Taimyr Peninsula, and the Olenek Gulf. Concentrations in the Russian soils were similar to levels in 'background' sites in the Canadian Arctic (0.9-1.7 ng/g dw), but direct comparison is problematic because the latter results are based on the sum of 80 congeners (Bright *et al.* 1995a).

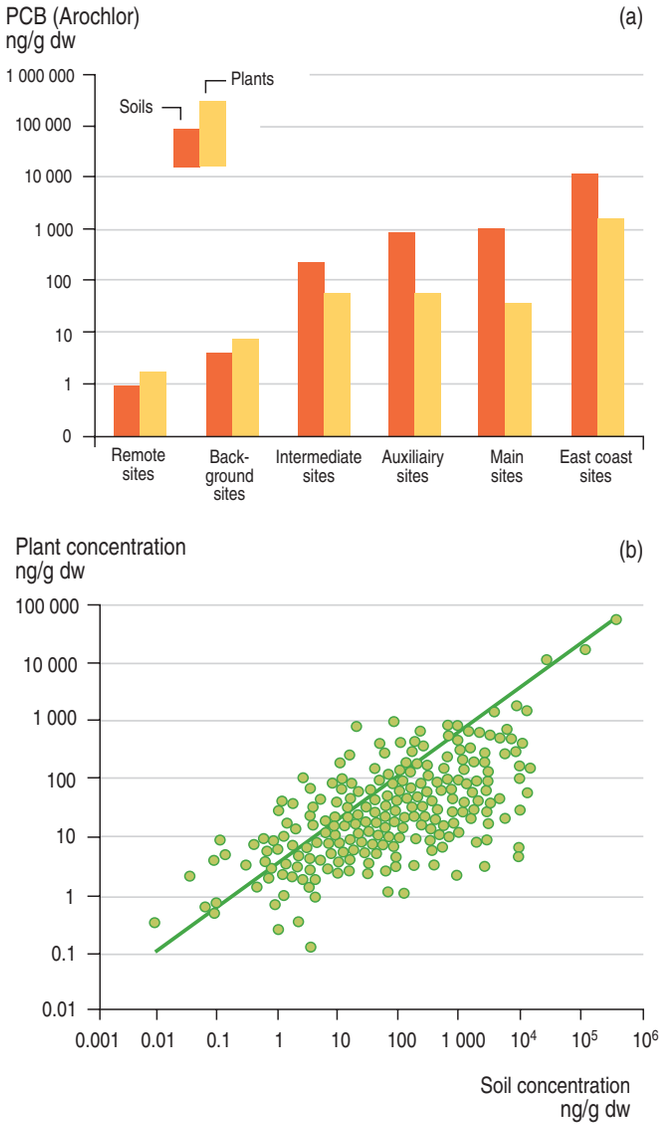



Figure 6·8. Concentrations of Aroclor PCBs in soils and plants (ng/g dw) associated with military radar sites in the Canadian Arctic. a) Mean PCB concentrations in soils and associated plants from remote (>20 km), background (<20 km), intermediate, auxiliary, main, and east coast DEW line radar sites. Intermediate sites were abandoned in the 1960s. Auxiliary and main sites were used until the 1990s. b) Soil–plant PCB relationships at background and radar sites.

• *Plants*

Plants are valuable indicators of contaminants entering the terrestrial ecosystem. They accumulate air- and soil-borne pollutants and form the base of the Arctic terrestrial food web. Perennial mosses and lichens are particularly good indicators of atmospheric deposition in remote and northern areas (Thomas *et al.* 1992, Ross 1990, Grodinska and Godzik 1990, Nash and Gries 1995a, 1995b). Annual vascular plants are also important because they are important components in the summer diets of herbivores.

Results for OC contaminants (including some less persistent OC pesticides) in plants are available from Canada, Russia, Finland, and Norway. Mean levels of all OCs in plants in both North America, Scandinavia, and Russia are generally low compared to levels in terrestrial fauna (Annex Table 6·A3). Excluding plant samples collected in the immediate vicinity of military radar sites, the dominant OC contaminants are PCB, DDT, and HCH with overall ranges of < 0.1-7.8, 0.12-9.1 and < 0.1-12.4 ng/g dw, respectively. Chlordanes and HCB were present in lower levels (range < 0.1-2.44 and < 0.1-3.1 ng/g dw, respectively). Dieldrin levels were lowest in plants (range < 0.01-1.0 ng/g dw). Toxaphene was found to be the most abundant OC in lichen (8-43 ng/g dw) and saxifrage (2-43 ng/g dw) from Ellesmere Island (France *et al.* 1997). There are no results for toxaphene in plants from other Arctic areas with which to compare these results.

No geographical trend in ΣPCBs is evident in the Russian data for lichen, bryophytes (mosses), and willows (*Salix*). Mean ΣPCB concentrations ranged from less than 0.1-7.8 ng/g dw. These results are based on seven congeners which, in the case of *Sphagnum* sp. analyzed by Himberg and Pakarinen (1994), accounted for 50% of PCB in samples from N. Finland. Assuming a factor of two times to allow comparisons with other locations, it can be concluded that PCB levels in Russian plants are generally lower than in plants from northern Norway and Finland, but higher than found in the Canadian Arctic (Figure 6·9).

Concentrations of α-HCH, HCB, γ-HCH, *p,p'*-DDE, and *p,p'*-DDT in lichens from southern Ellesmere Island were lower than those for lichens from Europe and southern Canada, but similar to residues measured in lichens from other remote polar sites (central NWT in Canada, Spitsbergen, and Antarctica). Total chlordane and toxaphene were present at higher concentrations on Ellesmere Island than observed in southern Ontario lichen (Muir *et al.* 1993). HCB and total chlordanes in Ellesmere Island lichens were proportionally higher than those found from more southern growing species. Calamari *et al.* (1991) actually found higher HCB residues in polar compared to temperate vascular plants (mosses) and lichen, and suggested that this was due to the farther northward transport of this volatile OC. Simonich and Hites (1995) also found higher levels of HCH isomers and HCB in tree bark from northern latitudes (40-70°N) and relatively low levels in the tropics.

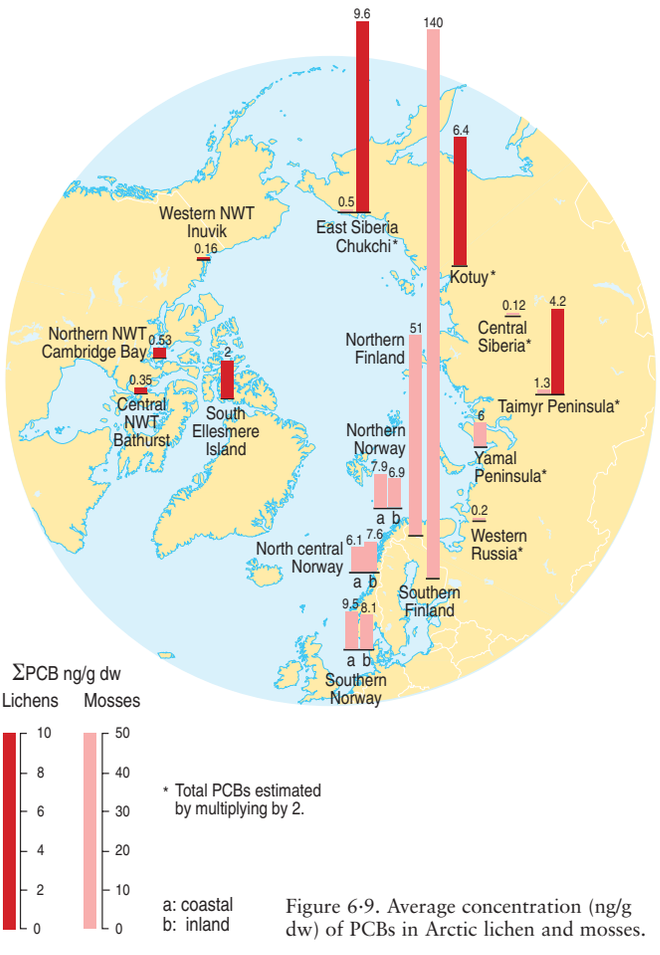



Figure 6·9. Average concentration (ng/g dw) of PCBs in Arctic lichen and mosses.

A major geographical survey of Arctic/subarctic mosses (*Hylocomium splendens*) was conducted by Lead *et al.* (1996a) along a transect from southern to northern Norway. Temporal trends in this survey are discussed in section 6.7.2.1. ΣPCB concentrations (sum of 33 congeners) were lower in samples from the two northern Norway areas (coastal, 6.9 ± 2.6; inland/continental, 7.9 ± 2.3 ng/g dw) than in southern Norway (9.5 ± 2.3 ng/g dw) (Figure 6·9). Differences were most pronounced for octachlorobiphenyls (0.30 ± 0.47 ng/g in the south vs. 0.12 ± 0.14 ng/g in the north). Himberg and Pakarinen (1994) also found much lower mean concentrations of PCBs (49 congeners) in mosses (*Sphagnum* sp.) from northern Finland (51 ± 14 ng/g dw) compared with southern Finland (140 ± 17 (SE) ng/g dw), and saw a uniform decline, of about three-fold, in all PCB homologue groups. The much higher levels of PCBs in the study by Himberg and Pakarinen (1994) may be due to air-drying of the sphagnum in the laboratory, a technique that Lead *et al.* (1966a) showed contributed to elevated levels of PCBs.

It is noteworthy that, even across a relatively small geographic area such as from northern to southern Finland and Norway, and northern to southern Ellesmere Island, a north-south gradient of OC levels is detectable (Himberg and Pakarinen 1994, Lead *et al.* 1996a, France *et al.* 1998). At Ellesmere Island, this is unlikely to be related to air concentrations, which are quite similar over large geographic areas, but may be related to gradients in wet precipitation. In Finland and Norway, however, the differences may be related to the proximity of the southern sampling sites to urban areas (Figure 6·9).

France *et al.* (1998) detected the less persistent, but widely used OC pesticides, tetra- and pentachlorophenol (as anisoles), endosulfan, methoxychlor, trifluralin, and triallate in lichen and saxifrage from Ellesmere Island. Concentrations of these pesticides in saxifrage were in the 0.1-1.0 ng/g (dw) range, similar to levels of other individual OCs such as dieldrin and chlordane isomers (France *et al.* 1997). Levels of pentachloroanisole (PCA) in the lichen from Ellesmere Island were about ten times lower than observed in southern Ontario lichen (Muir *et al.* 1993), suggesting that there are local sources of PCA in populated areas. In contrast, Simonich and Hites (1995) found PCA concentrations increased with latitude (40-70°N) in tree bark, similar to HCH and HCB.

### 6.6.2.2. **Terrestrial herbivores**

#### 6.6.2.2.1. **Caribou and reindeer**

The reindeer of northern Europe and Asia and caribou of North America are considered to be a single species. They inhabit the Arctic tundra, subarctic taiga, and coniferous forests. Summer diets include grasses, sedges, twigs, leaves, and mushrooms, while winter diets include horsetails and sedges, twigs, and lichens. Lichens are the mainstay of the species' winter diet. In contrast, High Arctic species, such as Peary caribou and Svalbard reindeer, feed extensively on grasses and sedges, as well as mosses (Staaland *et al.* 1988). Differences in selected food at sampling areas have a minor influence on the concentrations of POPs found in reindeer tissue (Kelsall 1968, Parker 1978). The defined ranges and distribution of caribou and reindeer herds, and the simple plant-herbivore food web make it a good species for the examination of temporal and geographical variation in terrestrial contaminant deposition.

Caribou herds have distinct migratory ranges which may cover hundreds of kilometers. In Canadian mainland tundra subspecies, the tundra summer ranges and forested winter ranges may be as much as 1300 km apart (Banfield 1974). Packs of wolves will have migration patterns similar to the caribou herd. However, most populations of caribou and reindeer move over considerably smaller distances. In Norway, Sweden, and Finland, reindeer are semi-domesticated and their traditional ranges depend on which Saami village the herd belongs to.

Recent data on levels of OC contaminants in *Rangifer* tissues are available from Russia, Canada, Svalbard, northern Norway, and Sweden, almost representing the circumpolar distribution of the species (Annex Table 6·A4). Unfortunately, data are difficult to interpret for a number of reasons. Tissues used and compounds measured were not always identical, sample sizes ranged from one to 35 individuals per location, and detection limits varied. Liver was the tissue common to most studies and OC levels were most commonly measured as liver wet weight concentrations. Data for the most common OCs from all locations are presented graphically (Figure 6·10; next page), after the application of some course correction factors to convert all data to liver ww concentrations if otherwise reported.

The Russian data are sparse, consisting of only one reindeer at each site. Analytical results are available for two consecutive years (1994 and 1995) and the OC levels differ considerably between these years. This makes it difficult to compare the Russian results with those of other countries, as the 1994 results are generally higher than for Canadian caribou and Svalbard reindeer, while the 1995 levels are lower or comparable. For both years, mean ΣPCB and ΣDDT levels are higher in the Russian samples than in Canadian *Rangifer*. It should be noted that the ΣPCBs in the Russian reindeer are based on only seven congeners (Melnikov *et al.* 1996a, 1996b). These seven, CBs 28, 52, 101, 118, 153, 138, and 180, were major congeners in Canadian caribou fat, comprising about 66% of ΣPCB (Figure 6·12; section 6.6.2.5).

In Canada, the predominant OCs in caribou fat and liver are ΣHCH, chlorobenzenes, ΣPCB, and chlordanes. In Russia, ΣPCB dominates in both data sets, followed by ΣDDT, HCB, and ΣHCH. Higher relative levels of ΣDDT and ΣPCB are also seen in Russian lemming, ptarmigan, and brant geese (Annex Table 6·A4). Reindeer from Svalbard have lower POP levels in their fat and liver and the predominant POPs are HCB and HCH followed by ΣDDT. PCB levels were below detection limits (which at 14 ng/g ww were much higher than other studies) in samples from Svalbard. Swedish reindeer also have low levels of HCH, PCBs, and ΣDDT, similar to the levels at Svalbard. When the data sets from Svalbard, Canada, and Sweden are compared on a lipid weight basis, levels of most persistent OCs are very similar (Annex Table 6·A4).

Most of the Canadian data are from recent studies by Elkin and Bethke (1995), who examined OC levels in several herds of free-ranging caribou in the Northwest Territories. This data set is of interest because it is capable of showing spatial variation in levels, and also because the various herds are followed and preyed on by wolves, which were also examined. A wide range of OC contaminants were detected, with most compounds found at very low levels and with less toxic compounds (e.g., HCH isomers) predominating. Moderate levels of HCB and α-HCH were detected at all study sites, as in other terrestrial herbivores in the Canadian Arctic (MacNeil *et al.* 1987, Thomas *et al.* 1992, Salisbury *et al.* 1992). In general, a significant west to east trend in concen-

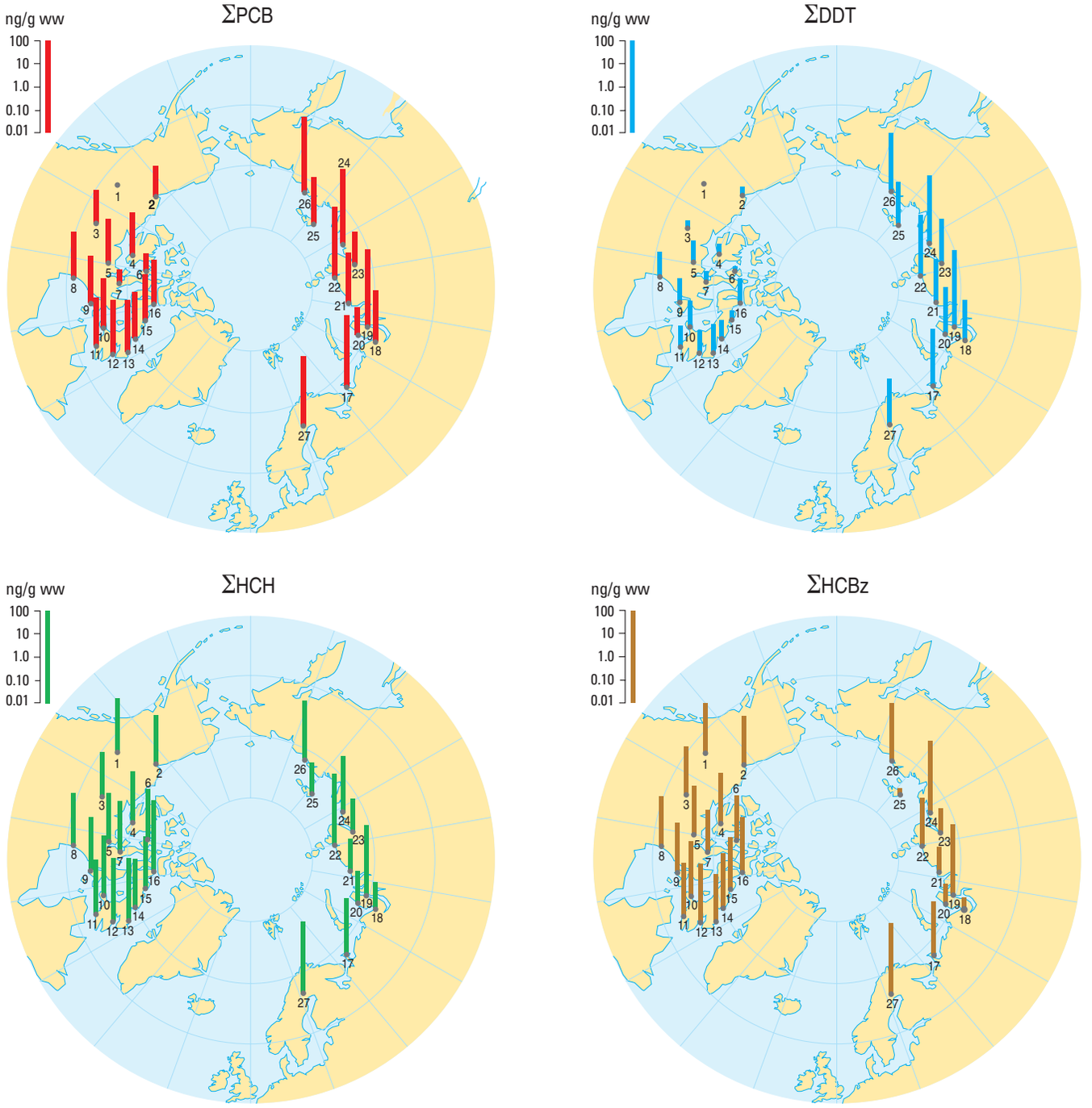



Figure 6·10. Observed and predicted organochlorine concentrations (ng/g ww) in caribou and reindeer (*Rangifer tarandus*) liver. All data are presented in, or were derived from data in Annex Table 6·A4. The liver wet weight concentrations were predicted from lipid wet weight with the linear equation: liver ww = k × lipid ww, in which the k's were derived from raw data from three Canadian Arctic locations (Elkin pers. comm.) and were as follows: 0.0675 for HCBz, 0.0792 for ΣDDT, 0.2675 for HCH, and 0.0973 for ΣPCB.

trations of OCs was observed with lowest levels in caribou from the western (Inuvik) and central herds (Bathurst and Qamanirjuaq (Arviat) and highest levels in the eastern Arctic herds from Cape Dorset and Lake Harbour on southern Baffin Island (Figure 6·10, Annex Table 6·A4). Total HCH (ΣHCH) ranged from 3.3 ng/g lw from Inuvik caribou to 39.8 ng/g at Cape Dorset, and consisted almost entirely of α-HCH. HCB residues ranged from a mean of 20 ng/g lw in fat of Taloyoak caribou to 129 ng/g in Lake Harbour animals. Total PCB residues (sum of 43 congeners) ranged from a mean of 0.55 ng/g lw in fat of Inuvik caribou to 32 ng/g at Cape Dorset (Annex Table 6·A4).

Oxychlordane, a major metabolite of several compounds in the pesticide technical chlordane, was the major chlordane-related compound detected. Total chlordane levels ranged from 0.04 ng/g lw in Inuvik caribou to 5.0 ng/g at Cape Dorset, with oxychlordane and to a lesser degree heptachlor epoxide predominating. ΣDDT, comprised largely of

*p,p'*-DDE, was below detection limits in Inuvik caribou, and ranged from 0.11 ng/g lw in Beverly caribou to 2.58 ng/g at Cape Dorset. Total toxaphene levels found in 12 pooled fat samples from four sites ranged from non-detectable to 23 ng/g lw.

The PCB congener patterns in fat were similar in all herds in the Canadian Arctic, with greater accumulation of more highly chlorinated congeners, especially CB 153 (Figure 6·12). Higher levels of trichlorobiphenyl and tetrachlorobiphenyl compounds found in the caribou, relative to other terrestrial mammals and birds, may be a product of the direct air → lichen → caribou food chain.

Levels of PCDD/Fs, and planar PCBs in *Rangifer* from Canada, Sweden, and Norway are summarized as TCDD TEQs and presented in Annex Table 6·A22. The Canadian study was made using subcutaneous back fat from female caribou in the Northwest Territories and the Yukon (Hebert 1994, Hebert *et al*. 1996). The samples were analyzed for 27 PCDD/F congeners plus five PCBs (CBs 77, 126, 169, 105, 118). PCDD/F and planar PCB levels were also determined in homogenates of male reindeer kidney fat collected from two sites in the Swedish Arctic, at Aitejokk, near Abisko (1992) and at Ammarnäs (1992-94), and in kidney fat from two sites in Norway, Jarfjord and Stilla (Schlabach and Skotvold 1996a, 1996b). Jarfjord is located in the vicinity of the Syd-Varanger smelter near Kirkenes. The samples from Stilla, in west Finnmark served as references for comparison with Jarfjord.

In the Canadian study, levels of PCDD/Fs and planar PCBs were extremely low in animals from all herds. Only 2,3,7,8-substituted PCDDs were observed although non-2,3,7,8-substituted PCDD congeners were also determined. TCDD was found in only two fat samples (Cape Dorset, 0.73 pg/g ww and Lake Harbour, 0.14 pg/g) despite very low detection limits ranging from 0.02 to 0.43 pg/g. OCDD was the only PCDD or PCDF that was found in the majority of fat samples, at levels ranging from <0.38 pg/g to 4.69 pg/g ww in the Cape Dorset sample. Among PCDF congeners, 1,2,4,7,8-PnCDF was present in all samples except Finlayson, at levels ranging from 0.24 to 0.74 pg/g. Five other non-2,3,7,8-substituted PCDF congeners, 2,3,6,8-TCDF, 2,3,6,7-TCDF, 1,2,4,6,8-PnCDF, 1,2,4,6,7,8-HxCDF and 1,2,4,6,8,9-HxCDF, were found only in the Lake Harbour sample at levels <1 pg/g. TEQs (PCDD/Fs plus planar PCBs) ranged from 0.33 to 3.3 pg/g ww and were higher in the eastern Canadian Arctic herds, consistent with results for *ortho*-substituted PCBs and OC pesticides.

In Sweden, the samples from Aitejokk had no measurable amounts of PCDD/Fs (< 1 pg/g ww), but all five planar PCBs were found (Annex Table 6·A22). The concentrations ranged from 1.5-1.9 pg TEQ/g lw (1.3-1.6 pg TEQ/g ww) for the planar PCBs. Two of the three samples from Ammarnäs, which is farther south than Aitejokk, contained 2,3,7,8-TeCDF (0.48 pg TEQ/g ww). All three homogenates contained all three nPCBs with TEQ levels of 0.91-1.03 pg/g ww (C. de Wit unpubl. data).

In Norway, the samples from Jarfjord had higher PCDD/F levels (7-13 pg TEQ/g ww) than the samples from Stilla (0.58-0.81 pg TEQ/g ww). Measurable amounts of most 2,3,7,8-substituted PCDD/Fs were found in samples from both sites, with PCDFs dominating (Schlabach and Skotvold 1996a, 1996b). The TEQ levels found in the Stilla reindeer samples are similar to those found in Sweden and Canada.

The presence of 2,3,7,8-TeCDF in the more southern Swedish herd, at Stilla, and in eastern Canadian Arctic herds is consistent with an atmospheric transport source probably originating from urban areas to the south. OCDD, PeCDD/Fs, and HxCDD/Fs were also higher in eastern Canadian Arctic samples and in the samples from Stilla, which is consistent with long-range transport from the south.

#### 6.6.2.2.2. **Waterfowl and other terrestrial birds**

The Arctic has over 150 species of breeding land birds, most of which fly south to warmer climates, either to warmer tundra or warmer coastlands or forests, to avoid harsh winter conditions (Stonehouse 1989). Some species, such as many ducks and geese, migrate only as far south as is necessary or possible, and for these species the degree of migration may differ in different areas of the Arctic or may differ among years. Most of these species will put on lipid stores before their flight south, with species that have long flights acquiring larger reserves. Some species may overwinter in the Arctic (e.g., in Iceland), especially if forced to do so, or may move to subarctic or northern temperate habitats. Small seed-eaters such as snow buntings and redpolls, and other species including corvids, larger birds of prey, and some ptarmigan are in this category. Some species are very transient in the Arctic, for example, some shorebirds that stay only long enough to mate or hatch eggs and then start their gradual migration south. Their southern overwintering habitats include some shorelines, estuaries, sewage lagoons, garbage dumps, bodies of water receiving agricultural or industrial runoff, and agricultural fields, all of which can be very contaminated.

The migratory birds of most interest include shorebirds, waterfowl, including geese, swans, and ducks, and birds of prey, which may include owls, accipiters, buteos, and falcons. Waterfowl and terrestrial game birds and their eggs are harvested to varying degrees for consumption (Coad 1994). Waterfowl, small passerines, and shorebirds also constitute major prey species for birds of prey such as the peregrine falcon, whose population status is of concern.

Most data available are from Canada and Russia, the countries with the largest Arctic land areas. For the most part, data collections are scattered geographically, and many species were sampled, thus, it is difficult to make generalizations. OC levels in Canadian waterfowl are given in Table 6·9; OC levels from body analyses of prey species of the peregrine falcon from near Rankin Inlet, NWT, and from Sweden are included in Annex Table 6·A4; and levels of OCs in eider ducks and gulls are presented in section 6.6.4.6.

To simplify the presentation of the Canadian data, values were grouped by trophic levels and geographic area. Northern collection sites were grouped into western Arctic (north of 60°N west of 95°W) or eastern Arctic (north of 55°N east of 95°W, so as to include Hudson Bay, northern Quebec, and northern Labrador). Baker Lake was kept with the eastern sites because of its proximity to Hudson Bay, and Spence Bay was kept with the western sites because of the influence of western Arctic waters. Terrestrial birds were grouped according to trophic level as follows: browsers – ground dwellers such as grouse and ptarmigan that feed mainly on terrestrial vegetation; grazers – geese that graze mainly on aquatic and terrestrial vegetation; omnivores – surface-feeding ducks with a varied diet consisting mainly of aquatic vegetation; molluscivores – diving ducks feeding mainly on invertebrates; piscivores – diving ducks feeding mainly on fish.

Levels of OCs in Canadian waterfowl were generally quite low, in the low ng/g range, with exceptions (Table 6·9). The highest OC levels were found in birds feeding at the upper trophic levels, the molluscivores and piscivores. The browsers and grazers contained the lowest levels of organic

Table 6·9. Range of organochlorine residue levels in breast muscle of waterfowl and game birds harvested in northern Canada in 1988-94 (ng/g ww). All data are from Braune (1994 unpubl. data).

| Trophic category[a] | Region | N[b] | ΣPCB | ΣDDT | ΣCHL | ΣCBz | ΣHCH | Mirex | Dieldrin |
|---|---|---|---|---|---|---|---|---|---|
| Browsers | East | (5, 9, 39) | <1.0-3.1 | <1.0 | <1.0 | <1.0 | <1.0 | <1.0 | <1.0 |
| | West | (12, 22, 91) | <1.0-9.6 | <1.0-1.5 | <1.0-2.5 | <1.0-1.9 | <1.0 | <1.0 | <1.0-5.0 |
| Grazers | East | (16, 39, 234) | <1.0-44.9 | <1.0-30.0 | <1.0-4.5 | <1.0-1.6 | <1.0 | <1.0 | <1.0-3.2 |
| | West | (7, 11, 59) | <1.0 | <1.0-3.1 | <1.0-4.7 | <1.0-1.9 | <1.0 | <1.0 | <1.0-2.0 |
| Omnivores | East | (2, 4, 30) | <1.0-87.0 | <1.0-31.5 | <1.0-2.3 | <1.0-2.0 | <1.0 | <1.0-1.6 | <1.0-1.3 |
| | West | (8, 31, 149) | <1.0-115 | <1.0-650 | <1.0-12.8 | <1.0-62.4 | <1.0-1.0 | <1.0-1.6 | <1.0-15.9 |
| Molluscivores | East | (14, 30, 175) | <1.0-1076 | <1.0-403 | <1.0-102 | <1.0-35.9 | <1.0-27.8 | <1.0-222 | <1.0-120 |
| | West | (12, 42, 178) | <1.0-129 | <1.0-127 | <1.0-13.2 | <1.0-15.4 | <1.0-26.5 | <1.0-1.7 | <1.0-3.8 |
| Piscivores | East | (7, 16, 55) | <1.0-1695 | <1.0-951 | <1.0-105 | <1.0-120 | <1.0-2.4 | <1.0-268 | <1.0-54.7 |
| | West | (2, 5, 5) | 1.1-909 | 32.0-530 | 2.9-75.7 | 1.6-18.3 | <1.0-9.9 | <1.0-9.2 | 1.0-17.4 |

a. Species are grouped by feeding habit: Browsers (grouse, ptarmigan); Grazers (geese, tundra swan); Omnivores (black, mallard, teal, pintail, wigeon, shoveller); Molluscivores (eider, scoter, scaup, oldsquaw or long-tailed duck, ring-necked duck, goldeneye, bufflehead, canvasback); Piscivores (mergansers, loons or divers).
b. Numbers in brackets refer to: (total number of sites, total number of collections, total number of birds).

ΣCBz = Sum of 1,2,3,5 and 1,2,3,4 tetrachlorobenzene, pentachlorobenzene and HCB.
ΣHCH = Sum of α-, β-, and γ- hexachlorocyclohexanes.
ΣCHL = Sum of oxy-, *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor, and heptachlor epoxide.
ΣDDT = Sum of *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT.
ΣPCB = Sum of PCB congeners, standardized to the following 29 congeners: 28, 31, 44, 52, 60, 66/95, 87, 97, 99, 101, 105, 110, 118, 138, 141, 146, 153, 170/190, 171, 172, 174, 180, 182/187, 183, 194, 195, 201, 203, 206.
Mirex = Sum of photo-mirex and mirex.

contaminants. The highest OC levels were found for ΣPCB and ΣDDT in piscivores and molluscivores collected from eastern Arctic communities. These generally contained higher levels of organic contaminants than those from comparable collections from western sites. The wider range of contaminant concentrations found in the eastern birds reflects a greater risk of exposure to contaminants in overwintering or staging areas due to the greater prevalence of contaminated areas in eastern North America versus western North America. Historically, some of the eastern overwintering areas, such as the Great Lakes, the Gulf of Mexico, and the eastern American seaboard, have been much more contaminated than some of the western areas. This difference is clearly illustrated by mirex, a contaminant characteristic of the lower Great Lakes (Kaiser 1978, Comba *et al.* 1993). Mirex is virtually non-detected in western Arctic birds, but is quite measurable in eastern Arctic birds (Table 6·9).

### 6.6.2.3. **Birds of prey**

Arctic and temperate birds of prey are particularly susceptible to the effects of contaminants because most prey species contain contaminants from overwintering areas. In addition, prey species such as marine invertebrates are often also carnivorous, hence the potential for biomagnification is great. Over the past 40 years, similar trends in contaminant levels (section 6.7.2.3) and reproductive effects (section 6.8.1.3) have been seen in both Eurasia and North America. Recent data are discussed here, and data collected between 1960 and the present are given in Annex Table 6·A5.

#### 6.6.2.3.1. **North American peregrine falcon**

The tundra peregrine falcon (*Falco peregrinus tundrius*) breeds in the tundra regions of Canada, Alaska, and Greenland. The main breeding areas are southern Baffin Island, Ungava Bay, western Hudson Bay, the central Arctic coast, and the interior barrens of the Northwest Territories (White 1968). Its overwintering range extends from the Great Lakes in Canada, through the USA to Texas, and even to Uruguay in South America (Court *et al.* 1988). The American peregrine (*F. p. anatum*) breeds in forested areas from the treeline south to California and Mexico.

The peregrine falcon population near Rankin Inlet on western Hudson Bay was examined in the time periods 1965-1987 and 1980-1987 (Court *et al.* 1990, Peakall *et al.* 1990), and 1991-1994 (Johnstone *et al.* 1996). PCBs and DDE were the most abundant residues in peregrine falcons in eggs and blood plasma (Annex Table 6·A5). A total of 28 eggs, representing 20 clutches, were collected for analysis between 1991 and 1994. Mean concentration of PCBs (as Aroclor 1254:1260, 1:1) and *p,p'*-DDE in eggs were 8.3 and 4.4 μg/g ww, respectively. These levels were lower than reported in other recent studies of tundra peregrines. Ambrose *et al.* (1988) studying *F. p. tundrius* in Alaska, reported mean *p,p'*-DDE levels of 9.3 μg/g while Peakall *et al.* (1990) reported a mean of 6.8 μg/g for 26 eggs collected between 1980 and 1986 in Canada. Levels in gyrfalcon, an Arctic resident believed to feed mainly on Arctic hare and ptarmigan, are considerably lower than in peregrines, approximately 30 to 70 times lower (Thomas *et al.* 1992).

Results of analyses of several waterfowl prey species of the peregrine falcon from the 1991-1994 period, from near Rankin Inlet are given in Annex Table 6·A4 and Table 6·9. The relatively high levels in these migratory prey species have implications for several predators, particularly avian and human, for which the migrating birds provide a food source. There are, however, sources of relatively clean prey for peregrines in Rankin Inlet. Up to one-third of the total biomass of prey consumed are mammalian species, namely ground squirrels and microtine rodents (Bradley and Oliphant 1991) (Annex Table 6·A4). These are non-migratory species, so any contamination in their tissues represents contamination of the immediate environment around Rankin Inlet.

Mean ΣPCB and DDE levels in blood plasma of peregrine nestlings (0.12 and 0.06 ng/g ww, respectively) were about one-half or less than the levels in adult males (0.20 and 0.32 ng/g) and almost one-tenth the levels found in adult females (0.95 and 0.6 ng/g) (Annex Table 6·A5). The difference in contaminant levels between female and male peregrines may be partly attributed to differences in diet. Female peregrines, being larger than males, eat larger prey. In Rankin Inlet, the larger prey species included oldsquaw (long-tailed duck, *Clangula hyemalis*), black guillemot, and pintail (*Anas acuta*), which have higher contaminant levels than the smaller prey species in the area (Bradley and Oliphant 1991).

#### 6.6.2.3.2. **Eurasian peregrine falcon**

The Fennoscandian population of peregrine falcon (*Falco peregrinus*), some of which inhabit the Arctic, declined from 2000-3500 pairs pre-1950 to about 65 known pairs in 1975 (Lindberg 1995a). The species inhabits both Arctic and southern areas of Fennoscandia. The Arctic populations feed on a broad spectrum of migratory birds that overwinter in Europe and Africa. Arithmetic means of DDE, ΣPCB and dieldrin in eggs for 1991-1994 (32 clutches) were 2.9, 12, and 0.14 μg/g ww, respectively. Concentrations of DDE and PCB were not significantly different between eggs from the northern and the southern populations. Factors explaining this are the importance of the wintering areas for egg concentrations, the more aquatic-based food for the northern population, as well as the fact that the prey of the northern population largely consists of migratory species (Lindberg *et al*. 1985, Lindberg 1995a).

Studies on a population of peregrine falcon on the Kola Peninsula have been ongoing since 1977 (Henny *et al*. 1994). The production rate between 1987-1991 was 1.94 young per active nest. Egg-shell thinning (11.4%) was similar to levels in Alaska. The geometric mean concentration of DDE in eggs was 3.5 μg/g ww (Annex Table 6·A5) and the PCB concentrations were higher than those found in Alaskan peregrines (3-21 μg/g ww, geometric mean of 7.3 μg/g ww). Concentrations of PCDD/Fs and planar PCBs in eggs were also relatively high, with combined TEQ levels of 86-640 pg/g ww (Annex Table 6·A22).

#### 6.6.2.3.3. **White-tailed sea eagle in Norway and Sweden**

Norway is a stronghold for the white-tailed sea eagle (*Haliaeetus albicilla*), with a stable or increasing population of 1500 pairs, which is 40% of the European breeding population. It is distributed along the Norwegian coast north of 59°N to the Russian border. It is one of the bird species in Norway that has the highest OC levels (Nygård 1991). Temporal trends since 1974 and spatial trends of OCs have been examined (Nygård and Skaare 1996). Inverse relations were found between latitude and DDE and PCB concentrations in eggs. A surprisingly high maximum level of 4.2 μg/g chlordanes were found, but moderate to low levels of mirex, HCHs, dieldrin, and HCB were recorded (Annex Table 6·A5).

The white-tailed sea eagle has also been studied at Arctic sites in Sweden (66-69°N) at altitudes of 200-1000 m. Most pairs breed below 400 m, but also forage at higher altitudes (Helander 1983). During late winter and spring, prey consisted of 58% mammals by weight, and 33% birds. In summer, fish comprised 43%, birds 34%, and mammals 23%. Pike, ducks, and reindeer carrion were also important foods (Helander 1983). During winter, the birds leave the breeding area and migrate to coastal areas of the Fennoscandian Peninsula.

Studies on concentrations of OCs were carried out on eggs collected during the 1960s and 1970s. Arithmetic mean concentrations of DDE and ΣPCB were 4.6 and 11 μg/g ww, respectively. These levels were 4-7 times lower than those found in the Baltic Sea population (Helander 1983, Helander *et al*. 1982).

PCDD/F and planar PCB (CBs 77, 126, 169, 105, and 118) levels were determined in two Arctic white-tailed sea eagle eggs collected from Sweden in 1989. Concentrations ranged from 180-230 pg TEQ/g lw (7.5-8.4 TEQ/g ww) for PCDD/Fs and from 960-2000 pg TEQ/g lw (50-100 pg TEQ/ g ww) for the PCBs (de Wit *et al*. 1994).

#### 6.6.2.3.4. **Gyrfalcon in Iceland**

Òlafsdóttir *et al*. (1995) examined OC levels in samples from gyrfalcon (*Falco rusticolus*) found dead and collected between 1979 and 1992 in Iceland (Annex Table 6·A5). This species of falcon is basically non-migratory. Levels are slightly higher than Canadian values, however, a long time span is represented, and the sample is no doubt biased toward stressed birds. The study showed a strong correlation of contaminant levels with age. At hatching, ΣDDT levels were approximately 0.1 μg/g, and had increased almost 100-fold in ten months, and 1000-fold at 20 months. DDE constituted 95-99% of the ΣDDT. A linear relationship between the levels of contaminants and age suggests that contaminants originate from the same source, making the possibility of local contamination unlikely. It is likely that migratory prey species are contaminated.

The possibility that nutritional status contributed to the death of these birds was also tested by relating lipid levels in individuals to contaminant levels. Levels of ΣPCB and ΣDDT were 2-3 times higher in lean birds than in birds with more fat. Differences in HCB levels were not observed. It is believed that the mobilization of fat deposits, where most of the organic contaminants are stored, may have contributed to the death of the leaner birds by critically raising the levels of contaminants in vital organs (Òlafsdóttir *et al*. 1995, Walker 1990).

The three *F. rusticolus candicans* individuals, which originate in Greenland, had higher contaminant levels than the more sedentary *F. r. islandus*. Also, the oldest bird in the sample was 28 months, however, gyrfalcon can become nine years old in the wild. Thus, the reported levels are probably considerably lower than in the population on average.

#### 6.6.2.3.5. **Fennoscandian merlin**

The merlin (*Falco columbarius aesalon*) is the most common falcon in the Arctic parts of Fennoscandia, and the preferred breeding areas are found in the subalpine birch forest (Gjershaug *et al*. 1994). The Norwegian population of merlins overwinters from Germany and southwards to the Mediterranean countries. It arrives in the breeding areas in Fennoscandia in April/May at the same time as its main prey, migrating passerines and waders.

In eggs collected in 1988-1993 in Norway (mainly from Alta, northern Norway), the mean concentrations of ΣDDT and ΣPCB were 26 782 ng/g and 1820 ng/g ww, respectively. Low concentrations of other OCs were also found, among them mirex, which was used on a large scale in the USA (Nygård *et al*. 1994).

The ΣPCB levels are low compared to the ΣDDT levels, and according to the authors, probably have no direct effect on the merlin reproduction in Norway at present (Nygård *et al*. 1994).

### 6.6.2.4. **Other carnivores**

#### 6.6.2.4.1. **Mustelids**

American mink (*Mustela vison*) and marten (*Martes americanum*) feed primarily on small mammals and fish species throughout the forested regions of North America. American mink have been introduced to Europe and today are present as a wild species in most Scandinavian countries. Small mammals and fish form the greatest components of mink diet in most areas (Gilbert and Nancekivell 1982, Eagle and Whitman 1987), thus, mink are exposed to contaminants from both terrestrial and aquatic food webs.

Both mink and marten collected from five sites in the western Canadian Arctic between 1991 and 1993 had about ten times higher OC levels than caribou/reindeer when levels were converted to lipid weight values (Annex Table 6·A5). The OC levels were lowest around Inuvik, while levels in most sites farther south were higher, but do not follow a clear pattern. ΣPCB residues (43 congeners) ranged from a mean of 4.94 ng/g ww in the livers of Inuvik mink in 1994 (the most northerly collection site) to 25.8 ng/g in mink from Fort Liard (one of the southerly sites). The reason why ΣPCB levels in the Fort Providence samples were over three times higher (92.5 ng/g) than mink from any other community is unknown. Higher levels of PCBs, ΣDDT, dieldrin, HCB, and ΣCHL were found in mink muscle and liver from northern Quebec (Grand Baleine river basin). Higher PCBs are due in part to quantitation as Aroclor 1254 : 1260 (1 : 1) (Langlois and Langis 1995), but the results are consistent with higher levels of OCs generally observed in terrestrial mammals and waterfowl in the eastern Arctic.

Overall, contaminant levels in western NWT American mink (*Mustela vison*) were low in comparison with wild mink from other areas of North America, where ΣPCB (based on Aroclor equivalent or sum of individual congeners) and ΣDDT were generally 1-2 orders of magnitude higher (summarized in Poole *et al.* 1995). The liver of mink from the Lake Ontario region contained the highest levels of ΣPCB and DDT metabolites.

Replicate liver samples for mink were examined for six PCDD/F congeners (Poole unpubl. data 1996). Only TCDF (one of three samples, 5 pg/g ww) and HxCDD (mean 2 pg/g in two of three samples) were detected.

The European otter (*Lutra lutra*) is present in the Arctic and boreal regions of Scandinavia and along the marine coast in Norway. The prey is predominantly fish, but frogs, insects, crustaceans, birds, and mammals are also consumed. Studies on otters collected in Sweden during the 1970s revealed high concentrations of, particularly, PCB, with decreasing concentrations from south to north. Concentrations in the remote Arctic areas of Sweden were 38 μg/g lw (median) (Olsson *et al.* 1996b), whereas otters from the Norwegian Arctic coast had lower concentrations (17 μg/g lw) (Sandegren *et al.* 1980). Concentrations of ΣDDT were much lower (2.6 μg/g and 1.7 μg/g lw, respectively) despite the fact that the concentrations of ΣDDT were similar or even higher than the concentration of PCB in prey and in fish (Olsson and Reutergårdh 1986). It was suggested as early as the 1980s that PCB might have caused the rapid decline of the otter population in Europe (Sandegren *et al.* 1980, Mason and Macdonald 1986, Olsson *et al.* 1996b, Leonards *et al.* 1995).

Current PCB levels in otter in Arctic Sweden are 7.5 μg/g lw and ΣDDT levels are 0.14 μg/g lw. The PCB levels are somewhat higher than those found in Canadian mink, ermine, and marten. ΣDDT levels are comparable to those found in Canadian mustelids.

PCDD/F and planar PCB (CBs 77, 126, 169, 105, 118) levels were determined in two homogenates of otter muscle collected from the Swedish Arctic (Annex Table 6·A22). Trace amounts of PeCDD, one HxCDD, TeCDF, and one PeCDF were found, as well as high levels of all five planar PCBs. The concentrations ranged from 2.7-4.7 pg TEQ/g lw (0.04-0.21 pg TEQ/g ww) for PCDD/Fs and 100-270 pg TEQ/g lw (1.5-12 pg TEQ/g ww) for planar PCBs (C. de Wit unpubl. data).

### 6.6.2.4.2. **Red fox and wolf**

The food habits of canine carnivores differ greatly depending on geographic area and time of year. Red foxes most typically prey on smaller mammals, but also consume insects and invertebrates. Reptiles, fish, and berries form a major part of the diet in late summer (Banfield 1974). The wolf data reported here are from packs in Canada that follow and hunt caribou. However, small mammals constitute a large fraction of their diet, and they will occasionally consume fish or foxes.

Arctic fox is not included under terrestrial carnivores in this report because the one population studied was part of the marine food web (section 6.6.4.9).

Red fox and wolf samples from Canada contain measurable levels of POPs (Annex Table 6·A5). In red fox muscle and liver from northern Quebec, the dominant OC was PCB (Aroclors, 320-460 ng/g lw) followed by HCB (79-240 ng/g

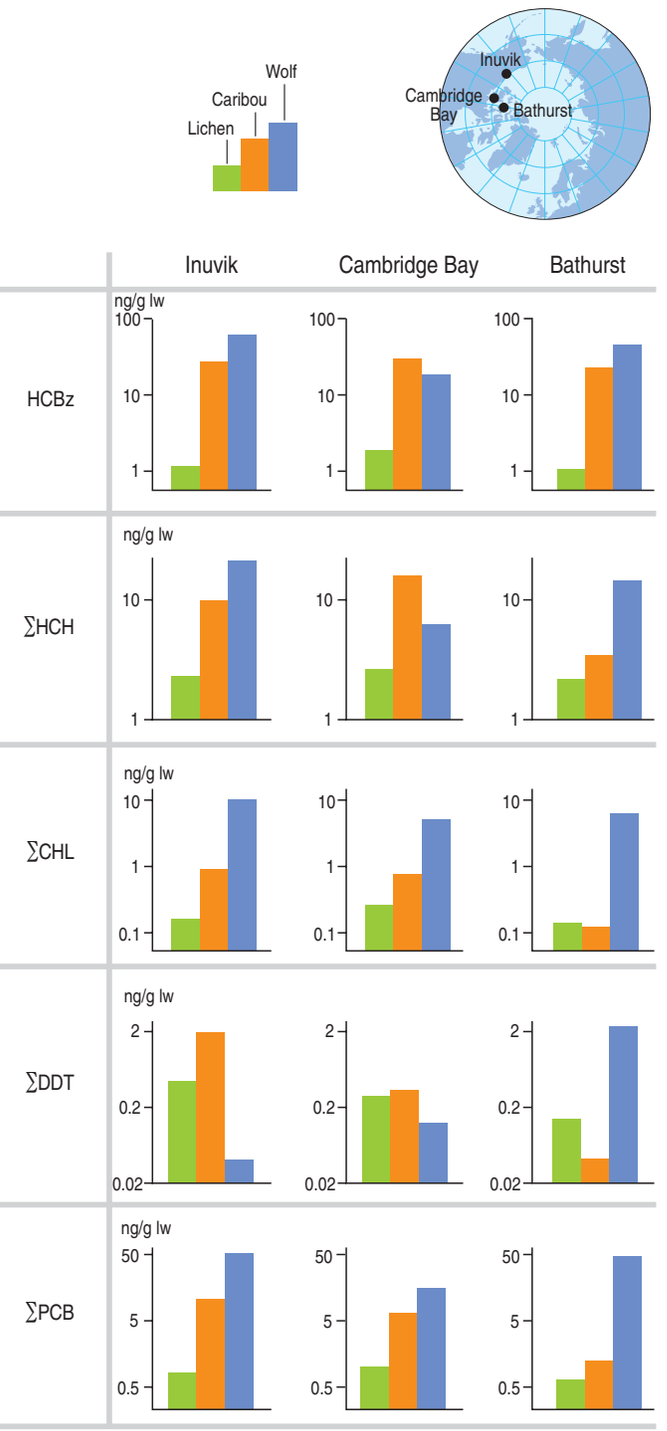



Figure 6·11. Relative tissue concentrations of organochlorine compounds detected in lichen, and in adipose tissue of caribou and wolf at three locations in the Northwest Territories, Canada.

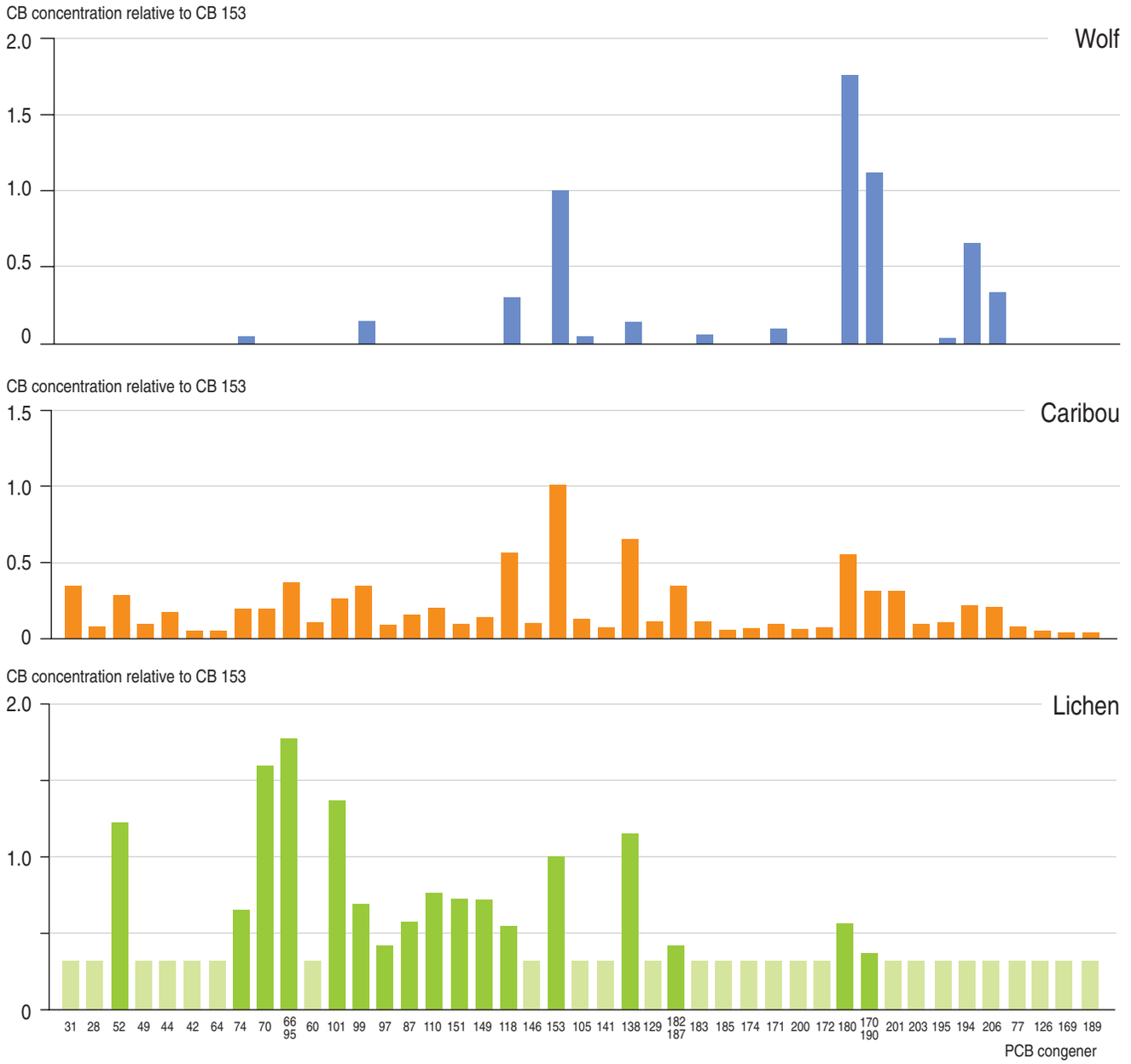



Figure 6·12. Concentrations of PCB congeners relative to CB congener 153 for lichen, caribou, and wolf in the Bathurst herd, Canada. The light bars denote that the congener was not detected.

lw), chlordanes (185-220 ng/g lw), and ΣDDT (n.d.-172 ng/g lw). ΣHCH, dieldrin, and mirex were also found, but in lower concentrations (22-79 ng/g lw). Similarly, the major POPs found in wolf muscle and liver, from three populations in NWT, are ΣPCB (24-113 ng/g lw), HCB (35-92 ng/g lw), and chlordanes (4.5-130 ng/g lw), followed by ΣHCH (9-35 ng/g lw). ΣDDT, dieldrin, and mirex were also found, but in lower concentrations (0.4-13 ng/g lw).

Levels of most persistent OCs were higher in red fox than in wolf. They reflect the spatial trends in OCs already noted for caribou and mink, i.e., higher levels in northern Quebec. Lipid weight OC levels were generally higher from Grand Baleine when compared to samples collected in the Northwest Territories.

### 6.6.2.5. An example of food chain transfer. The lichen → caribou → wolf food chain (Canada)

Elkin (1994) examined the transfer of OCs through the lichen → caribou → wolf food chain in the Bathurst, Bluenose, and Victoria Island caribou herds. Samples were collected with the cooperation of local hunters from Ndilo (Fort Rae), Yellowknife, Inuvik, and Cambridge Bay. For both caribou and wolves, samples were collected from 20 animals. Mean values of each OC at each trophic level are given in Annex Tables 6·A3, 6·A4, and 6·A5. The relative tissue concentrations of the predominant OC contaminants detected in lichen, caribou, and wolf are given in Figure 6·11. Levels of most OCs increased at the higher trophic level. In addition to increasing concentrations with trophic level, the pattern and relative amounts of PCB congeners (relative to CB 153 = 1) changed (Figure 6·12). PCBs in lichen were dominated by lower chlorinated congeners (CB 52, 66/95, 101). These congeners do not biomagnify in caribou or wolf. The PCB pattern in wolves is simplified due to metabolism of almost all congeners found in caribou muscle. Only the most recalcitrant congeners (2,4,5-substituted penta- and hexachloro-, for example, CBs 99, 118, and 153) along with 2,4,5- and 2,3,5- substituted hepta- (CBs 180 and 170) and octachloro- (CBs 195 and 194) remain at the top trophic level. A similar trend has been observed for the fish → seal → polar bear food chain (Muir *et al.* 1988b).

### 6.6.2.6. Summary and conclusions – terrestrial environment

The information available on contaminants in Arctic terrestrial mammals and waterfowl has increased significantly over the past five years. Previous reviews found relatively little data available on contaminant levels in the Arctic terrestrial environment (Thomas *et al*. 1992).

In general, the data available on OCs in the terrestrial environment conforms well to the (mandatory/optional) requirements set out in the original AMAP monitoring plan (AMAP 1993) in terms of media and contaminants that have been monitored, but not in terms of regions studied. Spatial coverage of OC levels in major species is good only for caribou/reindeer (*Rangifer*), where adequate numbers of samples have been analyzed from all major Canadian herds as well as herds in northern Norway, Svalbard, and Sweden. A few reindeer samples from several sites have also been analyzed in Russia.

On a regional basis, there is scattered coverage of waterfowl and game birds within northern Canada and mink populations within the Mackenzie River watershed in NWT. Individual populations of birds of prey have been studied for OC contamination in Sweden, Norway, Iceland, Russia, and Canada, but no single species can be compared on a circumpolar basis. Individual populations of otter in Sweden and Norway have also been studied.

Contamination of soils and humus by OCs has been investigated only in a limited number of regions with the majority of samples originating near military radar facilities.

North-south geographical trends of PCBs in plants have been examined only in Norway. OCs have also been determined in lichen, mosses, Bryophytes, and vascular plants from Finland and Russia, but there is much more limited data from Canada (lichen at three locations) and none from Alaska. Lichen and mosses from Alaska have been analyzed for heavy metals (Ford *et al*. 1995), but results are not yet available for OCs.

PCBs appear to be the most prominent contaminants in the animal species analyzed, especially in mustelids, waterfowl, and birds of prey. In caribou/reindeer (*Rangifer*), HCB and HCH isomers are present at concentrations similar to or greater than total PCB congeners in samples from Canada and Svalbard but PCBs are more prominent in samples from Russia. The relative proportions of OCs in *Rangifer* liver from Russia are not in accord with results from Sweden, Svalbard, or Canada. In the samples from Canada, the pattern of OCs (HCH = HCB > PCB) in *Rangifer* is similar to that in lichen, but this is not the case for the Russian samples. However, detailed comparison of the results from Russia with other regions or with plants is problematic because only single samples of reindeer liver were analyzed from each subregion, there is considerable between-year variation in the results, and percent lipid results were not available.

A significant west to east increase in ΣPCBs, HCB, and ΣHCH was found in caribou from the Canadian Arctic with highest mean levels in Cape Dorset and Lake Harbour herds and lowest in the Inuvik herd. Ten-fold differences in ΣDDT, HCB, ΣHCH, and ΣPCB concentrations between 1994 and 1995 samples from the Russian Arctic make it difficult to infer geographic trends.

A west to east trend of increasing PCDD/F and TEQs levels was also observed in caribou within the Canadian Arctic. TEQs in reindeer fat from the Swedish (Aitejokk, Ammarnäs) and Norwegian (Stilla) Arctic were within the range observed in Canada. Norwegian reindeer sampled near a smelter (Jarfjord) had higher levels. No data on PCDD/Fs are available for *Rangifer* in the Russian Arctic or Svalbard. Most of the TEQs in caribou are due to nPCBs. These results for PCDD/Fs and nPCBs in caribou showed very low levels, which are unlikely to pose a threat to either the caribou sampled in this study or to their human consumers. The levels observed can probably be considered to be background concentrations. TEQs in caribou fat are as low or lower than those reported in fat of domestic animals in Canada (Ryan and Norstrom 1991).

PCBs are the predominant OCs in red fox and wolf samples from Canada. Comparison of OC levels in the lichen → caribou → wolf food chain, from three Canadian herds indicated biomagnification of most OCs and highly selective bioaccumulation of PCB congeners.

Mink studied in NWT, Canada, had higher OC levels than caribou and wolf on a wet weight basis. Mink from the northernmost site in Canada had the lowest OC levels. A north-south trend is also seen in otter in Sweden. The highest OC levels are seen in mink, marten, and ermine from Grand Baleine, Quebec. PCDD/Fs and planar PCBs have also been found in mink from Canada and otter from Sweden.

Higher levels of PCBs and other OCs, particularly mirex, were also found in waterfowl, especially in molluscivores and piscivores, in the eastern compared to the western Canadian Arctic. In the case of birds, however, most overwinter at temperate latitudes and the east-west trends in OCs may, therefore, reflect migratory patterns and winter feeding locations rather than regional contamination differences.

Of the birds of prey for which analytical results are available, the lowest OC levels are found in Icelandic gyrfalcon. This is mainly because they are non-migratory birds, thus, their exposure is primarily of Arctic origin. Migratory species such as merlin, white-tailed sea eagle, and peregrine falcon have much higher ΣDDT and ΣPCB levels than gyrfalcon, reflecting accumulation of OCs at wintering grounds farther south, as well as accumulation in the Arctic from preying on migratory birds. Highest DDE levels were found in Canadian peregrine falcons as well as an increasing trend for heptachlor epoxide and oxychlordane. Highest PCB levels were found in peregrine falcons from the Kola Peninsula, followed by Canada and Fennoscandia, as well as in Norwegian white-tailed sea eagles. There was a significant south-north trend of decreasing OC levels in white-tailed sea eagle eggs with increasing latitude (from 61°30'N to 69°N) along the Norwegian coast.

Within the Canadian Arctic, higher levels of OCs in the east are probably the result of the predominant west to east/northeast atmospheric circulation pattern, which delivers these contaminants from industrialized regions of central and eastern North America to the Arctic via long-range atmospheric transport. The north-south trends seen in Norway and Sweden are probably the result of long-range transport from industrialized parts of Europe, combined with southerly/southwesterly atmospheric circulation patterns.

Significant contamination of soils and vascular plants by PCBs is observed in the immediate vicinity and within a 20 km radius of abandoned and recently active military radar (DEW line) sites in the Canadian Arctic. There is evidence for transfer of PCBs from plants to lemmings at former DEW line radar sites. This raises the possibility that 1) military sites of other circumpolar countries which contained significant amounts of electrical equipment could also have contaminated soils and dump sites and 2) terrestrial mammals and birds could be contaminated because of feeding, even infrequently, on resident plants or animals at these locations.

The work on PCBs in plants and soils near DEW line sites demonstrates how these contaminants can move off site over

time, presumably via volatilization and redeposition or on soil particles, so that concentrations can be elevated (compared to remote background locations) even several kilometers from the site. There was no evidence that large mammals, such as caribou, living in the general area of the DEW line sites had elevated levels of PCBs, however, it should be noted that the studies were not designed specifically to address this question.

Toxaphene was found to be the major OC contaminant in vascular plants (saxifrage) and lichen from the Canadian Arctic. Less persistent OC pesticides (endosulfan, pentachloroanisole, chlorothalonil) were also prominent contaminants. Little is known about circumpolar levels or trends of toxaphene or these less persistent contaminants in lichen/mosses, higher plants, and terrestrial animals.

## 6.6.3. Freshwater environment

### 6.6.3.1. Levels in water

#### 6.6.3.1.1. Organochlorines in river water

Information on contaminant loadings to the Arctic Ocean from northward-flowing rivers is limited. In North America, most of the focus has been on the Mackenzie River and its tributaries, since it is the largest river in the region flowing into the Arctic Ocean (Yunker and Macdonald 1995a, 1995b, Yunker *et al*. 1991, Thomas *et al*. 1986). In Russia, a limited number of OCs have been measured in water and suspended particulate matter (SPM) in major north-flowing rivers (Roshydromet 1995, Rovinsky *et al*. 1995, Melnikov *et al*. 1995). PCBs and γ-HCH were determined in water from nine rivers in Norway flowing into the Norwegian and Barents Seas (Holtan *et al*. 1993, 1994, 1995). There have been no measurements of OCs in water of northward-flowing rivers in Alaska or Iceland.

Jeffries *et al*. (1994, 1996) analyzed Canadian Arctic rivers for OCs over the period 1992-93. Samples were separated into dissolved and suspended solid fractions using continuous-flow centrifugation. HCH isomers were the major group of OCs detected in the Mackenzie River and 11 other north-flowing rivers of the Canadian Arctic (Table 6·10). Concentrations of many OCs were at or below their detection limits and hence the variability between replicates was high. ΣHCH levels (sum α- and γ- isomers) in the Mackenzie River (0.32 ng/L at Arctic Red River) were lower than the means in other northern rivers in Canada flowing to either the Arctic Ocean (1.2 ng/L) or to Hudson Bay (1.1 ng/L). Concentrations of ΣHCH in the Yukon River were similar to those in the Mackenzie River, but lower than in smaller north-flowing rivers (Alaee and Gregor 1994).

Circumpolar concentrations of ΣHCH in other north-flowing rivers are illustrated in Figure 6·13 (next page). In Russia, concentrations of ΣHCH ranged from 1.2 ng/L in River Bulun to 20 ng/L in River Ebitym (Rovinsky *et al*. 1995). Similar concentrations for ΣHCH (range 3 to 17 ng/L in 1992 and 1993 for all Russian rivers except the Ob River) were found in the survey by Roshydromet (1995). These levels are higher than those in Canadian rivers (0.3-1.7 ng/L); the Ob results were exceptionally high (43-59 ng/L) (Figure 6·13, Annex Table 6·A6). Ratios of γ-/α-HCH were also high (0.25-3.9), indicating significant inputs of lindane to the Russian rivers. Surveys of river water in major Norwegian rivers during 1992-1994 found γ-HCH was the major OC contaminant (Holtan *et al*. 1993, 1994, 1995) (Table 6·10). Concentrations of γ-HCH in Norwegian rivers (0.03-0.74 ng/L) were similar to those in Canadian Arctic rivers, but lower than results for Russian rivers. For purposes of comparison with other locations where both α- and γ-HCH were measured, the γ-HCH results from Norwegian rivers were multiplied by 2.

Table 6·10. Concentrations (ng/L) of selected contaminants in Canadian, Norwegian, and Russian rivers flowing to the Arctic and to Hudson Bay.

| | ΣHCH | ΣDDT | ΣPCB | ΣCHL |
|---|---|---|---|---|
| *Canada, rivers flowing to Arctic Ocean* (1993) (Jefferies *et al*. 1994) | | | | |
| Mackenzie, at Arctic Red | 0.32 | 0.10 | 0.64 | 0.01 |
| Mackenzie, east (mean) | 0.45 | 0.25 | 3.04 | 0.05 |
| Mackenzie, west (mean) | 0.35 | 0.00 | 4.83 | 0.07 |
| Andrews | 0.28 | 0.00 | 0.91 | <0.01 |
| Coppermine, above Copper Creek | 0.80 | 0.11 | 1.02 | <0.01 |
| Burnside, at mouth | 1.17 | 0.06 | 0.71 | 0.08 |
| Ellice, at mouth | 1.70 | 0.04 | 0.79 | 0.06 |
| Back, above Hermann River | 1.54 | 0.12 | 0.62 | 0.16 |
| Hayes, above Chantrey In. | 1.66 | 0.11 | 0.54 | 0.20 |
| Takini (tributary of Yukon River) | 0.49 | 0.02 | 0.15 | –[a] |
| Yukon, north of Whitehorse | 1.63 | 0.02 | 0.20 | – |
| *Canada, rivers flowing to Hudson Bay* (1993) (Jefferies *et al*. 1994) | | | | |
| Thelon, below Shultz Lake | 0.90 | 0.04 | 0.71 | 0.08 |
| Dubawnt, at Marjorie Lake outlet | 1.14 | 0.03 | 1.20 | 0.07 |
| Kazan, above Kazan Falls | 1.41 | 0.02 | 0.73 | 0.04 |
| Quoich, above St.Clair Falls | 0.78 | 0.03 | 0.77 | 0.08 |
| Lorillard (flow estimated) | 1.23 | 0.04 | 0.82 | 0.07 |
| *Norway* (1993) (Holtan *et al*. 1994) | | | | |
| Orkla | 0.29[b] | – | – | <0.05 |
| Vefsna | 0.29[b] | – | – | 2.70 |
| Alta | 0.15[b] | – | – | <0.03 |
| *Russia* (1992) (Rovinsky *et al*. 1995) | | | | |
| Lena (delta) | 2.3 | – | 27 | – |
| Bulun (Lena delta area) | 1.2 | – | – | – |
| Ebitym (Lena delta area) | 20 | – | 261 | – |

a. Dash indicates not determined or no data available.
b. Results for γ-HCH only.

ΣPCB and ΣCHL concentrations were also lower in the Mackenzie compared to other rivers flowing to the Arctic Ocean and Hudson Bay, respectively. The much longer residence time of water in the Mackenzie River system allows greater time for volatilization or degradation to take place than in smaller rivers. ΣDDT concentrations were approximately the same in the Mackenzie River (0.10 ng/L at the Arctic Red River) as in the other Canadian rivers flowing to the Arctic Ocean (0.09 ng/L). All rivers flowing to the Arctic Ocean have higher ΣDDT concentrations than those draining to Hudson Bay (0.03 ng/L). The contribution of the suspended solid fraction to the total concentration was <10% for all OC groups. Except for the Mackenzie River, concentrations of suspended particulate matter (SPM) were low, ranging from 1 to 10 mg/L. In the Mackenzie River, SPM was much higher, ranging from 50 to 200 mg/L.

Concentrations of OCs in the Yukon River, which flows into the Bering Sea, have only been measured within Canada (Alaee and Gregor 1994, Alaee 1996). ΣHCH (0.42 ng/L) and toxaphene (0.15 ng/L) were the major OCs in the Yukon River north of the city of Whitehorse. Based on work in Yukon Rivers (Alaee 1996), toxaphene may also have been a major OC contaminant in other Canadian rivers but it was not measured in the survey by Jefferies *et al*. (1994, 1996).

PCB congeners (CB 28, 52, 101, 118, 138, 153, and 180) were below the detection limit (0.03 ng/L) in 1993 and 1994, with the exception of one sample (Vefsna River, 2.7 ng/L) in 1993, in surveys of major Norwegian rivers during 1992-1994 (Holtan *et al*. 1993, 1994, 1995) (Table 6·10).

No assessment of contaminants in Arctic freshwaters would be complete without considering inputs from the Russian sub-continent, the rivers of which contribute more than 68% of total annual riverine input of water to the Arctic Ocean (see chapter 3). The data set on the concentrations of OCs in Russian rivers is very large, but limited in terms of numbers of OC contaminants measured (mainly HCH and

DDT). Although the analytical laboratories involved have participated in interlaboratory comparisons with Canadian and Norwegian laboratories, some results, especially those for PCBs, are difficult to reconcile with other measurements in Canada and Norway (Table 6·10).

Rovinsky *et al.* (1995) reported ΣDDT in the Lena River and nearby rivers (Bulun and Ebitym). A similar range of concentrations for ΣDDT was found in river waters sampled from 1992 to 1994 by Roshydromet (1995). Samples were collected 2-6 times during each year depending on the geochemistry of the basin and the likely use of the chemicals. Low concentrations (< 1-5 ng/L) of *p,p'*-DDT and *p,p'*-DDE were reported in most north-flowing rivers. Highest ΣDDT concentrations were found in the Ob River (mean 5 ng/L). Roshydromet (1995) did not include determination of PCBs. The herbicide 2,4-dichlorophenoxy acetic acid (2,4-D) was detectable in the Ob and Yenisey Rivers, but not elsewhere, at much higher levels than HCH isomers (< 100-65 000 ng/L).

Melnikov and Vlasov (1995) and Melnikov *et al.* (1996a, 1996b) reported relatively high concentrations of PCBs (< 0.1-26.6 μg/g dw) and ΣDDT (0.13-3.89 μg/g dw) in suspended particulate matter from several major Russian rivers including the Ob, Pechora, Yenisey, and Kolyma (Annex Table 6·A7). Highest PCB concentrations (sum of seven congeners) were found in the Pechora (mean 10 μg/g) and lowest in the Ob and Kolyma Rivers (0.5-0.7 μg/g). These concentrations are high when compared to other studies. Yunker and Macdonald (1995a, 1995b) were unable to detect PCBs (< 0.005 μg/g) or DDT (< 0.0001 μg/g) in SPM from the Mackenzie River. Pearson *et al.* (1996) found PCB concentrations averaging 0.14 μg/g dw on particles from Lake Michigan. Pham *et al.* (1993) found ΣDDT concentrations on SPM ranging from 0.008 to 0.382 μg/g in the St. Lawrence River which drains the Great Lakes and industrialized areas of Quebec. Thus, the data for PCBs on SPM in Russian rivers can either be considered inconsistent with other measurements, reflecting sample contamination problems, or are exceptionally high, even when compared with surface waters of urban areas of North America and western Europe. In either case, further investigation of the extent of PCB and DDT contamination of these rivers is needed.

Melnikov and Vlasov (1995), Melnikov *et al.* (1996a, 1996b), and Skotvold (1996) have measured persistent OCs in bottom surficial sediments of major Russian rivers (Annex Table 6·A8). In the Indigirka River delta and Pechora River, samples were taken independently at approximately the same locations. The Norwegian study found higher levels of PCB and other OCs than did Melnikov *et al.* (1996a, 1996b) at these two locations. These levels are consistent with high ΣDDT and ΣPCB levels, which have been consistently reported in surveys by Roshydromet. Previous work in the late 1980s by Vlasov and Melnikov (1990) found highest concentrations of ΣHCH and ΣDDT along the coastal areas of the Kara, Laptev, and East Siberian Seas in zones influenced by inflow of major rivers (Ob, Yenisey, Pyasina, Khatanga, Lena, Indigirka, and Kolyma Rivers). Concentrations of ΣDDT reached 5 ng/L in the mouth of the Yenisey River in the vicinity of the town of Dikson. Elevated levels of ΣHCH were also found in the Ob and Gydan River mouths (4-4.5 ng/L). The concentrations of ΣDDT and ΣHCH were about ten times and three times higher, respectively, than average concentrations in the Kara Sea. The older results are consistent with more recent observations for river water from Roshydromet (1995).

### 6.6.3.1.2. Organochlorines in lake water

Knowledge of levels of persistent OCs in lake waters in the circumpolar Arctic is limited. The largest data set comes from the work of Alaee and Gregor (1994) and Alaee (1996) who compared OC levels in water and suspended solids from five lakes in the Yukon River system. Amituk Lake (Cornwallis Island) in the central Canadian Archipelago and Peter Lake near the west coast of Hudson Bay have also been studied. Samples consisted of large volume water samples (20-100 L). Samples were generally filtered or centrifuged to separate 'dissolved' and suspended phases and the filtrate/centrifugate extracted with solvent or via XAD resin columns to isolate the OCs. Results are also available for samples (1 L) collected from two lakes on the Taimyr Peninsula (Melnikov and Vlasov 1995).

HCH isomers were the most abundant OCs in lake waters at all Canadian locations (Table 6·11). Concentrations of α-HCH (0.34-0.88 ng/L) and γ-HCH (0.07-0.17 ng/L) in the Yukon lakes in 1993 were lower than found in Amituk Lake by Falconer *et al.* (1995a) (1.3 ng/L for α- and 0.28 ng/L for γ-HCH) or in Peter Lake in 1994 (α-HCH = 0.67, γ-HCH = 0.19 ng/L). ΣHCH concentrations in deep waters of two lakes on the Taimyr Peninsula were similar to those in Amituk Lake but γ-/α-HCH ratios (0.48-0.64) were high-

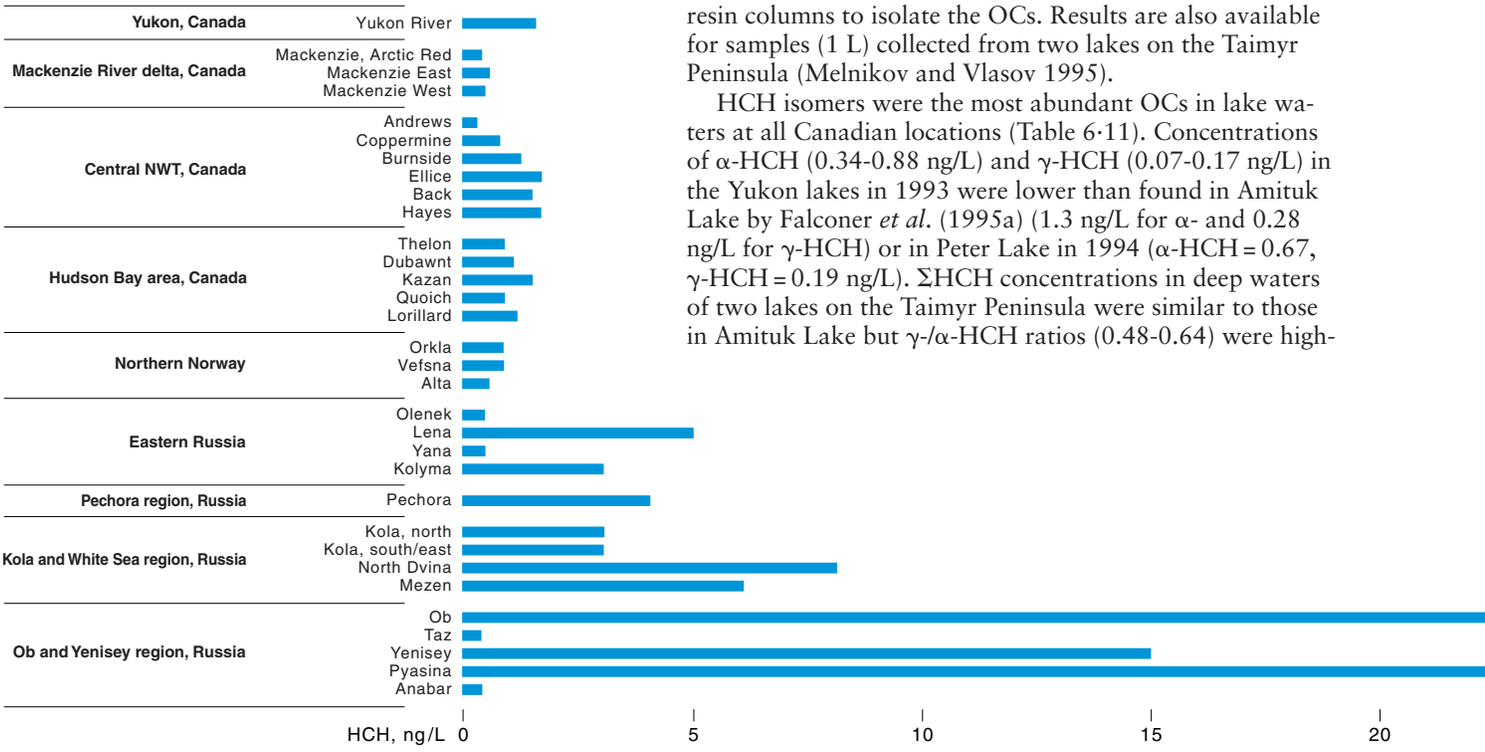



Figure 6·13. Average concentrations (ng/L) of ΣHCH in Arctic rivers in 1993.

Table 6·11. Average (dissolved) concentrations (ng/L) of major OCs in Arctic lake waters.

| Location | Latitude/longiude | α-HCH | γ-HCH | ΣDDT | Toxaphene | ΣPCB | ΣCHL |
|---|---|---|---|---|---|---|---|
| *Canada*[a] | | | | | | | |
| Atlin Lake[b], Yukon | 60°00'N, 133°50'W | 0.39 | 0.2 | 0.01 | 0.27 | 0.018 | – |
| Tagish Lake[b], Yukon | 60°10'N, 134°20'W | 0.39 | 0.15 | <0.005 | 0.02 | 0.116 | – |
| Bennett Lake[b], Yukon | 60°06'N, 134°52'W | 0.34 | 0.11 | 0.02 | 0.11 | 0.206 | – |
| Marsh Lake[b], Yukon | 60°25'N, 134°18'W | 0.48 | 0.2 | 0.005 | 0.18 | 0.147 | – |
| Kusawa Lake[b], Yukon | 60°20'N, 136°22'W | 0.42 | 0.07 | 0.02 | 0.20 | 0.219 | – |
| Fox Lake[b], Yukon | 61°14'N, 135°28'W | 0.39 | 0.15 | 0.09 | 0.11 | 0.090 | – |
| Lake Laberge[b], Yukon | 61°11'N, 135°12'W | 0.88 | 0.17 | 0.06 | 0.21 | 0.100 | – |
| Amituk Lake[c], NWT | 75°03'N, 93°46'W | 1.2-1.4 | 0.23-0.34 | 0.004-0.04 | 0.15 | 0.43-0.47 | 0.4-0.12 |
| Peter Lake[d], NWT | 63°00'N, 92°00'W | 0.67±0.05 | 0.19±0.01 | 0.13±0.06 | 0.34±0.12 | 0.70±0.08 | 0.05±0.01 |
| *Russia (Taimyr Peninsula)*[e] | | | | | | | |
| Levinson-Lessing Lake (95 m) | 74°28'N, 98°38'E | 1.51 | 0.96 | 2.52 | – | 4.06 | 0.30 |
| Taimyr Lake (18 m) | 74°32'N, 101°43'E | 1.09 | 0.53 | 0.16 | – | 0.67 | 0.14 |

a. All samples are from surface waters (0-1 m depth). All are filtered (≤0.5 µm) except those from Peter Lake.
b. Alaee (1996).
c. Results from Falconer *et al.* (1995a) and Semkin (1996).
d. Muir *et al.* 1995b; n = 4 unfiltered water; mean ± SD.
e. Results from Melnikov *et al.* (1995). Unfiltered water samples analyzed by capillary GC-ECD analysis.

er than in most Canadian Arctic lakes (0.17-0.51). Higher concentrations of HCH and lower γ-/α-HCH ratios have generally been found in the Great Lakes. Strachan *et al.* (1995) reported α-HCH of 1-2 ng/L and γ-HCH of 0.5-0.75 ng/L in water samples from Lake Ontario. McConnell *et al.* (1993) found concentrations of α-HCH ranging from 0.98-1.40 ng/L and γ-HCH from 0.31-0.45 ng/L in surface waters of the Great Lakes in 1990. γ-/α-HCH ratios averaged 0.33 in Great Lakes surface waters.

Evidence for the selective breakdown of HCH in Arctic lake waters was found in water in Amituk Lake on Cornwallis Island (Falconer *et al.* 1995a, 1995b). Optically-active pesticides like α-HCH are produced as racemates, which contain equal proportions of right- and left-handed optical isomers (enantiomers). In the environment, these enantiomers are broken down at different rates by enzymatic activity. Preferential metabolism of either (+) or (–)α-HCH has been found among different species of birds, marine mammals, and terrestrial mammals (Hummert *et al.* 1995, Hühnerfuss 1994, Müller *et al.* 1992, Mössner *et al.* 1992), and in water from different regions of the North Sea (Faller *et al.* 1991). These changes are assumed to be caused by microbial activity, since abiotic mechanisms such as hydrolysis and photolysis are not enantioselective. Selective degradation is expressed by the enantiomer ratio, ER = (+)α-HCH/(–)α-HCH. ERs in Amituk lake waters ranged from 0.77 to 0.93 indicating some biotransformation of the (+)α-HCH.

The less persistent OC pesticides, endosulfan, methoxychlor, and pentachloroanisole, are detectable in lake waters in the Arctic. Muir *et al.* (1995b) found all three compounds present at similar, low concentrations (0.017-0.023 ng/L) in water from Peter Lake, a large oligotrophic lake near Rankin Inlet (NWT). The presence of these compounds is consistent with their presence in Arctic air (Barrie *et al.* 1997). Concentrations were low relative to another current use OC, γ-HCH (0.14 ng/L).

Toxaphene concentrations in Yukon lake waters ranged from 0.02 to 0.27 ng/L. These levels were similar to those observed by Falconer *et al.* (1995a) for Amituk Lake (0.15 ng/L), but much lower than found by Muir *et al.* (1995b) for Peter Lake (0.34 ng/L) near Hudson Bay (Table 6·11). Toxaphene was also detectable in SPM from Lake Laberge at 9 ng/g dw.

A wide range of PCB concentrations is found in Canadian lake waters (0.05-0.7 ng/L), with lowest concentrations in headwater lakes of the Yukon River. A similar range of ΣPCB concentrations is observed in the Great Lakes. Jeremiason *et al.* (1994) found average concentrations of 0.18 ng/L for ΣPCB in Lake Superior (1992) while Pearson *et al.* (1996) reported average ΣPCB levels in the open waters of Lake Michigan (in 1991) of 0.47 ng/L.

The similarities among toxaphene concentrations and patterns in the Yukon River basin lakes suggest that the main contaminant source is atmospheric. Similar HCH concentrations in the lakes and rivers also indicate that there are no major local sources to the lakes. On the other hand, ΣDDT concentrations were three times higher in Lake Laberge than other lakes, suggesting continuing inputs possibly from past local use. Compared to the Canadian lakes, very high levels of ΣDDT were found in the two lakes on the Taimyr Peninsula (Table 6·11), especially in Levinson-Lessing Lake. Whether these DDT levels are accurate or due to contamination problems during sampling deserves further investigation.

### 6.6.3.2. **Freshwater surface sediments**

The presence of PCBs, persistent OC pesticides, PAHs, and other POPs in Arctic and subarctic freshwater and terrestrial environments in North America and Europe has been well documented in recent years (Gubala *et al.* 1995, Muir *et al.* 1995b, Skotvold 1996, Vartiainen *et al.* 1997). Although there have been significant local sources, such as military radar sites (Bright *et al.* 1995a, 1995b, 1995c, Reimer *et al.* 1991) (section 6.2.4.1) and use of DDT and HCH for insect control, the predominant pathway for persistent, semi-volatile POP inputs to Arctic lakes and ponds is thought to be via atmospheric transport and deposition from sources in temperate, industrialized regions.

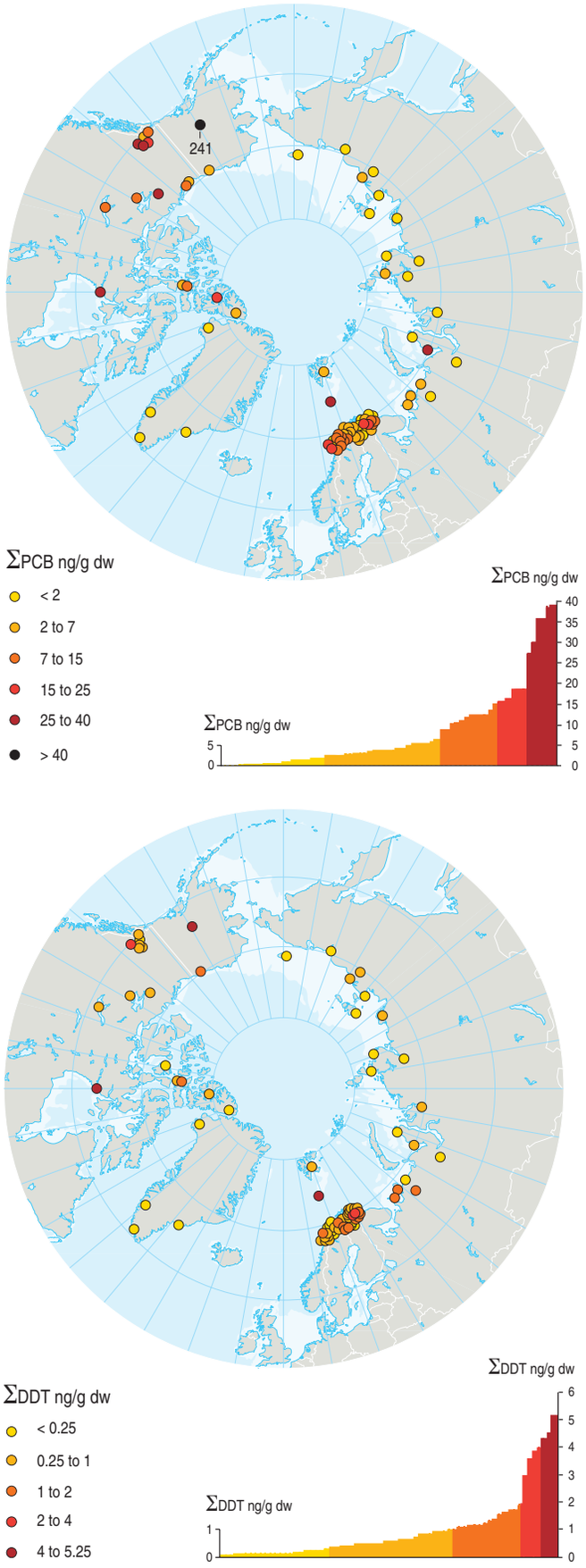



Figure 6·14. Concentrations of ΣPCB and ΣDDT in freshwater surface sediments (from grab samples or from top slices of cores) on a circumpolar basis, and the ranked distribution of values. Most samples were collected from remote lakes with little or no human activity. ΣPCB represents the sum of 13 to 60 congeners, but all studies determined a common suite of 10 to 12 (CBs 18, 28, 52, 66/95, 101, 105, 110, 118, 138, 153/132, 170, 180). ΣDDT = sum of *p,p'*-DDE, -DDT, -DDD, and *o,p'*-DDT.

6.6.3.2.1. **Spatial trends of organochlorines**

PCBs, DDT, HCH, and HCB were detectable at low concentrations (0.01-40 ng/g dw) in almost all samples of freshwater surface sediments from Alaska, northern Canada, Greenland, Norway, Finland, and Russia (Annex Table 6·A8). The majority of these samples were from lakes and ponds, although a few were from Russian rivers. The samples consist of both surface grab samples (using dredge type devices) or surface slices of sediment cores (10-15 cm in diameter). The majority of samples were collected from remote locations with no obvious industrial sources. Some were also collected near or downstream from urban areas, particularly in the Russian Arctic. Samples were not size-fractionated prior to analysis.

Circumpolar trends of ΣPCB and ΣDDT are shown in Figure 6·14. These data are taken from Gubala *et al.* (1995), Muir *et al.* (1995a, 1996b), Vlasov and Melnikov (1995), Cleeman *et al.* (1996a, 1996b), Vartiainen *et al.* (1997), and Skotvold (1996). Different numbers of PCB congeners were determined in each study: seven (Melnikov and Vlasov 1995), 13 (Cleeman *et al.* 1996a, 1996b), 26 (Skotvold 1996), and 60 (Muir *et al.* 1996b) congeners. However, most of the results for ΣPCB are comparable because a common suite of about 12 congeners (CB 18, 28, 52, 66/95, 101, 105, 110, 118, 138, 153/132, 170, and 180) were determined in most studies and they were found to represent greater than 75% of total PCBs (Muir *et al.* 1996b, Skotvold 1996). The sum of seven congeners determined in the Russian monitoring program represented from 11 to 65% of PCBs determined in the Canadian program. Concentrations of ΣPCB and ΣDDT were not correlated with percent organic carbon of the sediments ($p > 0.10$).

Lowest concentrations of PCBs and ΣDDT (both less than 0.1 ng/g dw) were found in surface sediments from four lakes in Greenland (Figure 6·14). Low PCB (< 0.1-10 ng/g) and ΣDDT levels were observed in Russian lakes, especially those east of the Taimyr Peninsula. Highest PCB (10-35 ng/g) and ΣDDT levels were observed in northern Norway, Bear Island (Bjørnøya), at one site (western Yamal) in the western Russian Arctic, and in several Canadian locations in the NWT. The highest reported PCB concentration in Arctic lake sediments (240 ng/g) was in Wonder Lake (central Alaska). This latter site had disproportionately high levels of PCBs relative to ΣDDT, which may indicate local contamination.

Other OCs detected in most freshwater sediments were HCH isomers, HCB, and *cis*- and *trans*-chlordane. Highest ΣHCH results (3-15 ng/g dw) occurred in Russian lakes/ponds (68-70°N, 45-67°E), in Finnmark, Norway, and in Alaska. No circumpolar trend for HCH was evident. It is possible that local or regional use of lindane for biting fly control might explain the elevated levels.

Variations among lakes in sedimentation rates, which may be influenced by lake morphometry including watershed area, steepness of the watershed, and water column depth, as well as phytoplankton biomass (Blais and Kalff 1995) probably have a major influence on the observed concentrations in these freshwater sediments. In this respect, dated sediment cores corrected for focusing, are far superior for assessment of spatial trends.

There is no obvious variation of major OCs reported in Annex Table 6·A8 in freshwater surface sediments with longitude or latitude. However, when results from mid-latitude lakes in Canada are included, concentrations of ΣDDT in surface sediments declined significantly with latitude from 9.7 ng/g (dw) in sediments from two lakes near 49°44'N, 93°45'W in northern Ontario (L382, L375) to 0.10 ng/g in

Lake Hazen at 81°45'N, 71°30'W on Ellesmere Island. In the same lakes, the proportion of di-/trichloro- congeners of ΣPCB increased significantly with latitude, while total levels of octachlorobiphenyls declined (Muir *et al.* 1996b). The lower chlorinated congeners, especially CB 28 and CB 52, were also the most abundant congeners, accounting for 20-30% of the 24 PCB congeners determined in lake sediments from northern Norway (Skotvold 1996).

With the exception of Wonder Lake, the results show that surface sediment samples from remote Arctic lakes have similar or lower concentrations of ΣPCBs and ΣDDT than those reported for mid-latitude lakes in North America and Europe (Swackhamer *et al.* 1988, Sanders *et al.* 1992, 1993, Muir *et al.* 1995a, 1996b), and far lower levels than lakes or reservoirs near industrialized areas in North America and western Europe (Eisenreich *et al.* 1989, Järnberg *et al.* 1993). Jeremiason *et al.* (1994) reported ΣPCB concentrations ranging from 7.8 to 17.6 ng/g dw in surface slices of six cores collected from Lake Superior in the 1980s. ΣPCB concentrations in surface slices of cores from four isolated Wisconsin lakes (2.6-89 ng/g dw) (Swackhamer *et al.* 1988), Siskiwit Lake (Isle Royale, Lake Superior; 48 ng/g) (Swackhamer *et al.* 1988), and four central Ontario lakes (12-54 ng/g) (Macdonald and Metcalfe 1991) were similar or slightly higher than found in Arctic lake sediments. Concentrations of other persistent OCs in freshwater sediments were generally higher than found in Arctic marine sediments by up to ten times (see section 6.6.4.2).

#### 6.6.3.2.1.1. Latitudinal trends of PCB fluxes in lake sediments

The majority of the freshwater data for OCs are from surface sediment samples (Annex Table 6·A8), however, sediment cores have been analyzed from Alaska, northern Canada, and northern Finland. Muir *et al.* (1995a, 1996b) collected sediment cores from eleven remote lakes in Canada between 49°N and 82°N and Gubala *et al.* (1995) analyzed two cores from Alaskan lakes. Sediment cores were also collected in Great Slave Lake (Evans *et al.* 1996) and in three lakes in northern Finland (Vartiainen *et al.* 1997). In all cases, sediment slices were dated using excess $^{210}$Pb and $^{137}$Cs and sedimentation rates determined from the decline of the radionuclide signal with depth using constant-rate-of-supply models (Robbins 1978). PCB fluxes (ng/m$^2$/y) were calculated by multiplying the sedimentation rates by the PCB concentration in surface slices and the inventories (ng/m$^2$) were calculated by summing annual fluxes. Sediment focusing factors, used to correct results to an average aereal deposition for the entire lake, were calculated as ratios of the depositional $^{210}$Pb fluxes or total sediment inventories of radionuclides to the regional atmospheric $^{210}$Pb fluxes, or decay-corrected regional radionuclide inventories.

The latitudinal trend of PCB fluxes is shown in Figure 6·15 using results from 25 sites (focus-corrected except for cores from Finland). Omitting results for Schrader Lake, a significant (six-fold) decline of ΣPCB flux with increasing north latitude over the 35° latitude range (46-81°N) was observed ($r^2 = 0.45$, $n = 24$, $p < 0.001$). This decline is mainly due to declines in penta- to octachlorobiphenyls because no significant decline in di-/trichloro-biphenyl fluxes was found for 11 Canadian mid-latitude and Arctic cores (Muir *et al.* 1996b). Fluxes of ΣPCB for six Lake Superior cores (841-1670 ng/m$^2$/y) (Jeremiason *et al.* 1994) were also in good agreement with the exponential decline. Results for Lake Ontario illustrate differences between a location receiving substantial riverine inputs compared with Arctic and mid-latitude sites receiving PCBs solely via atmospheric deposition. The results for Finland and Alaska are in general agreement with the approximately six-fold exponential decline observed for the Canadian data (Muir *et al.* 1996b). The predicted fluxes in the High Arctic agree well with observed PCB flux in a snow core from the Agassiz ice cap on northern Ellesmere Island (Gregor *et al.* 1995).

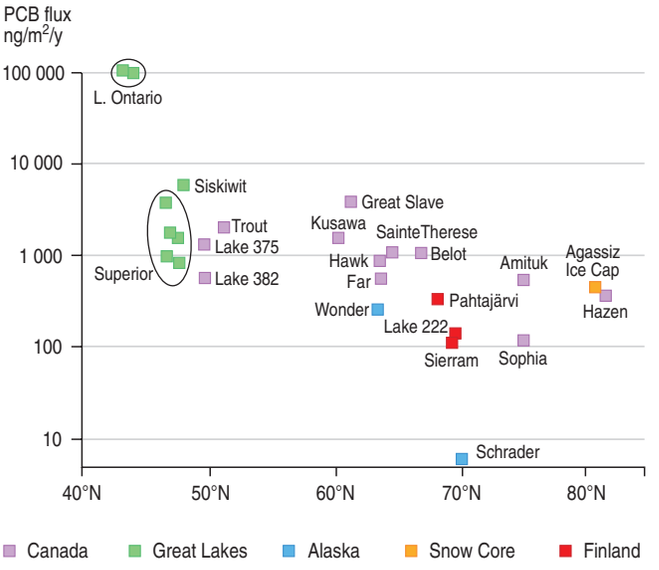



Figure 6·15. Latitudinal fluxes of ΣPCB in dated sediment cores from 25 locations including 13 Arctic locations (in Canada, Finland, and Alaska). All fluxes (except those from Finland) are corrected for focusing of fine particles in deep zones of the lake. There is a significant relationship ($r^2 = 0.45$) between latitude and PCB flux (omitting results for Schrader Lake and Lake Ontario).

#### 6.6.3.2.1.2. Concentrations and patterns of PCDD/F congeners in lake and river sediments

PCDD/Fs have been determined in a limited number of lake sediment cores in Canada and Finland, and in surface sediments from Arctic lakes in Norway and Sweden (Annex Table 6·A21). PCDD/F profiles with depth in sediment cores are discussed in more detail in section 6.7.3.1. Vartiainen *et al.* (1996) determined PCDD/F profiles in three lakes in northern Finland and found low TCDD TEQ concentrations ranging from 1.4 to 4.2 pg/g dw in the surface slices. PCDD/F TEQs in lakes Storvindeln and Akusjärvi in northern Sweden were very similar, at 3.0 and 4.1 pg/g dw, respectively (C. de Wit unpubl. data).

Schlabach and Skotvold (1996a, 1996b) studied the levels and spatial distribution of PCDD/Fs in lake sediments collected near the Syd-Varanger smelter works in Kirkenes, in Arctic Norway (section 6.2.4.3.1). TCDD TEQs in sediment from Hauksjøen, the reference lake (25 km from the smelter), were similar to the Swedish and Finnish lakes. The PCDD/F congener profile in Arctic lakes of Sweden, Finland, and Norway was dominated by OCDD. Levels of 2,3,7,8-TCDD/Fs were low or non-detectable in most of the sediment samples, however, other non-2,3,7,8-substituted tetra-CDD/F congeners were present.

Evans *et al.* (1996) determined PCDD/Fs in two cores from Great Slave Lake, as well as in surface grab samples from the same lake, and found TEQs in surface slices ranging from < 0.01 to ≈ 0.2 pg/g. The predominant PCDD/F congeners in Great Slave Lake surface sediments were OCDD (< 2-14 pg/g) and di- and trichloro-dioxin and -furan congeners (2,7/2,8-DiCDD, 2,8-DiCDF, 2,3,8-TriCDF). The toxic tetra-, penta-, and hexachloro- CDD/F congeners were

near or at detection limits in most surface sediments of Great Slave Lake (< 0.2-0.9 pg/g dw). This study was the only one to determine di- and trichloro- dioxins/furans in Arctic sediments. The presence of 2,7/2,8-DiCDD, 2,8-DiCDF, 2,3,8-TriCDF, and 2,3,7,8-TCDF, along with pentachloroanisole and chlorinated veratroles, in surface sediments suggests that bleached kraft pulp mills in the Peace-Athabasca-Slave River basin, which contributes much of the flow into the lake, are also important sources. The 2,7/2,8-DiCDD, 2,8-DiCDF, 2,3,8-TriCDF, and 2,3,7,8-TCDF congeners also predominated in bleached kraft mill effluent in the region (Pastershank and Muir 1995).

Concentrations of most PCDD/F congeners in surface sediments from the lakes studied in Finland, Norway, and Sweden were generally higher than in Great Slave Lake, by up to ten times. This may be due to much lower sedimentation rates (30-90 g/m$^2$/y) in these small lakes than in Great Slave Lake (550-690 g/m$^2$/y). OCDD was the predominant congener in all three lakes (28-76 pg/g dw), along with 1,2,3,4,6,7,8-HpCDF, but many non-2,3,7,8-PCDD/Fs were also present, representing 15-25% of total ($Cl_4$ - $Cl_8$) PCDD/Fs. Atmospheric deposition of combustion-related PCDD/Fs appears to be the only logical source of contamination of these lakes which are situated in uninhabited areas with no road access or where no local sources exist. Concentrations of PCDD/Fs in the Finnish lakes were similar to 'background' levels in some other areas of Europe. For example, PCDD/F concentrations were 5.3 and 3.3 pg/g dw in two cores from the northern part of Lake Ladoga (Särkkä *et al.* 1993). Much higher concentrations of PCDD/Fs, i.e., greater than 1000 pg/g total PCDD/Fs, are found in sediments near industrialized areas in Europe, in Newark Bay, USA and its tributaries (Ehrlich *et al.* 1994), and in several locations in the Great Lakes region of North America (Czuczwa *et al.* 1984, Czuczwa and Hites 1984, 1986, Smith *et al.* 1993b).

#### 6.6.3.2.1.3. PCDD/Fs in the Severnaya-Dvina River near Arkhangelsk, Russia

A study of PCDD/Fs in sediment of the Severnaya Dvina River in western Russia, which flows into the White Sea at the city of Arkhangelsk, was conducted by the Russian Ministry of Nature Protection and Natural Resources. This was part of a larger study to investigate the extent of contamination from chlorine-bleached kraft pulp and paper mills (Yufit and Khotuleva 1994). Analyses of PCDD/Fs were conducted by both the Institute of Evolution Morphology and Ecology of Animals (IEMEA) and the Bavarian Institute of Water. PCDD/F levels as TCDD TEQs in Dvina River sediments ranged from 0.08 to 0.12 pg/g dw at 'background' sites and from 0.30-0.60 pg/g dw in sediments collected within 2 km of chlorine-bleach pulp and paper mills at Syktyvkar and Kotlas. These TEQs were lower than reported in sediments from the Baltic Sea (Kjeller and Rappe 1995), but similar to levels found in Barents Sea sediments (Oehme *et al.* 1993). The PCDD/F congener pattern observed in Severnaya Dvina River sediments was typical of chlorine-bleach pulp and paper mills combined with chlorophenol sources. TCDD TEQs were mainly due to the contribution of 2,3,7,8-TCDD and -TCDF. OCDD, HpCDD/F, and HxCDFs were the predominant PCDD/F congeners. OCDD levels ranged from 2.4- 4.2 pg/g at 'background' locations. These levels are low compared to lakes in Norway, Sweden, and Finland (Annex Table 6·A21). Similar levels of OCDD were found in the western basin of Great Slave Lake which is more than 1000 km downstream of bleached kraft pulp mills in Canada.

#### 6.6.3.2.2. Modeling OCs in Arctic lakes

Preliminary chemical fate modeling of inputs and outputs of OCs has been conducted in Arctic lakes in Canada (Diamond 1994, Freitas 1994, Diamond *et al.* 1996). A mass balance model was developed, which accounted for inputs via stream inflow, gas exchange, and dryfall, and contaminant output via stream outflow, burial in sediments, and volatilization. Satisfactory agreement was obtained between predicted and measured concentrations of HCH isomers and other OCs following model calibration with field measurements at Amituk Lake (Cornwallis Island). A major adjustment to the model was the use of empirical rather than theoretical adsorption partition coefficients for sedimenting particles because of very low organic carbon. However, the modeling has raised questions about the fate of hydrophobic organics in Arctic lakes. It has shown that only a small fraction of PCBs and other particle-reactive OCs are sequestered in sediments, with the bulk of the snowmelt inputs passing through the lake to the outflow during the brief and rapid summer melt. Yet fluxes of OCs in sediment cores agree well with estimates of inputs from snow deposition monitoring.

### 6.6.3.3. Freshwater fish and invertebrates

Levels of OCs in freshwater fish from Canada, USA, Greenland, Finland, Norway, Sweden, and Russia are documented in Annex Table 6·A9. Levels from studies monitoring OC levels in fish in undisturbed lakes as well as levels in Arctic lakes with suspected contaminant problems are presented.

PCBs were generally present at 1.5 to 2-fold higher levels than DDT- or chlordane-related compounds. HCHs (sum of α-, β-, and γ-HCH isomers) and chlorobenzenes were present at much lower concentrations than the four major OCs (PCB, DDT, chlordane, toxaphene). Where it has been measured (mainly in samples from Canada and Greenland), toxaphene is a prominent OC present at concentrations similar to or greater than PCBs. Problems with the analysis of toxaphene have already been discussed (section 6.6.1.1). Determination of toxaphene is less accurate than for other OCs because of the lack of analytical standards for individual hepta- to nonachlorobornanes, the major components of toxaphene. Interlaboratory comparisons have shown agreement among participating laboratories within a factor of 2 (Andrews 1994). Highest toxaphene levels are generally seen in fish that are strictly piscivorous, such as lake trout and burbot in Lake Laberge and burbot in the east arm of Great Slave Lake.

PCDD/Fs and nPCBs have been measured in Arctic char, burbot, pike, and lake trout (Annex Table 6·A22) and are discussed in more detail below. Measurements of other planar OCs such as polychlorinated naphthalenes (PCN), chlorinated diphenyl ethers (PCDE) or their brominated analogs (PBDE) in Arctic fish are limited to work in Sweden (Sellström *et al.* 1993, Järnberg *et al.* 1993) and Finland (Koistinen *et al.* 1993). Levels of PCDEs in salmon from northern Finland (Tenojoki river) were about ten times lower than in salmon from southern Finnish lakes and rivers (Koistinen *et al.* 1993). PCDEs accounted for a lower proportion of TEQs than nPCBs and PCDD/Fs in the salmon. PCNs were detected in pike muscle from Lake Storvindeln. The major PCN congener, a hexachloronaphthalene, was present at similar concentrations as the non-*ortho* PCB congeners 77 and 126.

The less persistent OCs, such as methoxychlor, endosulfan, and pentachloro-anisole are frequently detected at low levels (relative to major persistent OCs) in Arctic freshwater fish. Methoxychlor concentrations ranged from < 0.01-0.17

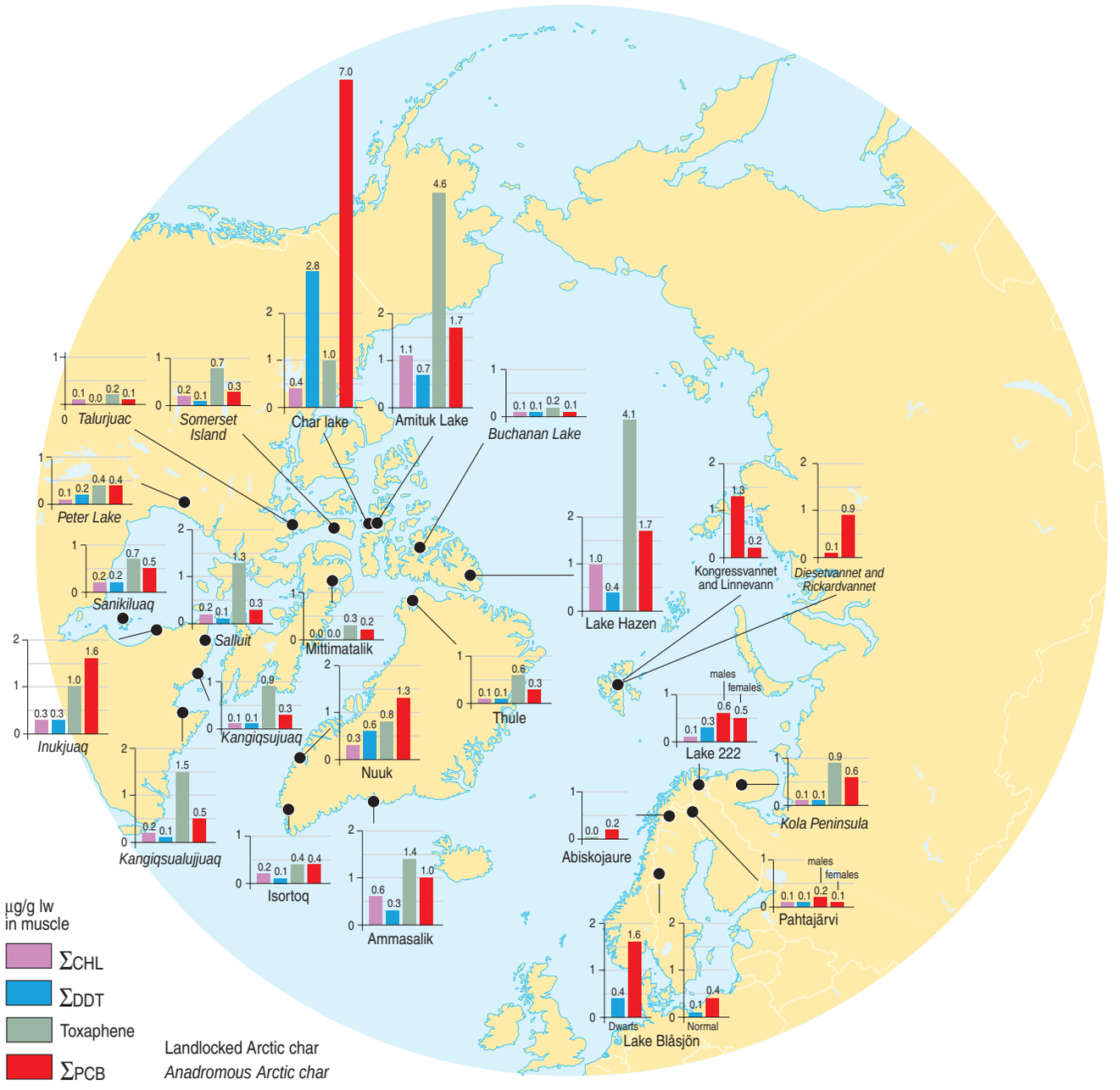



Figure 6·16. OC levels (µg/g lw in muscle) in landlocked and anadromous (italic labels) Arctic char.

ng/g ww in lake trout muscle from lakes in NWT (D. Muir unpubl. data 1996). Pentachloroanisole concentrations in lake trout and Arctic char in Peter Lake (NWT) ranged from < 0.01-0.1 ng/g ww while endosulfan levels ranged from < 0.01 to 1 ng/g ww. Concentrations of these compounds in Canadian Arctic fish were lower than levels reported in surveys of fish in the USA (USEPA 1991). A survey by the US Environmental Protection Agency (USEPA) in the lower 48 states showed methoxychlor concentrations in fish ranging from < 0.1 to 2.5 ng/g in 95% of fish sampled. The USEPA study did not include fish from Alaska.

Assessment of circumpolar trends in concentrations of persistent OCs in fish is difficult for several reasons. Most data for freshwater fish are very recent and have not been fully statistically analyzed for all possible correlations with ancillary data (such as percent lipid, size) or spatial trends. Most data are from studies with a limited number of samples per location, designed to examine regional spatial trends and estimate human dietary exposure. This situation is made more complicated by the observation that levels of many OCs are highly variable, due to differences in the food habits of individual fish and different species, and also in the productivity, food webs, and possibly pathways of contaminant entry in different lakes. Year-to-year variation is large even when sampling is standardized for covariates.

#### 6.6.3.3.1. Circumpolar trends in *Salvelinus* species, Arctic char and lake trout

Arctic char (*Salvelinus alpinus*) are the most widely examined fish from a circumpolar perspective. Char can be found as both landlocked and anadromous populations, and both forms can in fact occur in the same lake. Many populations of Arctic char occur as multimodal size classes with a wide range of ages represented in each size class. There are notable differences even between populations from the same geographic areas. The causes are assumed to be food web differences.

Levels of major OC groups in muscle in both landlocked (Annex Table 6·A9) and anadromous char (Annex Table 6·A17), expressed on a lipid weight basis, are presented in Figure 6·16. Anadromous char are discussed further in section 6.6.4.5.

Landlocked char from Canada, Greenland, Finland, Norway and Sweden have been analyzed for persistent OCs (Annex Table 6·A9). Results for PCBs are the most readily compared among sampling sites although there are minor differences in concentrations due to determination of different numbers of congeners. Mean concentrations of PCBs in landlocked char range from 1 to 290 ng/g ww, although levels at most sites are less than 10 ng/g ww. The lowest levels were found in Finnish Lapland (1-2 ng/g ww), while highest PCB concentrations were found in char from Char Lake, a lake on Cornwallis Island near the Resolute airport, which may have been contaminated by PCBs. Elevated levels of PCBs were also found in char from Amituk Lake in Canada (73 ng/g ww) and from Ammasalik Lake in Greenland (36 ng/g ww). Both are remote lakes with no obvious local sources of contaminants. The variations in levels of PCBs and other OCs are likely related to effects of age or growth rate on bioaccumulation. Hammar *et al.* (1993) found that slow growing char (dwarfs, mean weight 81 g, mean age 7.5 years) had significantly higher levels of ΣPCB and *p,p'*-DDE than faster growing char (normal, mean weight 239 g, mean age 4.9 years) in a Swedish Arctic lake (Annex Table 6·A9, Figure 6·16).

Despite differences in size and feeding behavior between anadromous and stationary populations, OC concentrations are similar in both forms when expressed on a lipid weight basis. This circumpolar comparison (Figure 6·16) demonstrates that toxaphene is the major OC in most char and that generally higher levels of ΣPCBs, toxaphene, and ΣDDT are found in landlocked char in Canada than in Scandinavia.

PCDD/Fs and nPCBs have been measured in landlocked Arctic char in Sweden, Norway, Finland, and Canada (Annex Table 6·A22). In char samples from Norway, Sweden, and Finland, TCDD TEQ concentrations ranged from 0.02 to 0.14 pg TEQ/g ww for PCDD/Fs and from 0.05 to 0.32 pg/g ww for planar PCBs. Higher TEQs (0.14-7.4 pg/g ww) were found in landlocked char from Arctic Canada.

Lake trout are widely distributed predatory fish that feed on forage fish and/or invertebrates. A large data set is available for OCs in muscle of lake trout from northern Canada and Alaska (Annex Table 6·A9). A wide variation in toxaphene and PCB levels is evident in lake trout in NWT and Yukon lakes. Even in lakes of similar size at the same latitude (Peter, Belot, and Colville; Travaillant, Raddi, and Fish) there were large differences in concentrations of toxaphene and, to a lesser extent, of PCBs. Within lakes, age and fish weight were significantly correlated with toxaphene levels in lake trout at several locations (Muir *et al.* 1997). By far the highest concentrations observed are found in muscle of lake trout from Lake Laberge (mean ΣPCB and toxaphene of 448 ng/g ww and 344 ng/g ww, respectively). Mean ΣPCB concentrations in lake trout from other Yukon and NWT lakes were lower, ranging from 10 to 128 ng/g ww. Converting the results to a lipid weight basis to eliminate the effect of different fat content of samples from different locations did not remove the lake-to-lake variation in mean PCB levels. This variation, which is mainly due to food chain length, is discussed in more detail in section 6.6.3.4. Lake trout muscle from the Grand Baleine region of northern Quebec had similar ΣPCB levels (Langlois and Langis 1995) to those from the NWT. Lake trout from five Arctic lakes in Alaska also had similar PCB levels (average 454 ng/g lw) to those in the NWT (Allen-Gil *et al.* 1997). Brown trout (*Salmo trutta*) from Lake 222 in Finland had lower levels of ΣDDT than lake trout, but higher total chlorobenzenes and ΣHCH.

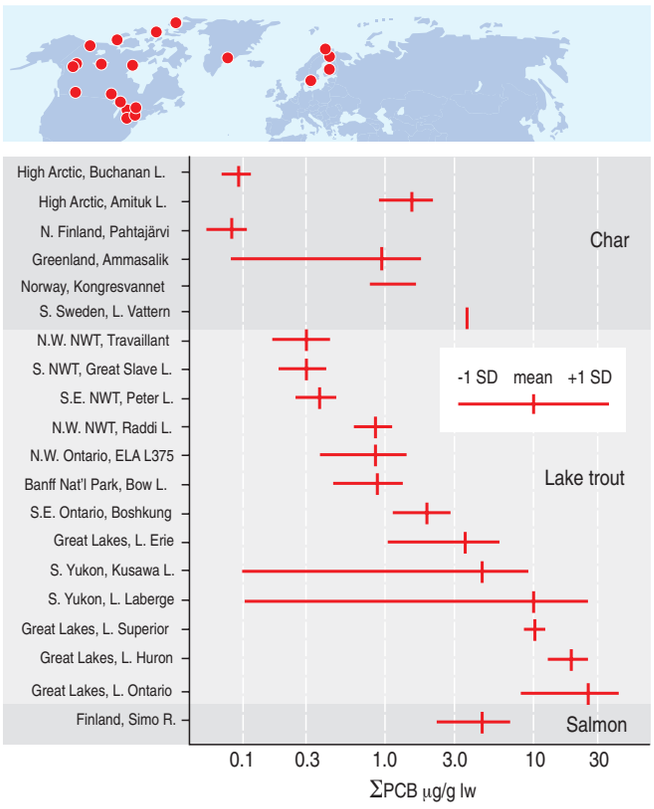



Figure 6·17. Comparison of ∑PCB concentrations (lipid normalized, calculated from geometric means) in Arctic versus temperate zone salmonid fish. See Annex Table 6·A9 for wet weight concentrations.

Levels of PCBs, toxaphene, and other persistent OCs have been measured in other regions of Canada and the USA, especially in the Great Lakes (DeVault *et al.* 1995). Figure 6·17 compares lipid-weight concentrations of PCBs in landlocked Arctic char from northern Canada, Greenland, Norway, Finland, and southern Sweden, in lake trout from the NWT, Yukon, and southern Canada/USA, and in salmon from Finland. Concentrations in Great Lakes lake trout are higher than in all char and most lake trout from the Arctic. An exception is lake trout from Lake Laberge which have similar levels (on both a wet weight and lipid weight basis) to those in Lake Superior. Lake trout from smaller remote lakes in Alberta (Bow L.) and northwestern Ontario (L. 375) have levels similar to Arctic salmonids.

### 6.6.3.3.2. **Burbot (*Lota lota*)**

OCs, including PCDD/Fs, have been measured in burbot liver from Canada, Sweden, Finland, and Russia. The majority of the data is from the Yukon and NWT in Canada where the widespread availability of the species, and consumption of the fatty liver by indigenous people, have made it a priority for monitoring OCs. Burbot is a predatory, bottom-feeding species of fish that inhabits deep lakes and Arctic rivers over much of Canada, Scandinavia, and Russia (Scott and Crossman 1973). Their sedentary nature and the high lipid content of the liver make the species suitable for monitoring lipophilic pollutants (Burgermeister *et al.* 1983). Burbot appear to accumulate similar levels of OCs as other predatory fish (Veith *et al.* 1977, Musial *et al.* 1979, Burgermeister *et al.* 1983).

Relatively high OC concentrations are found in burbot liver in Canada. Mean ΣPCB levels ranged from 27 to 1270 ng/g ww and toxaphene from 40 to 2300 ng/g ww. Highest concentrations were found in piscivorous burbot from large lakes in the Yukon and in Great Slave Lake in NWT. Lower

Table 6·12. Concentrations of major chlorinated hydrocarbon pesticides in burbot liver from remote lakes and rivers in Canada (Muir *et al.* 1990a).

| Compound | Concentration (ng/g lw) ± 95% confidence interval at each location[a] | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | Lake Winnipeg[b] 50°42'N, 96°34'W | ELA Lake 625 49°45'N, 93°48'W | Trout Lake 51°15'N, 93°30'W | South Indian Lake 56°47'N, 98°56'W | Mackenzie River, Fort Simpson 61°52'N, 122°21'W | Mackenzie River, Fort Good Hope 66°15'N, 128°38'W | Mackenzie River, Arctic Red River 67°26'N, 133°44'W | Peel River, Fort McPherson 67°26'N, 134°53'W |
| ΣCBz | 33.9±5.0 | 27.9±7.6* | 39.9±11.1 | 67.5±12.6** | 48.7±30.8 | 44.2±23.3 | 43.6±38.7 | 25.5±24.0 |
| HCB | 29.7±4.5 | 22.2±5.4* | 34.4±8.2 | 66.4±12.1** | 34.1±18.0 | 43 ±22.7 | 42.7±37.7 | 23.8±22.6 |
| ΣHCH | 69.9±18.4 | 72.3±14.8** | 35.9±7.8 | 54.6±11.0 | 27.1±24.4* | 30.3±5.3* | 29.0±17.4* | 18.8±9.8 |
| α-HCH | 53.6±14.9 | 43.2±16.9 | 30.6±6.7 | 38.8±4.3 | 24.5±14.8 | 19.7±4.5 | 24.5±22.0 | 15.3±8.6 |
| ΣCHL | 143 ±51.4 | 373 ±126 | 378 ±101** | 285 ±67.6 | 207 ±106 | 173 ±88.9 | 229±161 | 86.4±79.4* |
| *trans*-nonachlor | 34.3±14.5 | 109 ±57.7** | 92.8±25.4 | 68.3±19.1 | 56.4±25.4 | 56.7±27.1 | 83.5±58.0 | 25.9±23.5* |
| ΣDDT | 622 ±219 | 1490 ±601** | 1029 ±523 | 461 ±132* | 163 ±117 * | 95.2±57.4* | 101 ±67.4* | 50.7±46.1* |
| DDE | 350 ±123 | 1017 ±405** | 946 ±474 | 386 ±112* | 123 ±85.1* | 55.0±32.2* | 47.1±30.0* | 27.1±23.0* |
| Mirex | 10.1±3.0 | 14 ±4.1 | 16.9±7.6 | 17.4±4.5** | 8.6±2.3 | 7.0±3.6 | 5.2±4.2* | 3.7±3.1* |
| Toxaphene | 807 ±285 | 1723 ±541 | 2338 ±769 | 1467 ±324 | 1132 ±684 | 1570 ±1000 | 1700 ±1346 | 931 ±904 |
| Dieldrin | 41.2±13.3 | 60.1±9.8 | 70.8±21.2** | 34.8±8.9* | 14.0±11.6* | 13.5±6.1* | 16.2±11.3* | 7.1±6.3* |
| ΣPCB | 1941 ±682 | 1290 ±386** | 874 ±468 | 944 ±281 | 557 ±389* | 344 ±172* | 301 ±221* | 345 ±284* |
| *Ratios*[c] | | | | | | | | |
| DDE/ΣDDT | 0.57±0.06 | 0.68±0.05* | 0.92±0.02** | 0.84±0.04 | 0.76±0.08* | 0.59±0.10* | 0.47±0.08* | 0.55±0.14* |
| γ-HCH/ΣHCH | 0.16±0.02 | 0.13±0.04 | 0.15±0.01** | 0.14±0.03 | 0.09±0.02* | 0.08±0.03* | 0.11±0.02 | 0.08±0.01* |

a. Geometric means ± 95% confidence limits calculated from log transformed data. Results marked ** are significantly higher (Tukey test at $p < 0.05$) than those marked *. Unmarked results are not significantly different from the highest mean value, but may be significantly higher than the lowest values. Results for Lake Winnipeg were omitted from the means comparisons.
b. South basin of Lake Winnipeg.
c. Mean ratios ± standard deviations.

OC concentrations were found in burbot liver from Finland. A single burbot liver sample from the Yenisey River in Russia had higher ΣDDT and similar ΣPCB (sum of seven congeners) concentrations to burbot from the Mackenzie River. No geographic trends of OCs in burbot liver can be discerned from the NWT and the Yukon data. This is probably due to the confounding effects of fish size and food chain length. The effect of food chain length on OC concentrations in burbot is discussed in more detail in section 6.6.3.4.

Previous work by Muir *et al.* (1990a) examined OC levels in burbot in remote lakes and rivers in Canada, with sample sites ranging from northwestern Ontario to Fort McPherson on the Peel River in the Northwest Territories. Declines in concentrations of PCB congeners, ΣDDT, HCH, dieldrin, and mirex in burbot livers were found with increasing north latitude (Table 6·12). Mean ΣPCB concentrations ranged from 301 ng/g lw in samples from the Mackenzie River at Arctic Red River, NWT to 1290 ng/g lw in a remote lake in northwestern Ontario and 1941 ng/g in Lake Winnipeg, Manitoba. No significant differences in the mean concentrations of toxaphene, α-HCH, and tri- and tetrachlorobiphenyls were observed between southern and northern sampling sites. Toxaphene was the predominant OC residue in the northern fish samples, averaging 1400 ng/g lw at the three most northerly sites and 1723 ng/g lw in northwestern Ontario. The results were consistent with the hypothesis that inputs of semi-volatile OCs decrease with increasing north latitudes and distance from North American sources.

PCDD/F concentrations are relatively high in burbot liver compared to other fish tissues (Annex Table 6·A22). A pooled sample of burbot liver from Pajala, along the river Torneälv in Sweden, had TCDD TEQ concentrations of 3.75 pg/g ww for PCDD/Fs and 18.5 pg/g ww for planar PCBs. TEQ levels in burbot muscle were lower, 0.05 pg/g ww for PCDD/Fs and 0.28 pg TEQ/g ww for planar PCBs, due the low lipid content of muscle. Total TEQs in burbot liver from two lakes in Finland ranged from 5.4 to 8.6 pg/g ww. TCDD TEQs in burbot liver in Canada range from 0.8 pg/g in Great Slave Lake to 166 pg/g in Lake Laberge. In burbot from Scandinavia and Canada, TEQs due to planar PCBs were much greater than those due to PCDD/Fs.

#### 6.6.3.3.3. **Lake whitefish (*Coregonus clupeaformis*)**

A reasonably large data set exists on concentrations of persistent OCs in lake whitefish from Canada, Norway, and Sweden (Annex Table 6·A9). Lake whitefish, a widespread insectivorous fish, is of importance for commercial and subsistence fishing in northern Canada and Scandinavia. ΣPCBs are generally low in lake whitefish compared to piscivorous species such as lake trout. Mean ΣPCB concentrations ranged from 0.54 ng/g ww in Lake Ravdujav'n, Norway, to 280 ng/g ww in Lake Laberge, Yukon. Mean toxaphene and ΣPCB levels were also higher in lake whitefish from Great Slave and Kusawa Lakes in northern Canada, both large lakes with complex food webs compared to other small lakes in the NWT, northern Quebec, and Norway.

Data on PCDD/Fs and planar PCBs in lake whitefish are available for samples from Lake Storvindeln (Sweden) and Great Slave Lake (Canada), and for PCDD/F from Lake Førstevann (Norway) (Annex Table 6·A22). The Norwegian sample was collected in close proximity (1 km) to the Syd-Varanger smelter at Kirkenes. Lake whitefish muscle from Lake Storvindeln and Great Slave Lake had low or non-detectable levels of 2,3,7,8-TCDD, but the nPCBs (CB 77, 126, and 169) were present at low pg/g ww concentrations. Total TEQs in whitefish muscle from Lake Storvindeln and Great Slave Lake were identical, 0.47 pg/g ww. The Norwegian sample was more highly contaminated, with a PCDD/F TEQ of 8.3 pg/g ww.

PBDEs were detected in samples of lake whitefish from Lake Storvindeln in northern Sweden (Sellström *et al.* 1993). 2,2',4,4'-tetrabrominated diphenyl ether (TeBDE) was present at highest levels (15 ng/g lw) while two pentabromo- congeners (a congener of unknown structure and 2,2',4,4',5-PeBDE) were present at levels of 3.39 and 7.2 ng/g lw, respectively. Chlorinated paraffins were also determined in whitefish from Lake Storvindeln, but were not detected (<1 ng/g lw).

#### 6.6.3.3.4. **Invertebrates**

A reasonably large data set on OCs in zooplankton exists for Arctic lakes in Canada (Annex Table 6·A9). No results have been reported for other circumpolar countries. Plankton have proven to be useful biomonitors of PCBs and OC

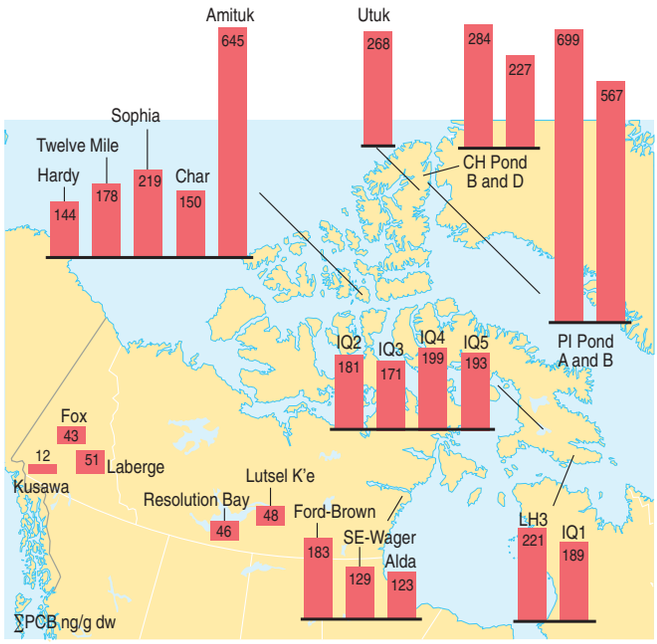



Figure 6·18. Geographic trends of ∑PCBs in zooplankton (>100 µm) in northern Canadian lakes. Data are from Evans (1994), Kidd *et al.* (1995a, 1995b) and Koenig (1996). All concentrations in ng/g dw.

pesticides in lakes (Taylor *et al.* 1991). A study by Koenig (1996) examined geographical and temporal variation of PCBs and OC pesticides in a series of 19 Arctic lakes between 63°N and 79°N. Kidd *et al.* (1995a, 1995b) and Evans (1994) have also determined OCs in zooplankton from Lake Laberge and Great Slave Lake, respectively. Highest PCB concentrations were observed in the northern-most lakes on Cornwallis Island and eastern Ellesmere Island (Figure 6·18). The lowest PCB levels observed by Koenig (1996) were found in samples from the Wager Bay area of northwestern Hudson Bay, but concentrations in zooplankton from Great Slave Lake and Yukon Lakes were even lower. These regional differences could be related in part to variations of zooplankton biomass and food web differences among lakes. Taylor *et al.* (1991) found an inverse relationship between OC concentrations and plankton biomass in 33 Ontario lakes. The results may also reflect differences in concentrations of PCBs in surface waters between the eastern and western Arctic. PCB and toxaphene levels are higher in lake waters from the eastern Arctic than in the Yukon (Table 6·11).

### 6.6.3.4. Examples of food web transfer – freshwater environment

Studies have been conducted to examine whether food web differences may explain elevated levels of PCBs and toxaphene in Lake Laberge and Great Slave Lake, compared to nearby lakes (Kidd *et al.* 1995a, 1995b, Evans 1994, 1996). In the case of Lake Laberge, it was hypothesized that the food web structure of Lake Laberge is different, resulting in elevated concentrations of atmospherically deposited contaminants in the fish. This hypothesis was based on the work of Rasmussen *et al.* (1990) who found that lake trout from Ontario lakes with longer food webs had higher concentrations of PCBs and ΣDDT.

The studies by Kidd *et al.* (1995a, 1995b) and Evans (1994, 1996) used stable isotope ratios to identify the trophic status of various organisms, to demonstrate differences in food web transfer in different lakes. The heavier isotope of nitrogen ($^{15}N$) is enriched 3-5 parts per thousand (‰) in an organism compared to its diet (Peterson and Fry 1987), and is used as an integrated measure of trophic level. This approach is informative because many freshwater Arctic organisms are opportunistic feeders. The use of nitrogen isotope ratios is an excellent method for identifying trophic status since it represents what is assimilated into an organism from its diet and integrates signals from food consumed over the time period of tissue turnover. Slopes from the logarithm (OC) concentration versus $\delta^{15}N$ regressions thus describe biomagnification factors within biological communities.

#### 6.6.3.4.1. Biomagnification in Lake Laberge, Canada

Lake Laberge, downstream from Whitehorse, a major community of 13 500 people on the Yukon River, Canada, was examined because of known contamination problems. The lake has been used for commercial, sport, and native subsistence fisheries for over a century. As described in section 6.6.3.3, analyses of lake trout, burbot, and lake whitefish revealed that levels of PCBs, DDE, and toxaphene were 30 times as high in Lake Laberge as in other Yukon lakes and lakes in the Northwest Territories (Muir *et al.* 1990a, Palmer 1992, Kidd *et al.* 1993, 1995b, Kidd and Schindler 1994). Levels were comparable to those found in the Great Lakes (Oliver and Niimi 1988, Borgmann and Whittle 1991).

There have been numerous hypotheses about the origin of the contamination in Lake Laberge, including contamination due to sewage, past use of toxaphene and DDT, and PCBs in transformer oils, and characteristics of the runoff to the lake (Kidd *et al.* 1995a, 1995b). The most current view is that past activities and local sources are not the cause of present-day contamination of the fish. Local contamination can be discounted because surface sediments contain levels of OCs similar to other regional lakes that do not contain contaminated fish. Long-range atmospheric transport and deposition are sources of toxaphene, HCH, DDT, and HCB to all of the lakes, however, an additional factor in Lake Laberge is that overfishing and increased nutrient input from the Whitehorse sewage lagoon may have changed the food web (Kidd *et al.* 1995b). The lake trout are solely piscivorous, unlike trout populations from other regional lakes, which is reflected in their high lipid content.

Contaminant levels in fish and invertebrates in Lake Laberge and other lakes in the Yukon are given in Annex Table 6·A9. Kidd *et al.* (1995a) examined biomagnification in the food web of Lake Laberge and two reference lakes, Fox and Kusawa, using $\delta^{15}$ nitrogen isotope measurements as an indicator of the trophic position of fish and invertebrates (Figure 6·19). The heavier $^{15}N$ isotope is enriched relative to $^{14}N$ from prey to predator and is expressed as $\delta^{15}N$ ($^{15}N/^{14}N$ stable isotope ratio standardized against air). Lake trout and burbot, mainly carnivorous species, have the highest trophic status and level, while snails and trichopterans (caddis flies), which feed mainly on periphyton and other plant material, occupy the lowest trophic level. There are differences in the food web structures of Laberge, Fox, and Kusawa Lakes (Figure 6·19). The top predators in Lake Laberge feed at a higher trophic level than those from the reference lakes, a factor that has been previously implicated in higher contaminant concentrations in lake trout (Rasmussen *et al.* 1990).

Examination of the relationship between concentrations of ΣHCH, ΣDDT, and toxaphene over the range of trophic levels in Lake Laberge (Figure 6·20) showed that toxaphene and ΣDDT were significantly correlated with trophic level measured by $\delta^{15}N$ (Kidd *et al.* 1995a, 1995b). ΣHCH was also correlated with $\delta^{15}N$, but showed less of an increase with trophic level consistent with its known lower persistence in the environment. Similar relationships were ob-

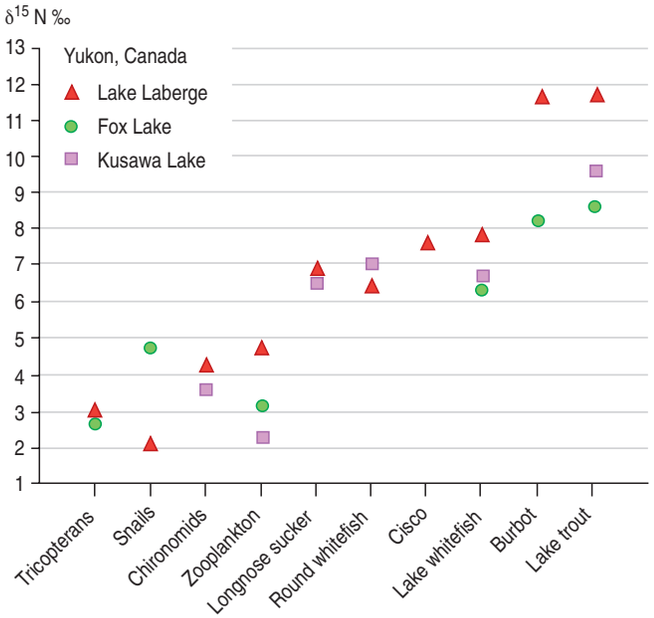



Figure 6·19. Mean $\delta^{15}$ nitrogen (‰) for invertebrates and fish collected from Laberge, Fox, and Kusawa Lakes.

tained for $\delta^{15}$N and OCs in the food webs of the reference lakes. The trophic difference between Laberge and the reference lakes provides one explanation for the higher concentrations of toxaphene, DDE, and PCBs in the fish from Lake Laberge.

### 6.6.3.4.2. Great Slave Lake and Slave River studies, Canada

Fish in Great Slave Lake, the seventh largest freshwater lake in North America, also have higher concentrations of persistent OCs and PCDD/Fs than the same species in the smaller surrounding lakes such as Trout, Gordon, and Alexie Lakes. Evans (1994) and Evans *et al.* (1995) determined levels of OCs in food web organisms in the eastern and western basins of Great Slave Lake and showed that OC concentrations were strongly associated with trophic level. Slopes for the regressions were similar although a little less steep than the 0.16-0.32 slopes observed by Kidd *et al.* (1995a, 1995b) for Lake Laberge in the Yukon. These slopes indicate that organic contaminants may not biomagnify as strongly in Great Slave Lake food webs as in the Lake Laberge food web.

$^{15}$N values were similar in burbot from Great Slave Lake and Lake Laberge, suggesting that burbot feed at similar positions in the food webs in both lakes. Toxaphene concentrations were substantially lower in Great Slave Lake burbot liver, averaging 244 ng/g ww for the Slave River delta and 762 ng/g for East Arm burbot, than the 2820 ng/g observed for Lake Laberge. This suggests that trophic level alone cannot account for the high toxaphene concentrations in Lake Laberge burbot liver. There were also differences in concentrations in invertebrates between Lake Laberge and Great Slave Lake. It is difficult to assess whether these inter-lake differences in contaminant concentration reflect true differences between the systems or are more a reflection of the inherent variability in such measurements. The resolution of this is important because the concentrations of organic contaminants in fish appear to be a function not only of trophic level but also of other aspects of the lake ecosystem. Thus, while there is some evidence of similarities in the slope of contaminant concentration as a function of trophic level ($^{15}$N) for a variety of lakes, the intercepts, representing overall contaminant levels, differ.

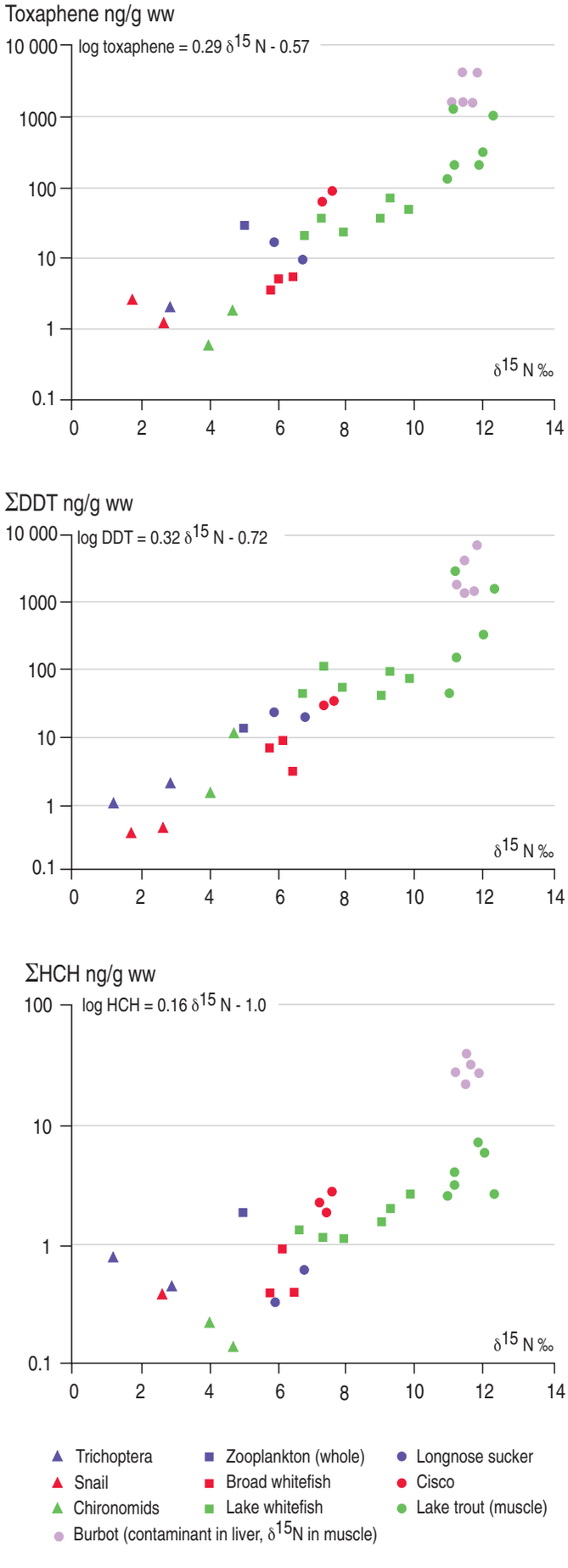



Figure 6·20. $\delta^{15}$ Nitrogen isotope ratios versus Toxaphene, DDT, and HCH for fish and invertebrates from Lake Laberge, Yukon Territory. (Kidd and Schindler 1994).

Great Slave Lake receives most of its water flow from the Slave River, which drains a large area of northwestern Canada (983 000 km$^2$). The western basin of Great Slave Lake, as well as the Mackenzie River, are greatly influenced by flows of the Slave River. The Slave River Environmental Quality Monitoring Program, conducted between 1990 and 1995, was designed to characterize the baseline aquatic ecosystem conditions in the Slave River at Fort Smith, NWT (Peddle *et al.* 1995). Water, suspended sediment, and fish samples were collected from the Slave River at Fort Smith and at two control sites and analyzed for organic and inorganic contaminants, including PCDD/Fs, PCBs, and persistent OC pesticides such as toxaphene (Peddle *et al.* 1995). Levels of these OCs in fish are given in Annex Table 6·A9 and PCDD/Fs in Annex Table 6·A22.

Toxaphene and PCBs were the predominant OC contaminants in walleye and burbot liver from the Slave River and the control lakes. A wide range of toxaphene concentrations were found in burbot liver, however, in general, concentrations were similar to levels observed in burbot liver from Great Slave Lake, but higher than in smaller control lakes (e.g., Alexie Lake).

Higher levels of toxaphene, DDT, and PCBs in walleye and burbot tissues compared to control lakes were likely due to longer food chains in the Slave River and Great Slave Lake, where these species migrate, compared to the small control lakes.

In walleye (whole fish) and burbot liver, 2,3,7,8 TCDD and 2,3,7,8 TCDF isomers generally made up 100 percent of the total TCDD and TCDF detected (nPCBs were not determined). The TCDD and TCDF levels in walleye from the control lakes were similar to those from the Slave River. In burbot liver, however, the levels were higher in the Slave River than the control lake. The TCDF/TCDD ratio suggested that the major source of these contaminants was from bleached kraft mill discharges that occurred upstream (see also section 6.2.4.3.2).

### 6.6.3.5. Summary and conclusions – freshwater environment

#### 6.6.3.5.1. Abiotic environment

Recent studies of contaminants in water, sediments, and biota from Arctic lakes and rivers have greatly expanded the information available on OC levels. Particularly important for understanding spatial trends are new data on OCs in lake sediments and fish from Alaska, Greenland, Norway, Finland, Sweden, and Russia. With the exception of river and lake waters, the original minimum monitoring objectives of AMAP for POPs in freshwater matrices (AMAP 1993) have been met in all circumpolar countries.

Knowledge of concentrations of OCs in northward flowing rivers is useful for estimating contaminant loadings to the Arctic Ocean and for identifying local/regional point sources. This information is still extremely limited spatially and temporally. Loadings of HCH to the Arctic Ocean can be estimated with reasonable accuracy for the Mackenzie River, for three Norwegian rivers (Orkla, Vefsna and Alta; for γ-HCH only), and for major Russian rivers. Concentrations of γ-HCH in Russian rivers are similar to results for Norway and Canada. However, higher γ-/α-HCH ratios are found in the Russian rivers suggesting continued inputs of the pesticide lindane in these areas.

Data on PCBs and ΣDDT in Russian rivers are problematic. PCB and ΣDDT concentrations in river water and suspended solids reported in recent surveys of Russian rivers are ten to 100 times higher than found in Canadian or Norwegian rivers. PCB and ΣDDT levels on suspended solids in the Ob and Yenisey Rivers are higher than found in river water near industrialized areas in North America. Although there may be unidentified quality assurance problems with the PCB and DDT data for Russian rivers, results from independent Russian and Norwegian studies of bottom surficial sediments in the Indigirka River delta and Pechora River were roughly in agreement considering the heterogeneous nature of the bottom material. The Norwegian study actually found higher levels of PCB and other OCs than did Melnikov *et al.* (1996a, 1996b) at these two locations. Thus, the data for PCBs and DDT in Russian rivers, while reflecting some sample contamination problems, may in fact be extraordinarily high even compared with surface waters of urban areas of North America and western Europe. In either case, further investigation of the extent of PCB and DDT contamination of these rivers is needed.

The emphasis on measurements of OCs in suspended solids in Russian river waters rather than in the dissolved phase also limits the usefulness of the data for estimating loadings to the Arctic Ocean. In the Mackenzie River, suspended solid loads represented only about 10% of total loadings of PCBs and < 1% for ΣHCH because most of the OCs were in the dissolved (or dissolved organic carbon associated) phase.

The AMAP project directory lists joint USA/Russian projects (by Geochemical and Environmental Research Group (GERL) of Texas A&M University) in which water samples were collected in the Ob and Yenisey Rivers for analysis of POPs. However, results from these studies are not yet available.

Information on POPs in lake water is confined to a small number of lakes in the Canadian Arctic and to samples from two lakes on the Taimyr Peninsula (Melnikov and Vlasov 1995) so that little can be said about circumpolar trends. HCH, HCB, and toxaphene, as well as several less persistent OCs, were readily detectable in lake water, which suggests that the water column is an important reservoir for the more polar OCs (i.e., log $K_{ow}$ < 5.5). Although these OCs are prominent in lake waters, their levels in Arctic lakes do not correlate well with levels in fish, which underlines the importance of food web rather than direct water pathways for uptake by fish, especially for toxaphene.

ΣHCH concentrations in deep waters of two lakes on the Taimyr Peninsula were higher than in most Canadian Arctic lakes, suggesting greater inputs of lindane as was also observed in river water. Very high levels of ΣDDT and ΣPCB were found in the two lakes on the Taimyr Peninsula relative to Canadian Arctic lakes or the Great Lakes in general. As noted for river water, there may be quality assurance problems with these PCB and DDT data that precludes their use for assessment purposes at the present. Whether or not levels are indeed this high deserves further investigation.

The less persistent OC pesticides endosulfan, methoxychlor, and pentachloroanisole are detectable in lake waters in the Arctic. This is consistent with their presence in Arctic air and plants. Levels are, however, low relative to HCH. Circumpolar data on levels of these current use OCs in water or sediments are not available.

Concentrations of ΣPCBs in Arctic lake waters in Canada exceeded the USEPA Great Lakes Water Quality guideline for protection of aquatic life of 17 pg/L, but ΣDDT did not exceed the guideline limit of 870 pg/L (Table 6·13). Levels of ΣPCB reported in water of Lake Levinson-Lessing on the Taimyr Peninsula exceeded Canadian environmental quality guideline limits of 1 ng/L.

Circumpolar coverage of POPs in freshwater lake surface sediments is, in comparison with information for freshwa-

Table 6·13. Selected criteria, action levels, or guidelines for critical pollutants in the Great Lakes (modified from De Vault *et al*. 1995).

| Contaminant | USFDA(1) Fish[a] | IJC (2) Water | GLI (4) Water | OMEE(5) Water | IJC (3) Fish tissue[a] | EQG (6) Fish tissue[a] | USEPA (7) Fish tissue[a] | ERL (8) Sediment[b] | ERM (8) Sediment[b] | EQG(6) Water | EQG (6) Sediment[b] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 2,3,7,8 TCDD | 25 pg/g | | 0.0096 pg/L | | | 1.1 pg/g | 0.5 pg/g | | | 0.02 pg/L | 0.091 pg/g |
| DDT | 5 mg/g | 0.003 µg/L | 0.00087 µg/L | 0.003 µg/L | 1.0 µg/g[c] | 0.0063 µg/g | 0.039 µg/g | 1.6 ng/g | 46 ng/g | | |
| Total PCBs | | | 17 pg/L | 0.001 µg/L | 0.1 µg/g[c] | 0.0076 µg/g[d] 0.006µg/g[e] | 0.16 µg/g | 23 ng/g | 180 ng/g | | |
| Mirex | 2 µg/g | < detection | | 0.001 µg/L | | | | | | | |
| Toxaphene | 5 µg/g | 0.008 µg/L | | 0.008 µg/L | | | | | | | |
| Aldrin/dieldrin | 0.3 µg/g | 0.001 µg/L | | 0.001 µg/L | 0.3 µg/g[c] | | | | | | |

a. All values for fish are based on wet weight.
b. All values for sediment are based on dry weight.
c. Whole fish.
d. Freshwater fish.
e. Marine fish.

(1) USFDA (US Food and Drug Administration) action levels in edible portions of fish for regulation of interstate commerce.
(2) (3) International Joint Commission, Annex 1 objectives for protection of aquatic life and wildlife.
(4) USEPA (US Environmental Protection Agency) Great Lakes Water Quality Guidance, proposed criteria for protection of wildlife (USEPA 1995).
(5) Ontario Ministry of Environment and Energy (OMEE 1993).
(6) Draft Canadian Environmental Quality Guidelines for protection of animals that consume aquatic biota (Environment Canada 1996).
(7) USEPA guideline values for assessment of hazards to fish-eating wildlife (USEPA 1995).
(8) ERL = Effects range low and ERM = effects range median for sediments (Long *et al*. 1995).

ters, reasonably complete, with some results from every circumpolar country. No geographical trend in concentrations of any OC is discernible. This is not surprising given the influence of lake sedimentation rates, which in turn may be influenced by lake nutrient status, morphometry (steepness, mean depth), and organic and inorganic inputs from the watershed. In general, surface sediment samples from remote Arctic lakes have similar or lower concentrations of ΣPCB and ΣDDT as compared to mid-latitude lakes in North America and far lower levels than lakes or reservoirs near industrialized areas in North America and western Europe.

Concentrations of ΣPCB and ΣDDT in Arctic lake surface sediments generally do not exceed guideline limits for effects on aquatic life (Table 6·13). Sediments from only a few locations exceed the Effects Range Low (ERL) or Effects Range Median (ERM) values. Values below the ERL are referred to as being in the minimal effects range and those between the ERL and ERM as in the low probability of effects range. ERL and ERM were derived by Long *et al*. (1995) from a review of contaminant levels and biological effects data for sediments from the USA and Canada. In the case of ΣPCB, sediments from five lakes in Canada, one in Norway (Bear Island), and one in Russia (Yamal Peninsula) exceed the ERL, and only one location, Wonder Lake (Alaska), exceeded the ERM. The ERL for ΣDDT of 1.6 ng/g dw was exceeded at about ten locations (Annex Table 6·A8), but the ERM (46 ng/g dw) was not exceeded. ERL and ERM values are not available for HCH isomers, HCB, or TCDD. TCDD TEQ levels in all sediments exceeded Canadian Environmental Quality guidelines for protection of aquatic life of 0.09 pg/g dw.

In the case of PCDD/Fs, there are higher concentrations of TEQs in Finnish than in Canadian (Great Slave Lake) sediment cores. However, this difference may be due to much greater dilution from high sedimentation rates in the Great Slave Lake cores. Much higher concentrations of PCDD/Fs have been found in sediments in the Great Lakes and near industrialized areas of Europe. The high proportion of OCDD in all Arctic lake sediments suggests that the major source of PCDD/Fs is combustion-related.

The availability of results for OCs in dated lake sediment cores has provided, for the first time, regional estimates of current and historical fluxes to freshwater and terrestrial environments. It has also provided insights into temporal trends (section 6.7.3.1). Significant latitudinal decline of fluxes of penta- to octachlorobiphenyls, as well as DDT-related compounds, with increasing north latitude, and the lack of a decline in fluxes of di-/tri-chlorobiphenyl fluxes in Canadian mid-latitude and Arctic sediment cores are consistent with predictions of the 'cold condensation' hypothesis (Wania and Mackay 1993). The results for PCB fluxes in Finland and Alaska sediment cores are in general agreement with the Canadian data (Muir *et al*. 1996b). Because the results for dated sediment cores are limited to Canada, Alaska, and Finland, there is insufficient information with which to estimate OC fluxes on a circumpolar basis.

The fact that fluxes predicted from lake surface sediment in the Canadian High Arctic agree well with observed PCB flux in a snow core from the Agassiz ice cap on northern Ellesmere Island (Gregor *et al*. 1995) and with recent estimates of PCB fluxes at other sites in the Canadian Arctic (Barrie *et al*. 1997) was unexpected. Mass balance calculations suggest that retention of OCs in Arctic freshwater lakes during summer snow melt is very inefficient because most of the meltwater does not mix and net sedimentation rates are generally low (Diamond 1994, Semkin 1996).

#### 6.6.3.5.2. **Biota**

Toxaphene is the major OC contaminant in all freshwater fish and invertebrates that have been analyzed from the Canadian Arctic and West Greenland. Although relatively little data on toxaphene levels in freshwater environments are available for Norway, Finland, and Russia, results from Finland (Paasivirta and Rantio 1991), as well as measurements in anadromous char and whitefish from Russia (section 6.6.4.5.1), suggest that toxaphene is also a prominent OC pesticide contaminant in fish in the European Arctic. No data are available for toxaphene in surface waters, sediments, or lower food web organisms in the European Arctic.

Because the source of most OC contaminants in Arctic freshwaters is atmospheric, prevailing air concentrations are relatively similar (within a factor of 2) at the four polar air-sampling stations, and fluxes to lake sediments vary over a narrow range, similar levels of OCs might be expected in fish over a wide geographic area. However, this is not the case. Highest toxaphene and PCB levels are generally seen in fish that are strictly piscivorous such as lake trout and burbot. Toxaphene and PCB levels also show significant correlation with fish size within lakes (Muir *et al*. 1997). In the case of lake trout and whitefish data from the Canadian Arctic,

levels of OCs are not strongly correlated with lipid content (Muir and Lockhart 1996), possibly because lipid levels in individuals fluctuate seasonally. Thus, for Canadian freshwater fish, lipid normalization does not help to improve understanding of geographical trends although it is useful for qualitatively examining differences among species and among regions.

On a lipid weight basis, POP levels in freshwater fish are generally higher than levels in terrestrial herbivores and are similar to or higher than levels in mammalian carnivores.

Differences in levels of bioaccumulating contaminants, such as PCBs in lake trout in Ontario lakes have been attributed to food chain length and lipid content (Rasmussen *et al.* 1990). Toxaphene and PCB concentrations within the food webs of all lakes examined so far (Fox, Kusawa, Laberge, and Great Slave Lakes) correlate significantly with $^{15}N$ in muscle (Kidd *et al.* 1995a, Evans 1996). The possibility exists that PCB and toxaphene levels in Arctic lakes with lake trout or burbot as top predators could be predicted with this empirical relationship. While there is some evidence of similarities in the slope of contaminant concentration as a function of position in the food chain ($^{15}N$) for a variety of lakes, the intercepts differ due to lake differences in the proportion of $^{15}N$ in lower trophic organisms. Therefore, it is not possible at present to predict levels of OCs in fish from $^{15}N$ except by thorough study of each food web.

With the exception of Arctic char (discussed in section 6.6.3.3.1), there is insufficient geographical coverage of any freshwater species to permit examination of circumpolar trends of OCs. Although a north-south trend in PCB (lipid-normalized) concentrations (but not for toxaphene or more volatile OCs such as HCH) was previously observed in burbot liver collected from a series of lakes and riverine sites in central and northwestern Canada (Muir *et al.* 1990a), no geographic trends of levels or patterns of OCs can be discerned for lake trout, burbot, or lake whitefish (lipid-normalized) from within NWT and the Yukon. There may be no trend or there may have been too few samples to detect a trend, especially considering the confounding effects of fish size and food chain length.

Char and lake trout from Arctic lakes clearly have lower levels of ΣPCBs and ΣDDT when compared to levels in the Great Lakes (Figure 6·17). Lake trout from smaller remote lakes in Alberta and northwestern Ontario, which receive contaminants solely from the atmosphere, have ΣPCB levels similar to Arctic salmonids, indicating that proximity to sources rather than north latitude, is a critical factor in explaining spatial trends. An exception is lake trout from Lake Laberge which have similar levels (on both a wet weight and lipid weight basis) to those in Lake Superior, one of the Great Lakes. Char from northern Norway and Finland had lower levels than char from southern Sweden (Lake Vättern).

The ΣPCB concentrations found in Arctic char, lake whitefish, and lake trout muscle generally exceed the most conservative guideline limits for protection of aquatic life. These limits range from 7.6 ng/g ww (Canada) to 160 ng/g ww (USEPA 1995) (Table 6·13). Most lake trout exceed the Canadian guideline, while Arctic char and lake whitefish do so only in a few locations. Fish from Lake Laberge are an exception; all three species exceed the USEPA guideline limit of 160 ng/g ww. No fish muscle samples were found to exceed the PCB guideline limit for human consumption and export of fish of 2 μg/g ww which is widely used in Europe and North America.

There now exists a relatively large data set on PCDD/Fs in freshwater fish from Canada, Norway, and northern Finland and Sweden. TCDD levels are low (typically < 1 pg/g) in comparison to levels in fish sampled near bleached kraft mills (Servos *et al.* 1994) or to species in the lower Great Lakes or the Baltic Sea. TCDD TEQ concentrations in fish muscle, based on PCDD/Fs only, did not exceed guideline limits for human consumption (20 pg/g ww) used in most circumpolar countries. Some fish tissues, especially burbot liver and lake trout muscle in Lake Laberge, approach or exceed the 20 pg/g ww limit if planar PCBs are included in the calculation of TCDD TEQs. With the exception of the lakes Pahtajärvi and Nitsijärvi in Finland, TCDD TEQ levels in fish tissue (based on PCDD/Fs) also did not exceed the USEPA (1995) guideline for protection of fish-eating wildlife of 0.57 pg/g ww.

Fewer measurements have been made of the toxic non-*ortho* substituted PCBs and other planar OCs, such as chlorinated diphenyl ethers (PCDEs) and polychlorinated naphthalenes (PCNs), in Arctic fish, but, in general, concentrations are an order of magnitude higher than those of PCDD/Fs. Where nPCBs have been measured along with PCDD/Fs, PCNs, and PCDEs, calculation of TCDD TEQs shows that nPCBs, especially CB 126, account for most of the TEQs. However, knowledge of levels of nPCBs in freshwater fish analyzed in the Arctic are very limited in both Canada and Scandinavia, and non-existent for Alaska, Iceland, Greenland, and Russia. Therefore, the priority for future measurements of toxic planar OCs in the Arctic should be additional measurements of nPCBs. More information is also needed on circumpolar trends of PCNs, PCDEs, and PBDEs because of their toxicological significance and the fact that they have been analyzed in very few samples. No data are available from Canadian or Alaskan freshwater sediments or biota for a wide range of planar OCs, including PCNs, PCDEs or their brominated analogs (PBDEs).

Although less persistent OC pesticides have been detected in Arctic air, plants, and lake waters, little is known about their presence in Arctic fish or sediments. The limited data suggest that compounds such as endosulfan, methoxychlor, and pentachloroanisole are not present at high levels in Arctic fish in comparison with persistent OCs. Although the data suggest that biomagnification is not occurring, little is known about circumpolar trends of methoxychlor and other less persistent pesticides because their analysis has been limited to fish in Arctic Canada.

### 6.6.4. Marine environment

The marine environment was the first component of the Arctic environment to be examined for the presence of persistent organic pollutants. As was noted in the introduction to this chapter, studies of the presence of organochlorine contaminants began in the late 1960s. Holden (1970) reported the presence of dieldrin and DDT as well as PCBs in blubber of ringed seals from the Canadian and Norwegian Arctic. Reviews of the information on persistent OCs in the Arctic marine environment that were available up to 1991 (Andersson *et al.* 1988, Muir *et al.* 1992b) identified a general lack of data on OCs in the abiotic Arctic environment, and limited information on circumpolar spatial trends in biota. The past five years has seen a large increase in the amount of data on persistent OCs available, especially from the European and Russian Arctic.

This assessment first examines levels of major OCs in Arctic seawater, ice, snow, and marine sediments. These data are summarized in: Annex Table 6·A7– OCs in suspended sediment in Russian rivers; Annex Table 6·A10 – OCs in seawater in Russia; Annex Table 6·A11 – OCs in suspended particulate matter from Russian seas; Annex Table 6·A12 –

Table 6·14. A summary of measurements of organochlorine concentrations (ng/m$^3$ or pg/L ) in seawater[a] (mean ± S.D.).

| | Norwegian Sea[b] | Barents Sea[c] (N=2) | Laptev Sea[d] (N=3) | Kara Sea[d] | Pechora Sea[d] | Canadian Archipelago[e] (N=8) | Bering Sea[f] (N=4) | Bering Sea[g] (N=7) | Chukchi Sea[f] (N=3) | Chukchi Sea[g,h] (N=2) |
|---|---|---|---|---|---|---|---|---|---|---|
| Year | 1985 | 1992 | 1994 | 1994-95 | 1992 | 1992 | 1990 | 1993 | 1990 | 1993 |
| α-HCH | 2750 | 477±85 | 260±270 | 120-460 | 330±250 | 4180±1600 | 1500 | 1990±390 | 1400 | 2060±360 |
| γ-HCH | 380 | 120±36 | 160±240 | 100-330 | 320±540 | 430±120 | 190 | 450±120 | 180 | 430±100 |
| Toxaphene | – | 14±4 | – | – | – | 48±8 | – | 22±6 | – | 15±1 |
| ΣCHL | – | 3±1 | 240±260 | <10-570 | 20±40 | 13±6 | 3.9 | 2.7±1.5 | 4.0 | 1.2±0.1 |
| Heptachlor epoxide | – | – | 40±40 | <10-70 | <10 | – | – | 2.4±0.5 | – | 2.8±0.1 |
| Dieldrin | – | – | 30±70 | <10 | <10 | – | – | 3.7±0.7 | – | 3.4±0.2 |
| Endosulfan | – | – | – | – | – | – | – | 2.1±0.4 | – | 1.8±0.1 |
| ΣDDT | <50 | 3±1 | 760±590 | 50-450 | 270±350 | 1.0±0.3 | 1.0 | – | 0.3 | – |
| ΣPCB | <500 | 38±15 | 2540±2940 | 510-6120 | 550±750 | – | 12 | – | 8.4 | – |

a. Samples of surface waters (0-1 m depth). Dash indicates not analyzed or results not reported.
b. Gaul 1989. Average of two sites, 77°30'N, 15°55'W and 77°00'N, 1°33'W.
c. Chernyak *et al.* 1995. Coastal waters near the Kola Peninsula.
d. Melnikov and Vlasov 1995. Surface waters 1 m depth (filtered). Results for the Kara Sea are range of means for three locations sampled in 1995 (Baydaratskaya Gulf, Yenisey Gulf, Ob Gulf).
e. Bidleman *et al.* 1995b. N=8 except for DDTs where N=4; HCH results from Falconer *et al.* 1995a (N=12).
f. Iwata *et al.* 1993.
g. Bidleman unpubl.; HCH results from Jantunen and Bidleman 1995 (N=14).
h. Siberian Chukchi, N=7 for HCH.

OCs in ice cover in Russian seas; Annex Table 6·A14 – OCs in marine sediments; and Annex Table 6·A21 – PCDD/Fs in freshwater and marine sediments. Recent data on OCs in marine and anadromous fish, seabirds, marine mammals, polar bear, and Arctic fox are also presented. These data are summarized in: Annex Table 6·A15 – OCs in marine invertebrates; Annex Table 6·A16 – OCs in seabird and shorebirds; Annex Table 6·A17 – OCs in anadromous and marine fish; Annex Table 6·A18 – OCs in marine mammals; Annex Table 6·A19 – PCDD/Fs and nPCBs in marine biota; Annex Table 6·A20 – OCs in polar bear and Arctic fox; and Annex Table 6·A22 – PCDD/Fs and nPCBs in terrestrial and freshwater biota.

### 6.6.4.1. **Seawater**

Data for POPs in the water column and ice published prior to 1991 have been reviewed by Muir *et al.* (1992b). Analysis of samples of seawater collected during the mid- to late 1980s showed the presence of OCs in waters of the Norwegian, Russian, Alaskan, and Canadian Arctic Ocean and in the Bering Sea. The most predominant chlorinated POP was HCH (Table 6·14). Of all the OCs measured, the most complete water-column database is for α- and γ-HCH. Gaul (1989) found average concentrations of 2750 and 380 pg/L for α- and γ-HCH, respectively, from two sites in the Norwegian Sea (73°30'N-80°48'N) in 1985 near the edge of the ice pack (Table 6·14). Gaul (1989) observed a gradient of ΣHCH (α- + γ-HCH) between the North Sea, where concentrations ranged from 4800 to 6200 pg/L, and the Norwegian Sea, where they ranged from 900 to 1500 pg/L.

In a large survey of OCs in surface seawater collected during 1986-1989 from the Kara, Laptev, East Siberian, and Chukchi Seas, Vlasov and Melnikov (1990) found ΣHCH concentrations ranging from 1580 to 2170 pg/L. In a repeat of this survey in 1994-1995, Melnikov *et al.* (1995) reported mean ΣHCH levels ranging from 180 to 2900 pg/L in the Pechora, Kara, and Laptev Seas. These ΣHCH results in the Laptev Sea were in good agreement with work by Jantunen and Bidleman (1995) and Chernyak *et al.* (1995). Chernyak *et al.* (1995) reported mean levels of ΣHCH of 720 ± 120 pg/L in samples collected in 1992 from the Barents Sea coast of Russia. In general, levels of ΣHCH in the Norwegian and western Russian waters were lower than in seawater from the Canadian, Alaskan, and east Russian Arctic (Table 6·14). An exception is the Pechora Gulf where the highest levels of ΣHCH were found along the Russian coast (means of 1270 and 1590 pg/L in 1995 for α- and γ-HCH, respectively) (Melnikov and Vlasov 1995).

The most thoroughly studied Arctic region, in terms of concentrations of OCs in seawater, is the Bering/Chukchi Seas (Table 6·14). Joint USA/Russian (Hinckley *et al.* 1991, Jantunen and Bidleman 1995) and Japanese cruises (Iwata *et al.* 1993) have determined OCs in seawater from this region. The study by Iwata *et al.* (1993) determined OCs in surface seawater during 1989-1990 from 73 locations in the northern hemisphere and found highest seawater concentrations of ΣHCH in the Bering/Chukchi Seas (1400-2100 pg/L) and Gulf of Alaska (1700-2100 pg/L) compared to almost all other locations (58-1600 pg/L). HCH levels appeared to increase in a smooth gradient with latitude from the tropical western Pacific Ocean to the Arctic Ocean (Figure 6·21). Wania and Mackay (1995, 1996) have suggested that this is evidence of the 'cold-condensation' effect. Other less volatile OCs (e.g., chlordanes, PCBs, DDTs) were present at lower concentrations in the Bering/Chukchi Seas than at more tem-

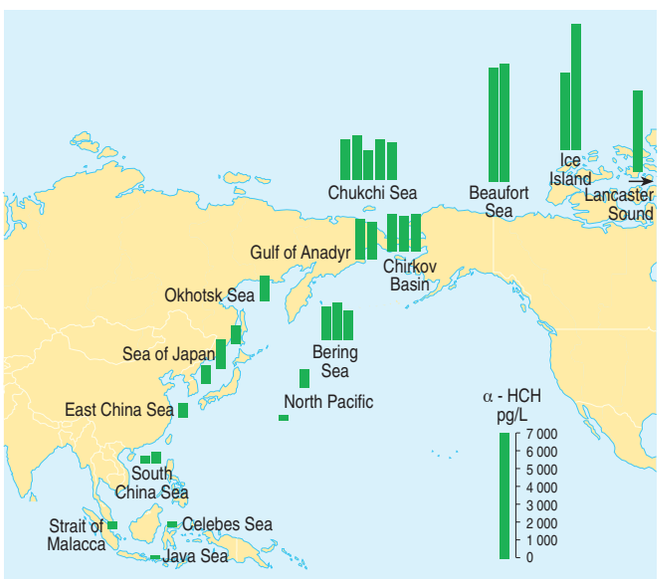



Figure 6·21. α-HCH (pg/L) in seawater: an illustration of the cold condensation effect (Wania and Mackay 1996, Iwata *et al.* 1993).

perate latitudes. ΣPCB concentrations decreased from means of 17 and 26 pg/L in the East China Sea and Japanese North Pacific, respectively, to an average of 8.4 pg/L in the Chukchi Sea (Iwata *et al.* 1993).

Most recent data confirm that the relative abundance of OCs in Arctic seawater is α-HCH > HCB > γ-HCH ≈ toxaphene > chlordanes ≈ PCBs > DDTs as described by Bidleman *et al.* (1990). Iwata *et al.* (1993) found α-HCH > γ-HCH > PCBs > chlordanes > DDTs in the Chukchi sea. The exception is the recent survey of Russian seawater which found ΣPCBs > ΣDDT > ΣHCH > chlordanes > chlorobenzenes. Levels of ΣPCB in the Russian Arctic seas are much higher than observed in other studies with seawater (Table 6·14), but are similar to some results for riverine inputs (section 6.6.3.1.1). ΣPCB concentrations ranging as high as 15 000 pg/L (Annex Table 6·A10) are difficult to reconcile with data from other studies (e.g., Hargrave *et al.* 1988, Iwata *et al.* 1993, Chernyak *et al.* 1995), which found PCB concentrations in seawater to be in the low pg/L range (< 4-38 pg/L). However, they are consistent with the results of Joiris *et al.* (1995) who reported whole water concentrations of 22-52 000 pg/L for PCBs (based on measurements in suspended particulate matter) of the Barents Sea and waters near the North Pole. The results are also consistent with other reports of elevated DDT and PCBs in suspended sediments of Russian rivers (see section 6.6.3.1.1). Unfortunately, there are no measurements of PCBs in whole water samples of Russian rivers available at present. Studies by Roshydromet (1995) included only OC pesticides. A project by Texas A&M University, listed in the AMAP project directory, mentions collection of water samples for analysis of POPs, but results from this work are not yet available. If confirmed by future measurements, this implies major inputs of DDT and PCB to the Arctic Ocean from Russian sources.

Despite possible quality assurance problems with the Russian data for PCB, some interesting spatial trends emerge for OCs (Annex Table 6·A10). Higher concentrations of most OCs, especially ΣHCH and ΣCHL, were found in seawater from shallow areas influenced by riverine inputs such as the Baydaratskaya, Ob, and Pechora Gulfs, than in samples collected in more northerly parts of the Laptev and Kara Seas.

HCHs are not uniformly distributed within the Arctic Ocean (Figures 6·22 and 6·23). A large proportion of HCH is contained in the top 200 m of the water column. The Canada Basin and other Canadian sites contain pools of surface water where HCH concentrations are elevated by a factor of 2 or more compared to other regions of the Arctic Ocean or oceans to the south (Figures 6·21). This pool of surface water in the Canada Basin has a relatively long residence time (chapter 3). Due to permanent ice cover, it does not easily outgas. The surface concentrations in the Canada Basin may, therefore, reflect conditions a decade or so ago when atmospheric HCH concentrations were much greater. Relatively high concentrations of HCH in water of the Canadian Archipelago waters (Falconer *et al.* 1995a) are, therefore, explained by the flow of surface waters from the Canada Basin.

Since 1985, a number of vertical profiles have been measured for both α- and γ-HCH (Figure 6·22), allowing assessment of the inventories of these compounds (Table 6·15, section 6.6.4.3.1.1) and their distributions in the ocean. Most of the data have been collected near the surface of the ocean with only relatively few points from water deeper than 400 m. Concentration ranges vary from 'very low' in the deep ocean to as much as 7 ng/L for α-HCH and 1.1 ng/L for γ-HCH at the surface. The concentrations of both forms of HCH are highest in the top 200 m of the water column. An important observation is that, horizontally, HCH is not uniformly distributed in surface waters; stations in the southern part of the Canada Basin contain relatively high concentrations, over twice those observed in the Chukchi and East Siberian Seas and in the Greenland Sea.

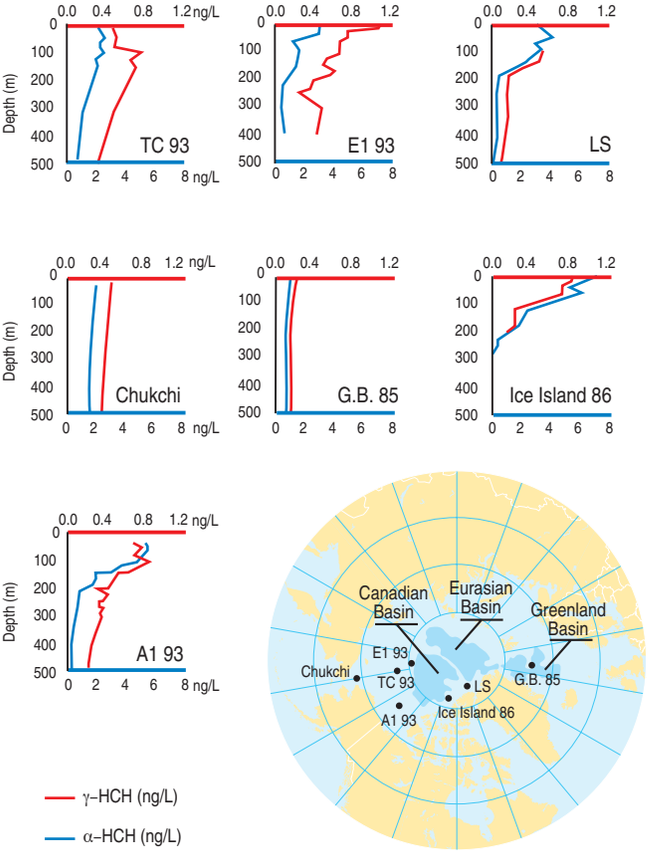



Figure 6·22. Vertical profiles of HCH for the Arctic Ocean. Data have been taken from Gaul (1989); Hargrave *et al.* (1988) (Ice Island); Jantunen and Bidleman (1995) (Chukchi); Macdonald (1994) (A1, TC, E1, LS).

There are too few data to assess the vertical distribution of OCs other than HCH. At 200 m depth at Ice Island, north of Axel Heiberg Island, at approximately 81°N (Hargrave *et al.* 1988), HCB, chlordane, and dieldrin decreased to one half or less than a half of their surface concentrations. Melnikov *et al.* (1995) found up to two-fold lower concentrations of ΣHCH and ΣDDT at depths of 15-30 m in the Laptev and Kara Seas than in surface waters, but no consistent trend for ΣPCB or ΣCHL.

Toxaphene is the second most prominent OC contaminant in Arctic Ocean water. It was first identified in seawater in the Canadian Arctic at Ice Island in 1986-1987. More recent measurements were made in the Canadian Archipelago in 1992-1993 (Bidleman *et al.* 1995a, Hargrave 1996) and on oceanographic cruises through the Bering and Chukchi Seas and across the polar cap (Bidleman *et al.* 1996) (Table 6·14). Figure 6·24 summarizes measurements of toxaphene in surface water on the Bering/Chukchi Seas (1993) and the transpolar cruises (1994). Concentrations of toxaphene increase from 15-30 pg/L in the Bering/Chukchi Seas to 90 to 120 pg/L at higher latitudes. In comparison, toxaphene in lakes within the Yukon River basin ranged from 20 to 270 pg/L. The spatial distributions of toxaphene and HCHs in the Arctic Ocean are similar in that both pesticides are elevated in more northerly waters. The range of toxaphene concentrations from 1992-1993 falls below the 1986-1987 Ice Island measurements. However, considering that only two surface samples were analyzed in the latter study, it is not possible to say whether a real decline has occurred. Unlike

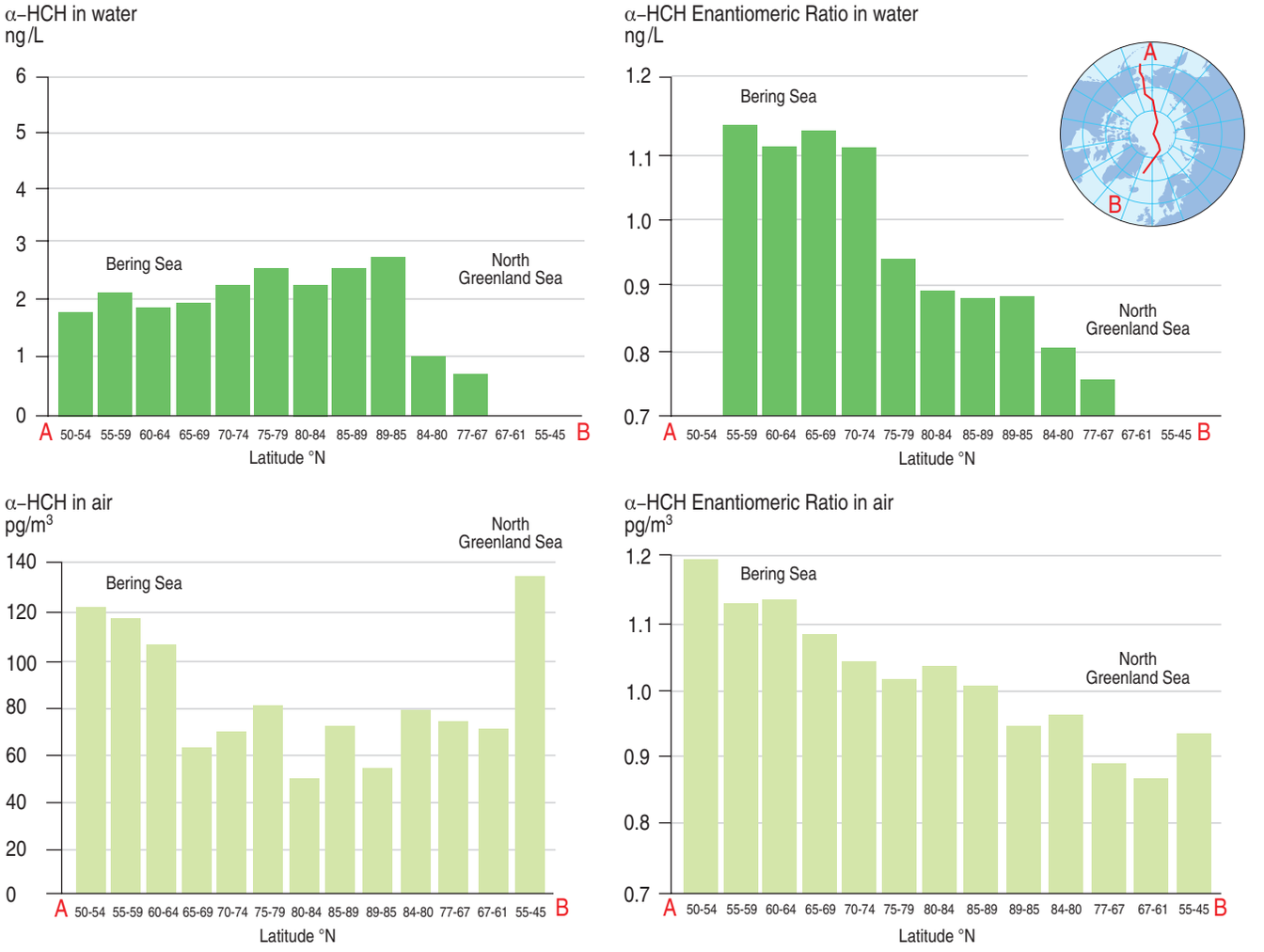



Figure 6·23. Trends in α-HCH and enantiomer ratios (ER) on a transect from the Bering Sea across the polar cap to the Greenland Sea during the Arctic Ocean Transect Study of July and August 1994. The pesticide is produced with a racemic ratio (ER = 1.00). An ER less than 1.00 implies selective breakdown of the (+)α-HCH enantiomer.

HCHs, there are no measurements of toxaphene in eastern Arctic waters, including the Norwegian and Barents Seas.

The following scenario is suggested to account for the elevated toxaphene and HCHs in the northern Canada Basin. Atmospheric levels of HCHs in the early to mid-1980s were about 4-8 times higher than present-day values (Bidleman *et al.* 1995b, Jantunen and Bidleman 1995, see Figure 6·43), and there is a suggestion that toxaphene levels were also higher in the last decade, perhaps by a factor of 2-6 (see section 6.7.1.1). Thus, it is likely that atmospheric loadings of these pesticides were greater in the past, especially to the regional seas which surround the central Arctic Ocean. These seas are largely unfrozen during the summer months and are susceptible to gas exchange, whereas exchange at higher latitudes is inhibited by the ice cover. Over time, water containing high concentrations of toxaphene and HCHs has been transported from the regional seas into the central Arctic Ocean. Today these regional seas have cleared themselves by outgassing and sedimentation and have become adjusted to reduced atmospheric inputs, but these processes do not operate efficiently in the High Arctic. Toxaphene and HCHs which are trapped under the polar cap are thus 'ghosts of the past' and will be slowly drained, largely through the Canadian Archipelago, on a time scale of decades.

Hargrave (1996) monitored OC pesticides in the upper 50 m of the water column in the Canadian Archipelago at Resolute Bay during 1993. Mean concentrations of HCHs (4100 pg/L) and toxaphene (85 pg/L) were similar to levels found under the polar cap on the transpolar cruise in 1994

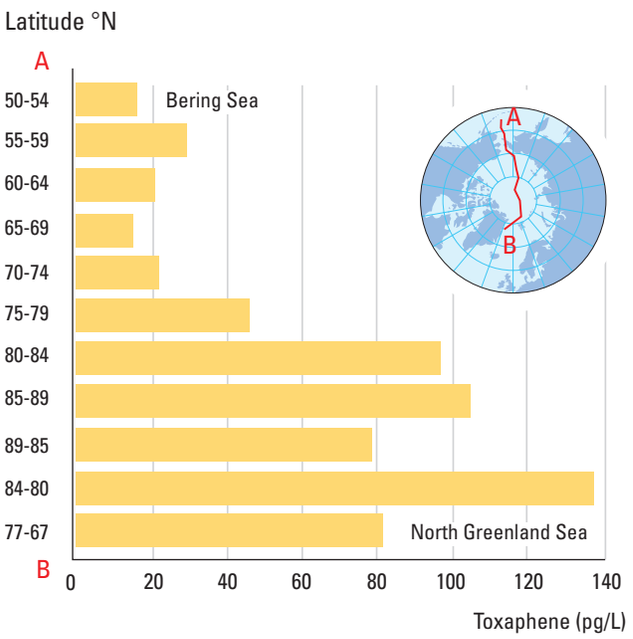



Figure 6·24. Measurements of toxaphene in surface water in the Bering Sea (1993) and along the transpolar cruise (1994). (Jantunen and Bidleman 1995, Bidleman unpubl. 1996).

(Figures 6·23 and 6·24). Hargrave (1996) also found that toxaphene levels in surface water were highest during winter-spring, and decreased in summer-fall. The decline paralleled an increase in phytoplankton productivity in July and

elevated concentrations of particulate organic carbon which persisted in the water column through early fall. This suggests that toxaphene is scavenged from the water column by settling particles during and following the productive season. Fugacity calculations show that enhanced air-to-water gas exchange accompanies the draw-down in toxaphene surface water concentrations, thus demonstrating the close coupling between the atmospheric and biotic components of the system.

#### 6.6.4.1.1. Sea ice, suspended particulates, surface microlayers, and fog waters

Sea ice may entrain dissolved and particulate bound OCs from ocean water and the surface microlayer during freezing. Some OCs may be lost during freezing due to rejection of brines and other dissolved substances (chapter 3). Ice, in general, has lower or equivalent concentrations of all OCs in comparison to those observed in seawater. Gaul (1989) found that concentrations of HCH isomers were lower in sea ice than in seawater (Norwegian Sea), while PCB congeners, DDT-related compounds, and HCB were at much higher concentrations. Gaul also found PCB concentrations of 1-2.5 ng/L in melted sea ice, which was attributed to the contribution from particulates associated with the ice. Hargrave *et al.* (1988) did not detect PCBs, chlordanes, or any DDTs in sea ice (detection limits 2-23 pg/L). Vlasov and Melnikov (1990) found generally 20% lower levels of ΣDDT and PCBs in sea ice than in sea surface waters in nearshore samples from the Kara, Laptev, and East Siberian Seas.

In more recent work, Melnikov *et al.* (1996a, 1996b) reported HCH and DDT-related compounds, and ΣPCB (sum of seven congeners), in sea ice and snowcover from the Kara, Laptev, East Siberian, and Chukchi Seas (Figure 6·25). PCB concentrations in sea ice were lower than in overlying snow by 1.5 to more than three times (Annex Table 6·A12). However, volatilization of OCs from fallen snow may result in large concentration changes after deposition and prior to melting. PCB concentrations in surface seawater were higher than in ice or snow at three of four sites where results for OCs in ice, snow, and seawater were available from the same location (although samples were not collected at the same time). There were fewer differences between seawater and ice for ΣHCH or ΣDDT. Seawater/ice ratios for PCB and ΣDDT for the Yenisey and Ob Gulfs differed from the other locations. Water at these sites may be of lower salinity than other locations because of the influence of freshwater from rivers and levels of suspended sediments entrapped in the ice may be higher.

Melnikov *et al.* (1995) reported relatively high concentrations of five major OC groups in suspended particulate matter from Russian coastal waters. PCBs were the most prominent OCs with means ranging from < 0.1 to 5.2 μg/g dw (Annex Table 6·A11). Expressed on a volume basis, these concentrations are similar to those reported by Gaul (1989) for sea ice, that is, in the 0.5-5 ng/L range with SPM content of 1-10 mg/L. Joiris *et al.* (1995) found mean PCB concentrations (as Aroclor 1254) of 110 ng/g dw on SPM from the Barents Sea near Svalbard. Expressed on a volume basis (that is, dissolved + SPM), the PCB concentrations in Arctic sea waters (80-90°N) were 22 ng/L, about 2.54 times lower than in mixed North Atlantic/Barents Sea waters near Svalbard, and three times lower than in Atlantic waters (73°N 0°W).

In a survey of current-use pesticides in the Bering/Chukchi Seas, Chernyak *et al.* (1996) detected atrazine (400 pg/L) and chlorpyrifos (170 pg/L) in sea ice, but did not detect other semi-volatile pesticides such as endosulfan, trifluralin, and chlorothalonil. The source of the contaminants in sea ice was thought to be snow because both compounds would be efficiently scavenged from the atmosphere by precipitation. Concentrations of chlorpyrifos in surface waters decreased with distance from the ice fields, suggesting that this organophosphate was degrading in seawater following release from the ice or simply that the ice was the only major source of the pesticide. Pfirman *et al.* (1995) have pointed out that ice movement and melting at the ice edge could be a major pathway of transport of contaminants deposited by precipitation or dryfall/gas absorption or associated with epontic (under ice) algae.

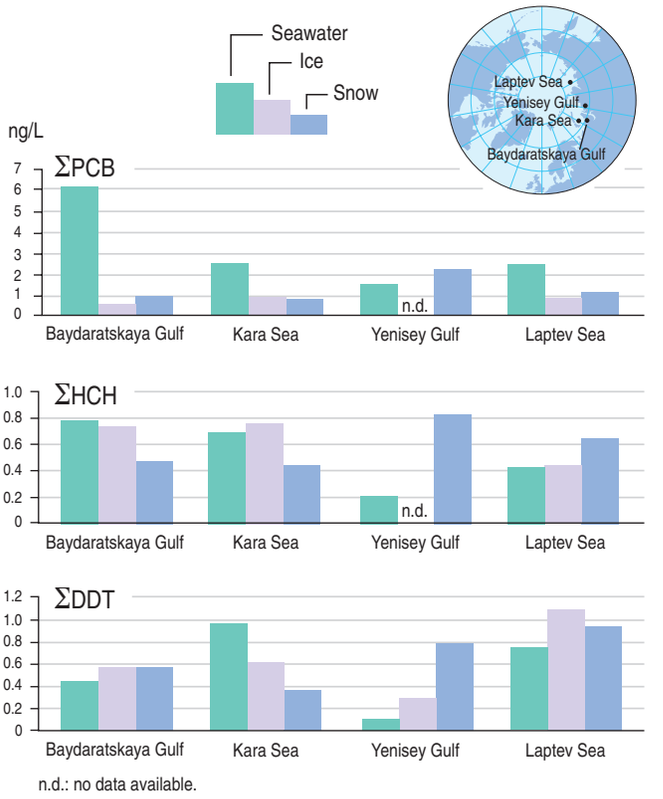



Figure 6·25. Concentrations of ΣPCB (sum of CBs 28, 52, 101, 110, 138, 153, and 180), ΣHCH, and ΣDDT in seawater, ice, and overlying snow collected at approximately the same locations in the Russian Arctic in 1993-95 (Melnikov *et al.* 1996a, 1996b).

Chernyak *et al.* (1996) made the first measurements of chlorinated pesticides in fog waters in the Bering/Chukchi Seas. Chlorpyrifos, an insecticide, and chlorothalonil, a widely used fungicide, were detected in almost all samples. Chlorothalonil was also detected in samples of the surface microlayer from the Bering and Chukchi Seas. Enrichment of these pesticides in fog and surface microlayers may reflect similar phenomena, i.e., the association of the organic contaminants with dissolved and particulate matter. Measurements of HCH isomers in surface microlayers from the Barents Sea (Chernyak *et al.* 1995) showed little enrichment for these relatively water soluble OCs. Additional measurements are needed to determine whether fog may represent a significant transport and deposition pathway of more hydrophobic chlorinated POPs and whether surface microlayers could be an important route of exposure for some Arctic biota.

### 6.6.4.2. Marine sediments

#### 6.6.4.2.1. PCBs and organochlorine pesticides

Organochlorine contaminants have been determined in a large number of surface grab samples of marine sediments from the Arctic Ocean and adjacent seas during the AMAP

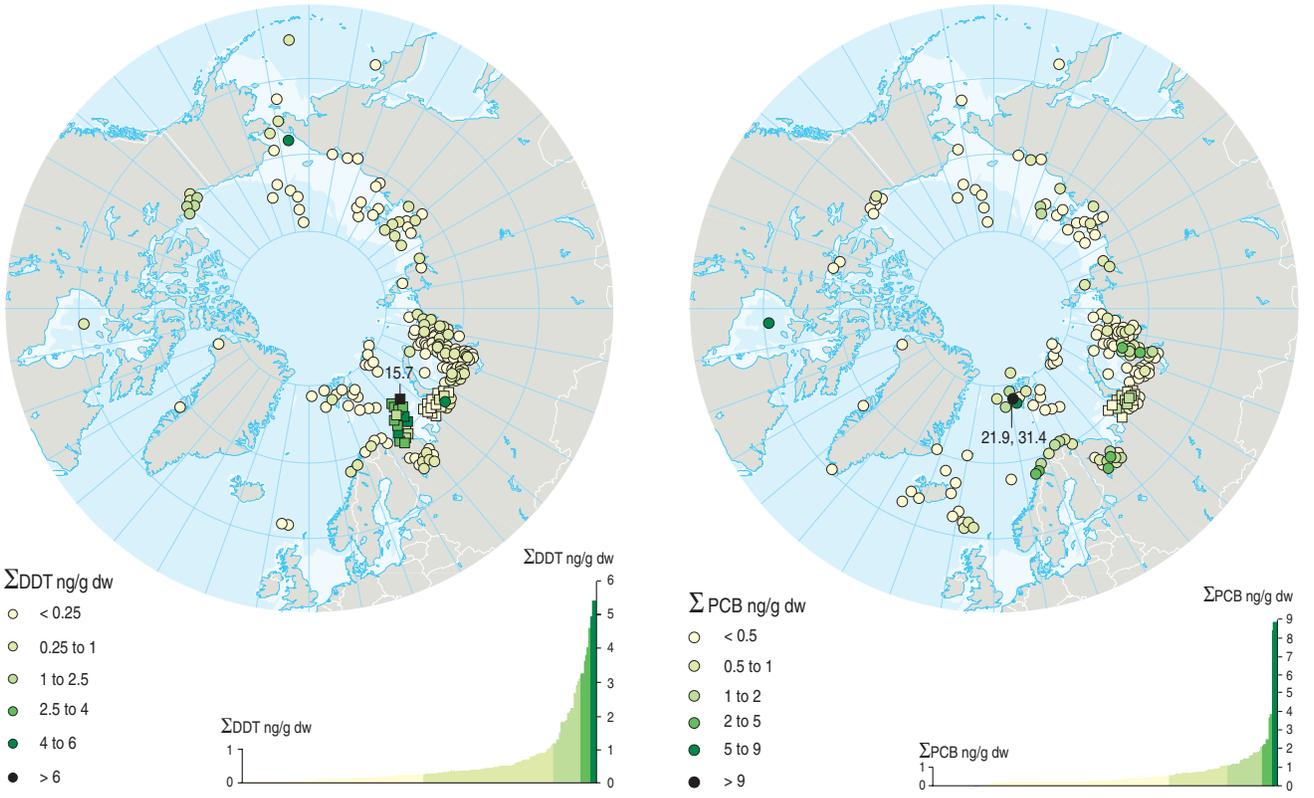



Figure 6·26. Concentrations of ΣDDT and ΣPCB in marine surface sediments (from grab samples or from top slices of cores) on a circumpolar basis, and the ranked distribution of values. Square symbols distinguish the dataset of Savinov and Savinova (reference 5 in Annex Table 6·A14), which has relatively high levels of ΣDDT and ΣHCH in samples from the Barents Sea. ΣDDT = sum of *p,p'*-DDE, -DDT, -DDD, and *o,p'*-DDT. ΣPCB represents the sum of 13 to 60 congeners, but all studies determined a common suite of 10 to 12 (CBs 18, 28, 52, 66/95, 101, 105, 110, 118, 138, 153/132, 170, 180).Full data set is in Annex Table 6·A14.

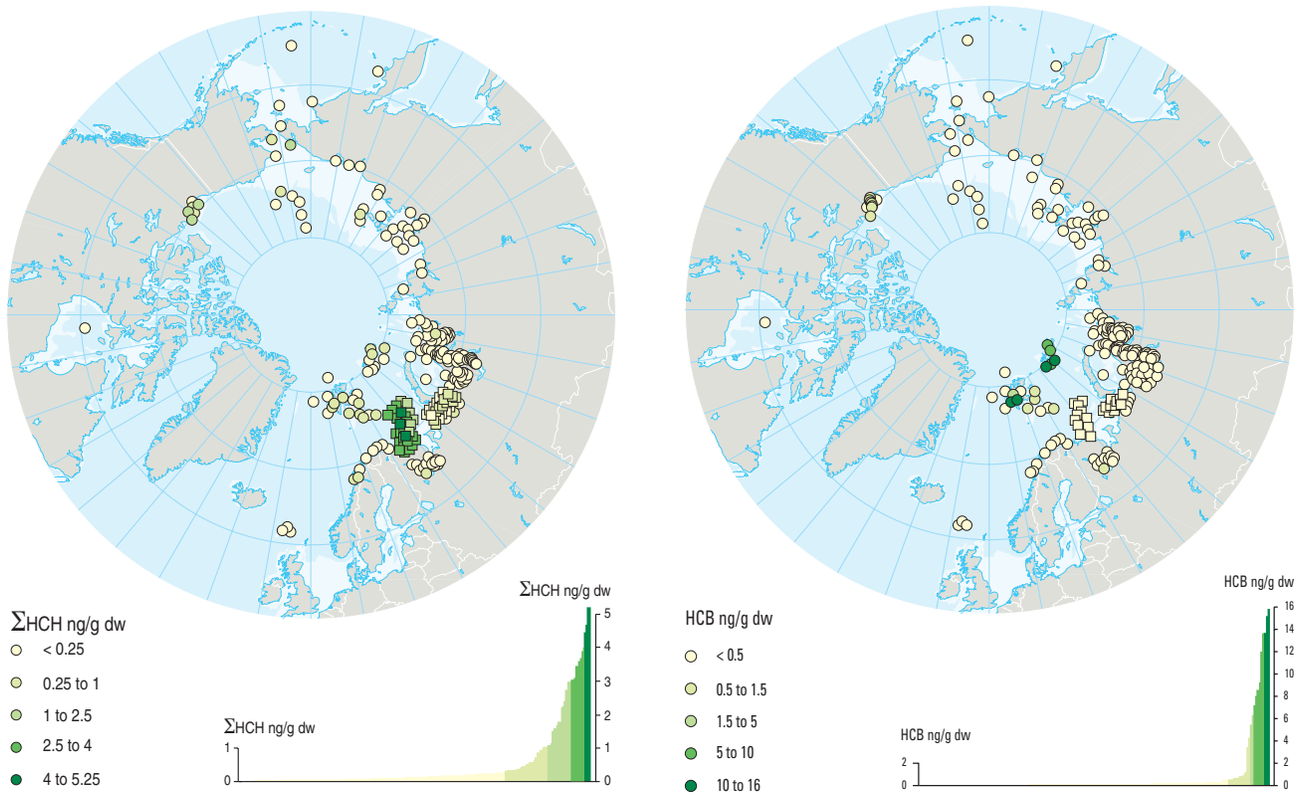



Figure 6·27. Concentrations of ΣHCH and HCB in marine surface sediments (from grab samples or from top slices of cores) on a circumpolar basis, and the ranked distribution of values. Square symbols distinguish the dataset of Savinov and Savinova (reference 5 in Annex Table 6·A14), which has relatively high levels of ΣDDT and ΣHCH in samples from the Barents Sea. Full data set is in Annex Table 6·A14.

program. Results for HCB and three major OC groups (ΣHCH, ΣDDT, and ΣPCB) are listed in Annex Table 6·A14 and plotted in Figures 6·26 and 6·27. Most samples are from nearshore areas in Norway, Russia, Greenland, the North American shelves of the Bering/Chukchi Seas, and the Beaufort Sea/Mackenzie River Delta area. All laboratories contributing data used capillary GC-ECD or GC-MS for determination of ΣPCBs and other OCs and, therefore, analytical results should be comparable. However, in the case of PCBs, various labs may have measured more congeners than

others, especially of lower chlorinated congeners, which contribute a significant fraction of ΣPCB in marine sediments. The data extracted from the ICES database were standardized to ten PCB congeners (CB 28, 52, 101, 105, 118, 153, 138, 180, 170, 206) while Melnikov *et al.* (1995) used seven congeners (CB 28, 52, 101, 118, 153, 138, 180). The results from the Beaufort/Chukchi Seas and High Arctic Ocean are based on the above ten congeners plus about 30-50 additional congeners. Intercomparison of results for HCB, HCH isomers, and *p,p'*-substituted DDT-related compounds is less of a problem because of the fewer components involved and the wide availability of analytical standards.

Spatial coverage of OCs in surface grab samples of marine sediments is relatively good in Norwegian and Russian waters, especially in the Barents Sea. The circumpolar distribution of ΣPCB and ΣDDT levels is indicated in Figure 6·26. In general, concentrations of all OCs in marine sediments are extremely low in comparison with freshwater sediments. Most sites have concentrations less than 1 ng/g dw. Data are lacking on OCs in marine sediments from the Canadian and Alaskan Arctic except for a limited number of samples from the southern Beaufort Sea and Bering Sea. Higher concentrations of ΣHCH, HCB, and ΣDDT were found in Mackenzie Delta sediments than in the Beaufort/Chukchi Seas, but ΣPCBs were present at similar levels.

A large data set for OC contaminants in sediment for the Russian Arctic is available as a result of studies by Melnikov *et al.* (1995) and Akvaplan-niva (dos Santos *et al.* 1996a, 1996b) (Annex Table 6·A14). ΣDDT, HCB, and ΣHCH are elevated in nearshore sediments from the White Sea, Pechora Sea, Baydaratskaya Gulf, and Khatanga Gulf relative to open ocean sites in the northern Barents and Kara Seas (Figures 6·26 and 6·27). ΣPCB concentrations range from 0.01 to 2.20 ng/g, but the majority are less than 1 ng/g dw. Levels of ΣHCH, HCB, and ΣDDT are also low, much lower than in Norwegian nearshore sediments or in the Mackenzie River Delta.

No geographic trend for ΣPCB and ΣDDT is apparent for offshore sites, that is those in the Beaufort/Chukchi Seas, and the northern Barents, Kara, and Laptev Seas (Figure 6·26). A distinct difference between offshore and nearshore sediments is evident at locations along the Norwegian Sea coast of Norway between Bergen (60°25'N, 5°E) and Tromsø (69°40'N, 19°E) (Annex Table 6·A14). Most of these locations are open areas near shore rather than in fjords, nevertheless, relatively high levels of ΣHCH (0.1-1.5 ng/g dw), ΣPCB (0.1-31.4 ng/g), and HCB (0.1-15.1 ng/g) were found at some of the sites. By comparison, open ocean sites in the Norwegian Sea (60-61°N and 3°E-7°W) had much lower levels of ΣHCH (0.01-0.18 ng/g), ΣPCB (0.35-1.13 ng/g), and HCB (0.10-1.15 ng/g). ΣDDT did not show a clear nearshore and open ocean trend. Higher levels of PCBs, HCH, and HCB at nearshore sites suggests possible influences from industrial and municipal effluents, however, none of these sites are in harbors. Given that HCH, HCB, and lower chlorinated CB congeners are the predominant OCs in precipitation and in the gas phase, it is possible that elevated levels in nearshore sampling sites may also be due to runoff and riverine flow from terrestrial drainage basins receiving only atmospheric inputs.

There have been fewer measurements of OCs in sediments of the Bering Sea. Rice *et al.* (1992) reported that HCH, PCBs, and DDT were the major OCs in upper slices of cores from four sites in the Bering/Chukchi Seas. Iwata *et al.* (1994a) measured relatively low ΣHCH concentrations in the Chukchi Sea and Bristol Bay (Bering Sea). Higher concentrations of ΣHCH were found in the Barents Sea and at some sites along the northern coast of Norway (1-5.2 ng/g dw; Annex Table 6·A14). In a highly productive region of the Chukchi Shelf, Macdonald *et al.* (1996) measured α-HCH concentrations in shallow-water surficial sediments as high as 0.23 ng/g, about ten times lower than in North American lakes dominated by atmospheric inputs (e.g., Lake Superior; Baker and Eisenreich 1990).

Bright *et al.* (1995c) found that sediments within Cambridge Bay harbor had higher concentrations of PCBs (0.14-45 ng/g dw, sum of 47 congeners) and a different congener pattern from those in two control sites (0.05-0.44 ng/g dw) about 60 km east and west of the harbor. Contamination of Cambridge Bay was attributed to improper disposal of PCBs from a military radar site. Sediments from control sites had higher proportions of lower chlorinated CBs indicative of atmospheric sources. This pattern may also exist along the Norwegian coast, however, a limited number of PCB congeners and a high frequency of non-detects makes the comparison of nearshore and open ocean sites difficult in this region.

#### 6.6.4.2.2. **PCDD/Fs**

Levels of PCDD/Fs have been determined in marine sediments from northern Norway (near Kirkenes), in the Mackenzie River Delta area, and in the Barents Sea. Oehme *et al.* (1993) analyzed surface sediments from the Barents Sea for PCDD/Fs and compared results with samples from the North Sea/Norwegian Sea. Concentrations of PCDD/F homologues in Barents Sea sediments are given in Annex Table 6·A21 and levels of OCDD and ΣPCDD/Fs at Barents Sea locations are compared with those in the North Sea in Figure 6·28. OCDD-concentrations are much lower in the Barents Sea than the North Sea. PCDD/F concentrations in the Barents Sea were low and fell within a relatively narrow range (12-32 pg/g dw for ΣPCDDs; 16-102 pg/g dw for ΣPCDFs). Non-2,3,7,8-substituted PCDD/Fs predominated. Average levels (11 sites) of 2,3,7,8-TCDF were 0.86 pg/g, while ΣTCDF averaged 14.1 pg/g. The proportion of more volatile PCDD/F congeners was higher in the Barents Sea than the North Sea, indicating a fractionation process (Oehme *et al.* 1993). Two samples closer to the Norwegian coast had higher levels of TCDD TEQs (1.1 and 0.72 pg/g) than other sites (0.31-0.63 pg/g) due to slightly higher levels of 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD than sites farther north. No correlations were found between PCDD/F levels and total organic carbon in the sediments.

Concentrations of PCDD/Fs in the Barents Sea were 10-20 times lower than those in the northern North Sea where TCDD TEQs ranged from 5.5 to 17.2 pg/g (eight samples; four locations). TCDD TEQs at one site in the Norwegian Sea between the Shetland Islands and Norway averaged 0.78 pg/g dw (n = 2), similar to levels in the Barents Sea.

Schlabach and Skotvold (1996a, 1996b) studied the levels and spatial distribution of PCDD/Fs in marine sediments collected from Bøkfjord near the Syd-Varanger smelter works in Kirkenes, in Arctic Norway and at a reference site (Heikenesset) on the Barents Sea coast. TCDD TEQs declined northward along the fjord from 2.5 pg/g dw near the smelter to 0.1 pg/g dw at the reference site. The PCDD/F congener profile at the reference site was dominated by PCDFs (ΣTCDF = ΣPeCDF = ΣHxCDF > OCDD > all other PCDD/F homologues) similar to observations in the Barents Sea (Oehme *et al.* 1993). PCDD/F isomer patterns were very similar for both the Barents Sea and North Sea samples and indicative of combustion sources. OCDD was the predominant congener, averaging 42% of ΣPCDDs at the 11 Barents Sea sites. Similar congener profiles were found in an unpolluted lake in Sweden (Kjeller *et al.* 1990) and in lakes in northern Finland (Vartiainen *et al.* 1997).

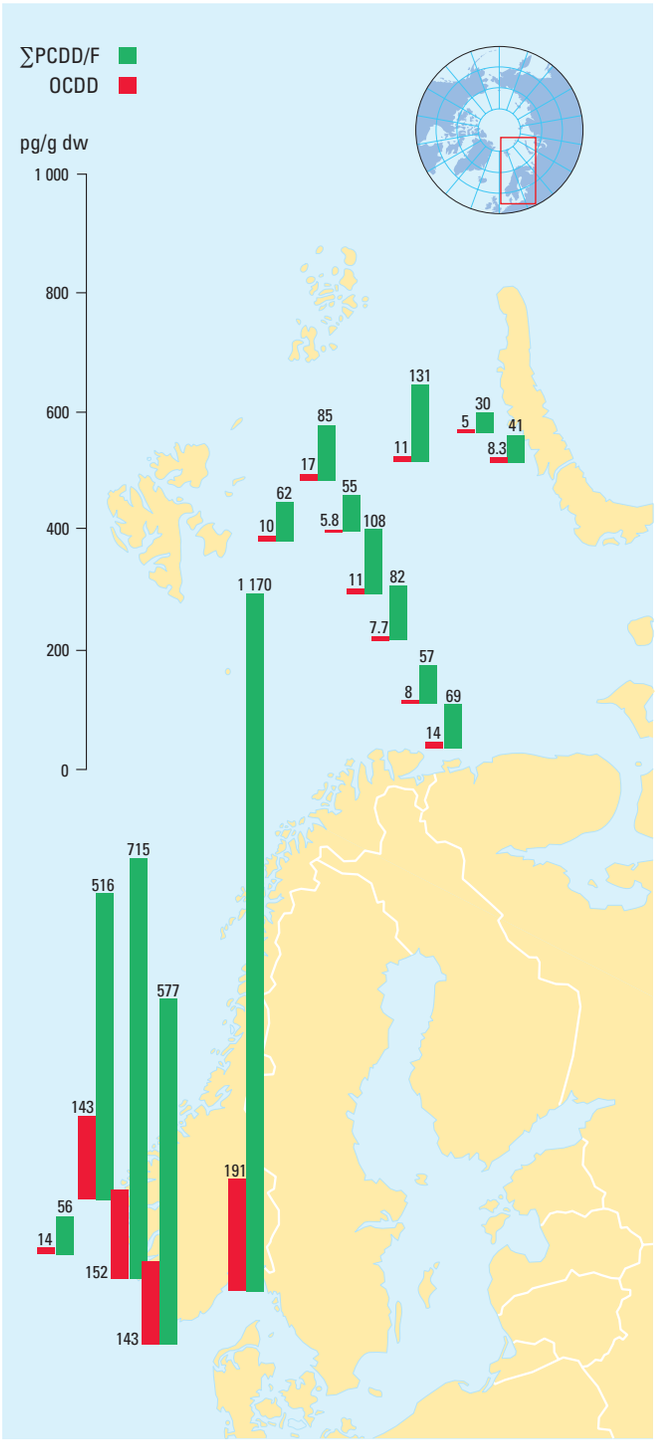



Figure 6·28. Concentration (pg/g dw) profiles for octachlorodioxin (OCDD) and total PCDD/Fs in marine sediments from southern and western Norwegian waters and from the Barents Sea (Oehme *et al.* 1993).

Macdonald (unpubl. data 1996) found a different PCDD/F pattern in sediments from the Eskimo Lakes (Husky Lake), estuarine lakes on the eastern edge of the Mackenzie River Delta, in which TCDD, PeCDD, and HxCDD congeners and OCDD predominated (Annex Table 6·A21). This pattern was quite different from PCDD/F patterns upstream in the Mackenzie River basin in Great Slave Lake. The proportion of OCDD found in the Eskimo Lakes sediments and in northern Norway and the Barents Sea was considerably lower than that found in Great Slave Lake (Evans *et al.* 1996) or mid-latitude lakes in the eastern USA (Czuczwa *et al.* 1984, Czuczwa and Hites 1986, Smith *et al.* 1993b). The reasons for this difference are unclear, but may reflect different combustion sources as well as greater contributions from chlorophenol use (which has a significant OCDD content) in North America compared with Europe.

#### 6.6.4.2.3. **Tributyltin**

There are few data on TBT concentrations in environmental media from the Arctic. Measurements made in coastal regions of northern Norway show sediment contamination in several harbors (Berge 1995). TBT concentrations are highly variable, as they are elsewhere, and are probably related to shipping activities and local rates of flushing and sedimentation.

### 6.6.4.3. **Mass balance modeling of organochlorines in the Arctic Ocean**

Constructing a mass balance for selected OCs helps to develop an understanding of the processes controlling the input and fate of POPs as well as to identify knowledge gaps critical to prediction of future trends. This has been attempted with four OCs: α- and γ-HCH, toxaphene, and PCBs (Barrie *et al.* 1992). Less information is available for the latter two compounds. As discussed in sections 6.6.1.1 and 6.6.4.1, HCHs are the most abundant pesticides in Arctic air and water. Toxaphene has air–water partitioning characteristics similar to the HCHs and would be expected to follow similar transport pathways. The difference between HCHs and less volatile, more hydrophobic OCs such as PCBs, chlordanes, DDTs, and toxaphene, is that the fate of the latter substances is influenced to a greater extent by wet and dry deposition of particles from the atmosphere and sedimentation in the water column. Barrie *et al.* (1992) formulated a mass balance for HCHs in the Arctic Ocean using air and water concentrations typical of the mid-1980s. The budget was based on limited data for HCHs in ocean water and rivers, and assumed that atmospheric levels remained constant over the year. Deposition of HCHs was calculated for two seasons, winter and summer. The estimated standing stock of HCHs in the upper 200 m of the water column was 8100 tonnes with a residence time of 20-30 years. Major inputs to the Arctic Ocean were by ocean currents (63%), atmospheric deposition (30%), and river runoff (7%). Losses were mainly by outflow of water to the North Atlantic through the Canadian Archipelago (78%), the East Greenland Current (16%), and to a lesser extent by other currents (4%) and ice export (2%).

The HCH budget of Barrie *et al.* (1992) has been updated here and extended to toxaphene and PCBs, taking into account advances in knowledge by: 1) using atmospheric concentrations measured in the early 1990s; 2) estimating atmospheric fluxes on a monthly or quarterly basis rather than biannually; 3) incorporating new data on the spatial variability of HCHs, toxaphene, and to a limited extent PCBs in the Arctic Ocean; 4) making use of more detailed information on the circulation, ice cover, and water budget of the Arctic Ocean; and, 5) updating estimates on the input of HCHs and PCBs (but not toxaphene) from rivers (especially in Russia).

For purposes of the budget, the 'Arctic Ocean' is defined as selected waters above 65°N, namely, the Canada and Eurasian Basins, the Canadian Archipelago, and the following regional seas: Barents, Kara, Laptev, East Siberian and Chukchi. Excluded are Baffin Bay, Hudson Bay, the Bering Sea, the Norwegian Sea, and the Greenland Sea. Even though portions of these waters lie above 65°N, they are not included because their circulation patterns place them outside of the Arctic Ocean regime. The Arctic Ocean was partitioned into two domains; the 'North American Arctic Ocean' (NAAO = $2.57 \times 10^6$ km$^2$), which includes the Canada Basin

and the Archipelago and accounts for approximately 25% of the total ocean area, and the 'Eurasian Arctic Ocean' (EAO = $7.72 \times 10^6$ km$^2$), which includes the Eurasian Basin and the regional seas.

The net exchange direction of gaseous OCs between air and sea is controlled by the fugacity (partial pressure) in surface water relative to air. The water/air fugacity (F) ratio is calculated (McConnell *et al.* 1993):

$$F = f_w / f_a = C_w H / C_a R T_a$$

where $f_w$ and $f_a$ are the fugacities of water and air, $C_w$ and $C_a$ are the dissolved and gaseous OC concentrations in water and air (ng/m$^3$), H is the Henry's Law constant at the water temperature (Pa · m$^3$/mol), $T_a$ is the air temperature (K), and R = 8.314 Pa · m$^3$/mol K.

The framework for the atmospheric portion of the budget and descriptions of the process parameters are presented by Barrie *et al.* (1992) and Cotham and Bidleman (1991). Deposition mechanisms include: 1) rain and snow scavenging of particulate OCs (associated with haze aerosols), 2) dry deposition of particulate OCs, 3) rain scavenging of vapor-phase compounds, 4) adsorption of vapors to falling snow, and 5) air-to-sea gas exchange. Processes which release OCs to the atmosphere from within the Arctic are: 6) volatilization from ocean surface water, and 7) volatilization from ice and snow. The equations used to calculate these air-surface fluxes as well as the values of process parameters required for the calculations are given in Annex Table 6·A13. The fraction of each OC on particles was estimated from the Junge-Pankow adsorption model (see section 6.2.1.1), using the subcooled liquid vapor pressure as a function of temperature. Details of these calculations are given by Cotham and Bidleman (1991). The atmospheric budget takes into account monthly (averaged quarterly) variations in the atmospheric concentration of OCs, haze aerosols, air temperature, precipitation, and the area of the Arctic Ocean that is ice-free. The total area used for calculating loadings by wet and dry deposition is $1.03 \times 10^7$ km$^2$.

Gas exchange fluxes were converted into the mass of each OC depositing or volatilizing by assuming values for the area of the ocean that is unfrozen and available for gas exchange. The extent of sea ice cover and area of open water within the ice pack has been measured by a multichannel microwave radiometer from the NASA Nimbus-7 satellite operated from 1978-1987 (Gloersen *et al.* 1992). Their estimates of the proportion of the entire Arctic Ocean that is ice-free ranged from 11% in February to 64% in August. These figures are heavily weighted by the large areas of open water in the regional seas, and we used them for the EAO. The ice cover in the Canada Basin is greater. Satellite imagery for 1987-1990 in the Beaufort Sea shows greater than 90% cover in January-February and 10-60% in August (LeDrew *et al.* 1992). Satellite data collected in August-September 1994 in connection with the AOS-94 cruise show 10-50% ice cover between 70-75°N and 90-100% at higher latitudes. The open water in the NAAO was assumed to be 5% and 25% of the total area in February and August, respectively. Linear changes in ice cover were taken for the summer-winter transitions in the EAO and NAAO.

The budget for each OC was constructed using the volumetric flows for freshwater, seawater, and ice into and out of the Arctic Ocean (chapter 3), together with the respective estimated concentrations of each compound (Table 6·16). Details of the new budget calculations are presented for each chemical group in the following sections.

#### 6.6.4.3.1. Hexachlorocyclohexane mass balance in the Arctic Ocean and regional seas

HCH compounds volatilize soon after application, especially in the tropics (Takeoka *et al.* 1991), and are atmospherically transported to the Arctic and other remote regions. The low Henry's Law constants of HCHs favor partitioning from air into water, especially at low temperatures. At equilibrium, the ratio of α-HCH concentration in seawater to that in air is 3000 at 25°C, but 22 000 at 0°C. The world's oceans are the major reservoir of HCHs. Global models estimate that approximately 20% of the HCH present in the environment is held in the ocean surface layer, which is taken to be the upper 75 m (Strand and Hov 1996) or 200 m (Mackay *et al.* 1995). Even though the heaviest use of HCHs has been in tropical and subtropical regions, levels in surface seawater are an order of magnitude higher in the Arctic (Iwata *et al.* 1993, Schreitmüller and Ballschmiter 1995). Concentrations of HCHs in tree bark increase with latitude, being relatively high in Canada, Nordic countries, Alaska, and Russia, and lower in tropical countries (Simonich and Hites 1995). Thus, the transport and distribution of HCHs exemplifies the 'cold condensation' effect (Wania and Mackay 1993, 1995).

##### 6.6.4.3.1.1. Budgets for water and dissolved HCHs

The results of the calculations and the HCH sources data are summarized in Table 6·15. One of the problems identified in constructing this budget is that there are no surveys which include measurements of HCH in all the important water masses within a time frame of a few years. Although HCH concentrations are now available for the regional seas in the Russian Arctic, recent values for the Eurasian Basin are especially lacking. The values selected for Table 6·15 incorporate data from Table 6·15 and other sources from the past decade. The burden of HCH estimated from available vertical profile data (Figure 6·22) is 2200-2600 tonnes of α-HCH and 280-420 tonnes of γ-HCH for the NAAO, and 2200-2400 tonnes of α-HCH and 500-630 tonnes of γ-HCH for the EAO.

For the removal of HCHs by ice flow through the Canadian Archipelago and Fram Strait, we have relied on the measurements of HCH levels in ice from the Russian coastal waters (Melnikov *et al.* 1996a, 1996b). Representative HCH concentrations for the Bering Sea have been determined (Jantunen and Bidleman 1995) which, when combined with reliable estimates of inflow through the Bering Strait, makes this the best-determined component of the HCH budget. Estimates of riverine inputs of HCHs are uncertain, particularly for the Russian rivers. Jeffries *et al.* (1994) measured a fairly uniform, low range of HCH concentrations in the Mackenzie River and eleven other small Canadian Arctic rivers. In contrast, average HCH concentrations for the Russian rivers in 1992-1993 (Roshydromet 1995) were well above those found in the North American rivers (Figure 6·13). In the budget, we have used 1994 α- and γ-HCH concentrations of 5 and 2 ng/L, respectively (the range is 0-55 ng/L), to take into account the high levels in Russian rivers. Our estimation of the exchanges between the Atlantic and Arctic Oceans is based exclusively on the data of Chernyak *et al.* (1995). Lacking data for HCH in the Norwegian Coastal Current, the results from Gaul (1989) were used which are based on measurements in 1985. These latter measurements are higher than levels reported for the Barents and Kara Seas by Chernyak *et al.* (1995) and Melnikov *et al.* (1995). For the East Greenland Current, we have partitioned the water column into two domains (Polar Water and Return Atlantic Water) (Foldvik *et al.* 1988) and have chosen HCH values

Table 6·15. Arctic Ocean input/output budget for HCH isomers and preliminary estimates for toxaphene and PCBs[a]. Standard deviations (SD) calculated from recent measurements (see Table 6·14). SD for model results calculated for seawater flows only, assuming variance is due to the contaminant measurement.

| | Water flow, $10^4$ km$^3$/year | Water concentration, µg/m$^3$ or ng/L | | | | | | | | Flux, tonnes/year | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | α-HCH | SD | γ-HCH | SD | Toxaphene | SD | ΣPCB | SD | α-HCH | SD | γ-HCH | SD | Toxaphene | SD | ΣPCB | SD |
| *Input* | | | | | | | | | | | | | | | | | |
| Bering Strait | 2.6 | 2.0 | 0.48 | 0.45 | 0.10 | 0.025 | – | 0.01 | – | 52 | 12.4 | 12 | 3 | 0.65 | – | 0.3 | – |
| Norwegian Coastal Current | 2.2 | 2.0 | 0.30 | 0.45 | 0.20 | 0.08 | – | 0.025 | – | 44 | 6.6 | 9.9 | 4.4 | 1.76 | – | 0.6 | – |
| Barents Sea | 3.8 | 1.0 | 0.09 | 0.25 | 0.04 | 0.08 | – | 0.025 | – | 37.8 | 3.40 | 9.5 | 1.5 | 3.0 | – | 0.9 | – |
| W. Spitsbergen Current | 6.3 | 1.0 | 0.09 | 0.25 | 0.04 | 0.08 | – | 0.025 | – | 63 | 5.67 | 15.8 | 2.5 | 5.04 | – | 1.6 | – |
| Rivers: N. America | 0.03 | 0.5 | 0.50 | 0.1 | 0.10 | 0.15 | – | 0.5 | – | 0.15 | 0.15 | 0.03 | 0.03 | 0.045 | – | 0.15 | – |
| Rivers: Eurasian | 0.3 | 5.0 | 5.00 | 2.0 | 2.00 | 0.10 | – | 5.0 | – | 15 | 15 | 6.00 | 6 | 0.30 | – | 15 | – |
| Gas deposit. – EAO[b] | | | | | | | | | | 34 | | 5.62 | | 7.28 | | 38.6 | |
| Dry fall – EAO | | | | | | | | | | 0.06 | | 0.04 | | 0.85 | | 6.21 | |
| Wet fall – EAO | | | | | | | | | | 0.77 | | 0.20 | | 0.57 | | 2.29 | |
| Gas deposit. – NAAO[b] | | | | | | | | | | 4.50 | | 0.75 | | 0.96 | | 2.05 | |
| Dry fall – NAAO | | | | | | | | | | 0.02 | | 0.01 | | 0.28 | | 0.89 | |
| Wet fall – NAAO | | | | | | | | | | 0.25 | | 0.07 | | 0.19 | | 0.32 | |
| *Total* | | | | | | | | | | 247 | | 59 | | 20 | | 66 | |
| *Output* | | | | | | | | | | | | | | | | | |
| East Greenland Current (Polar water) | 3.2 | 0.90 | 0.20 | 0.20 | 0.04 | 0.08 | – | 0.025 | – | −28 | 7 | −6.3 | 10 | −2.5 | – | −0.8 | – |
| East Greenland Current (Return Atlantic water) | 7.3 | 0.70 | 0.20 | 0.16 | 0.02 | 0.08 | – | 0.025 | – | −51 | 13 | −12 | 20 | −5.8 | – | −1.8 | – |
| Can. Archipelago | 5.4 | 4.5 | 0.50 | 0.60 | 0.12 | 0.09 | – | 0.01 | – | −241 | 47 | −32 | 45 | −4.8 | – | −0.5 | – |
| Ice | 0.52 | 0.50 | 0.50 | 0.20 | 0.20 | 0.05 | – | 0.038 | – | −2.6 | 3 | −1.0 | 1.0 | −0.3 | – | −0.2 | – |
| Volatilization – EAO | | | | | | | | | | −14.9 | | −1.2 | | −1.2 | | −0.7 | |
| Volatilization – NAAO | | | | | | | | | | −49.8 | | −6.7 | | −0.6 | | −18.9 | |
| Hydrolysis | | | | | | | | | | −55 | 14 | −6.0 | 10 | 0.0 | – | 0.0 | – |
| Sedimentation | | | | | | | | | | −0.06 | | −0.03 | | −0.03 | | −0.15 | |
| *Total* | | | | | | | | | | −443 | | −65 | | −15 | | −23 | |
| *Net input/output* (negative = output) | | | | | | | | | | −196 | | −6 | | 4 | | 43 | |

a. See Annex Table 6·A13 for details on calculation of each input and output process.
b. EAO = European Arctic Ocean; NAAO = North American Arctic Ocean.

based on recent measurements from the transpolar cruise (Figure 6·23).

As discussed in section 6.6.4.2, there are insufficient data at present to make a reliable determination of HCH flux to sediments as part of the budget. HCHs are neither very particle-reactive nor bioaccumulative and, therefore, are often below detection limits in suspended particles or in sediments (e.g., Hargrave *et al.* 1988, 1989b). Recent fluxes of ΣHCH in a core from the Gulf of Alaska were estimated to be 61 ng/m$^2$/y based on a sedimentation rate of about 1 mm/y (Iwata *et al.* 1994b). Assuming this rate is typical of the NAAO and EAO, this represents about 0.5 tonnes/y deposition of ΣHCH to sediments for the entire Arctic Ocean. This may be a high estimate. Net sedimentation rates in the Arctic Ocean are generally much lower than 1 mm/y. Average sedimentation rates for Ice Island site (7.3 g/m$^2$/y; Hargrave *et al.* 1989b) yield a sedimentation rate of α-HCH of < 0.7 ng/m$^2$/y or 0.008 tonnes/y extrapolated to the entire Arctic Ocean. For the mass balance calculations, we assumed a net sedimentation of 10 g/m$^2$/y in pelagic areas of the NAAO and EAO and 100 g/m$^2$/y in shelf areas. Even with this modification, sedimentation and burial of HCH isomers is negligible in comparison to the other values in Table 6·16. Based on these lines of reasoning, it appears unlikely that sedimentation plays much of a role in the overall HCH budget for the Arctic Ocean, even though there may be locations where the vertical fluxes are enhanced by high primary productivity.

The hydrolysis half-lives of α-HCH and γ-HCH in seawater at pH 8.1 are estimated to be 0.4 and 0.5 years at 25°C, but increase to 60 and 100 years at 0°C, using the hydrolysis rate constants derived by Ngabe *et al.* (1993). The fraction of the initial concentration that is hydrolyzed in one year is 0.0110 for α-HCH and 0.0064 for γ-HCH. Taking the surface layer burdens to be 5000 tonnes of α-HCH and 1000 tonnes of γ-HCH, hydrolysis accounts for annual losses of 55 tonnes of α-HCH and 6 tonnes of γ-HCH, amounts comparable to removal by other processes such as water outflows and volatilization (Table 6·15).

The first evidence that OC pesticides can be microbially degraded in Arctic waters has recently been obtained from the changing proportion of α-HCH enantiomers (optical isomers) (Falconer *et al.* 1995a, 1995b). Selective breakdown of (+)α-HCH was found in the Canada Basin and the Greenland Sea, with ERs for surface water ranging from 0.75-0.93. Loss of (+)α-HCH increased with depth, and the level of this enantiomer was near the detection limit below 750 m (Jantunen and Bidleman 1995). Water samples from the Bering and Chukchi Seas were depleted in (–)α-HCH, an opposite degradation preference from the Arctic Ocean (Figure 6·23). Reasons for this are not known, but may be related to different microbial populations in these regions. The α-HCH in air was also depleted in the (+) enantiomer for samples collected over the Arctic Ocean and the Greenland Sea, and in the (–) enantiomer over the Bering/Chukchi Seas (Figure 6·23). The appearance of the enantiomeric profile for surface water in the overlying air provides evidence of sea-to-air volatilization (see below). Because the degradation rates for this breakdown in seawater are currently unknown, this pathway was not included in the HCH budget.

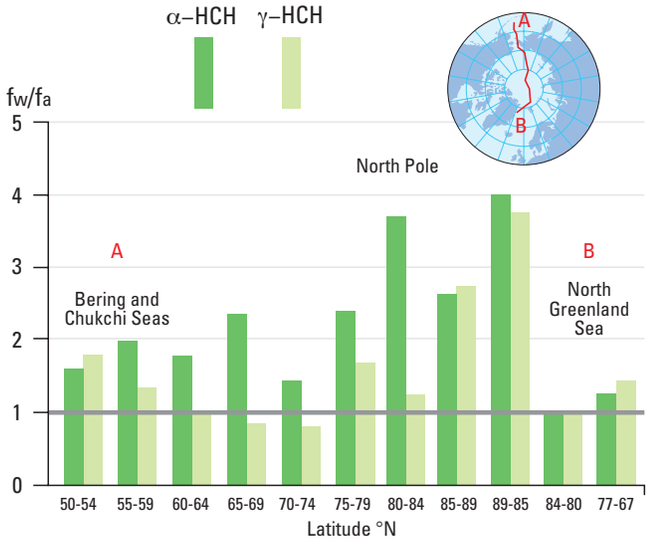



Figure 6·29. Water/air fugacity ratios (fw/fa) of α-HCH and γ-HCH on a transect from the Bering Sea to the Greenland Sea in July and August 1994. A fugacity ratio of 1 indicates air–water equilibrium. A ratio different than 1 indicates the potential for net deposition to, or volatilization from the ocean.

### 6.6.4.3.1.2. The atmospheric HCH budget

In the 1980s, mean fugacity (F) values of α- and γ-HCHs were 0.74 and 0.28, respectively, in the Arctic Ocean, and 0.76 and 0.55 in the Bering/Chukchi Seas. That is, the surface water was 28-76% saturated with respect to the partial pressure of HCHs in air. At that time, air-to-water deposition of gaseous HCHs was estimated to have contributed 80% of atmospheric loadings, or 63 tonnes/y, to the Arctic Ocean (Cotham and Bidleman 1991, Barrie *et al.* 1992). The more recent (1992-1994) F values, measured in the Canadian Archipelago (Falconer *et al.* 1995a), in the Bering/Chukchi Seas (Jantunen and Bidleman 1995), and on the transpolar cruise in 1994 (Jantunen and Bidleman unpubl. data 1996), were more than 1.0 at most stations (Figure 6·29). Lower F values were calculated for the EAO, where HCH concentrations are lower than in the Canadian Archipelago and Beaufort Sea. Average F values for α-HCH for the EAO ranged from 1.0-1.8 based on water concentrations of 2 ng/L and on air concentrations of 58-90 pg/m$^3$. This implies that waters throughout the Arctic Ocean are now oversaturated and outgassing HCHs to the atmosphere when not ice-covered.

Although the uptake and release of vapor-phase HCHs by snow and ice is expected to be significant (Hoff *et al.* 1995, Wania and Mackay 1995), we have not included these mechanisms in the atmospheric budget. The specific surface area of fresh snow is highly variable and changes by two orders of magnitude as the snow ages (Hoff *et al.* 1995). This large uncertainty precludes quantitative estimates of these processes. Furthermore, the revolatilized compound could be deposited into the water column by gas absorption. Thus, while omitting the snow scavenging pathway underestimates deposition to the ice surface, it prevents possible double accounting for inputs. Based on recent surveys of HCHs in surface seawater (Table 6·15, Figures 6·22 and 6·23), the concentrations in the Canada Basin and the Canadian Archipelago are about twice those in the Eurasian Basin and the regional seas.

Because HCHs are relatively volatile, even at Arctic temperatures, sorption to haze aerosols does not play a large part in their environmental distribution, and inputs by dry deposition are small (Table 6·16). Percentages sorbed to particles were highest during March, reaching 2% for α-HCH and 6% for γ-HCH.

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

### 6.6.4.3.1.3. Overall HCH budget for the Arctic Ocean

The mass budget of HCH in the Arctic is illustrated in Figure 6·30. Gas exchange is a dominant process, accounting for 88% of the atmospheric budget for α-HCH and 81% for γ-HCH. Loadings of α-HCH to the Arctic Ocean are 1.0 tonnes/y by precipitation, 0.1 tonnes/y by dry particle deposition, and 39 tonnes/y by air-to-sea gas transfer. These inputs are offset by –65 tonnes/y volatilization, and the net result is an output of about 196 tonnes/y. However, on a regional basis, volatilization is higher in the NAAO than the EAO due to the substantially higher α-HCH concentrations in the surface waters of the Canada Basin and the Canadian Archipelago (Figure 6·22). In addition, the high percentage of ice cover in the NAAO suppresses outgassing.

Inputs of γ-HCH by precipitation and dry particle deposition are 0.27 and 0.05 tonnes/y. Deposition and volatilization of vapor-phase γ-HCH account for 6.3 and –8 tonnes/y, respectively. Thus, the atmospheric budget for γ-HCH is close to steady state. For HCH (α- + γ-HCH) in the early 1990s, there is net output of 202 tonnes/y. By comparison, Barrie *et al.* (1992) estimated the net loading of α- + γ-HCH in the 1980s at 79 tonnes/y, 80% of which was by gas-phase deposition.

Ocean currents provide 79% of the total HCH input, with atmospheric deposition and rivers contributing 15% and 6%, respectively. Although HCH inputs from Russian rivers, based on recent measurements, were set ten times higher than from North American rivers, their influence on

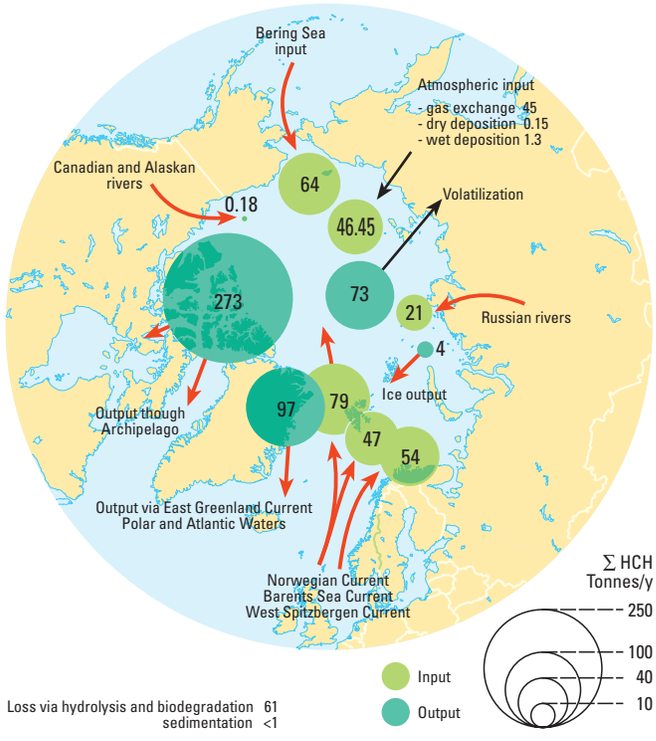



Figure 6·30. ∑HCH budget for the Arctic Ocean (tonnes/y).

the overall Arctic Ocean HCH budget is small. Outflow of HCHs occurs mainly through the Canadian Archipelago (52%) and the East Greenland Current (16%). Volatilization and ice export account for 14% and 1% of the loss, respectively. The only chemical loss quantified in this exercise was hydrolysis, estimated to account for 12% of the removal of total HCHs. However, the enantiomer data suggest that microbial breakdown may also be an important, as yet unquantified, sink.

It is likely that loadings of HCHs to the Arctic Ocean from the atmosphere as well as ocean currents were greater in the past, and were dominated by technical HCH mixtures that contained a high proportion of the α-isomer. The picture that emerges from this mass balance exercise is of the Arctic Ocean in steady state with γ-HCH and exporting α-HCH. Inputs and outputs of HCHs are dominated by ocean current advection, with about a 14% contribution by atmospheric processes, largely air-sea gas exchange (Figure 6·30). In the future, we can expect to see a decline in the large pool of α-HCH in the Canada Basin due to drainage out through the Canadian Archipelago and to a lesser extent by sea-to-air volatilization. Outgassing will also result in a slow decline of both HCH isomers in waters of the Eurasia Basin and the regional seas, assuming that atmospheric concentrations continue their downward trend.

#### 6.6.4.3.2. **Toxaphene mass balance in the Arctic Ocean and regional seas**

Processes of atmospheric deposition and volatilization of toxaphene were considered, using the framework developed for HCHs (section 6.6.4.3.1, Table 6·15). Because almost all toxaphene measurements have been made in the NAAO, the mass balance modeling was limited by lack of data for the EAO.

Since measurements of toxaphene in Arctic air and water have been made on a 'total toxaphene' basis, rather than of individual chlorinated bornane components, average properties for the toxaphene mixture must be used. The vapor pressure and Henry's Law constant of technical toxaphene measured at 20°C by Murphy *et al.* (1987) were extrapolated to Arctic temperatures (0°C for water) using equations for subcooled liquid vapor pressure versus temperature (logarithm $VP_L = 12.25 – 4487/T$) for two hepta- and octachlorobornanes (Hinckley *et al.* 1991) and for Henry's Law constant versus temperature (logarithm $H = 11.48 – 3416/T$) based on the work of Tateya *et al.* (1988) for PCBs. These equations were also used by Hoff *et al.* (1993) to estimate the atmospheric deposition of toxaphene to Lake Superior.

As discussed in section 6.6.1.1, current estimates of toxaphene concentrations in Arctic air differ by about a factor of 3. For mass balance purposes, airborne toxaphene concentrations adjusted to the NIMS scale were used (Table 6·6). Dry deposition was assumed to be highest during winter haze season, when about 90% of the toxaphenes are predicted to be associated with aerosols according to the Junge-Pankow model. The contribution of precipitation is fairly constant over the year due to particle scavenging during winter and gas scavenging during summer.

F values for toxaphene, based on air concentrations of 7.6-20 pg/m$^3$ and water concentrations of 80 pg/L in the NAAO and 40 pg/L in the EAO, range from 0.1 to 6. Low concentrations of toxaphene in air during winter leads to the higher F values. Little outgassing takes place during winter because of the small area of open water available for exchange. During summer, the atmospheric concentrations are higher, fugacity ratios are well below 1.0 (net deposition), and gas exchange is maximized because of the expanded area of open water.

Net atmospheric loadings of toxaphene were estimated to be 5 tonnes/y. With an uncertainty in atmospheric concentrations of about a factor of 3, the estimate of net loadings is also highly uncertain because the net input is primarily due to gas exchange, which responds to the water/air fugacity ratio. Other factors, such as limitations in icecover data, compound the uncertainty in the toxaphene loadings calculations and, at the present time, it seems fair to say that atmospheric loadings of toxaphene can be expressed to no better than an order of magnitude.

Overall, atmospheric inputs may represent 46% of total inputs of toxaphene. Volatilization is also an important loss process (11%), although export via the Canadian Archipelago and the Greenland Current account for most of the removal of toxaphene from the Arctic Ocean. These export fluxes are reasonably certain because they are based on recent measurements of toxaphene in seawater near Resolute, NWT, Canada (Table 6·15) and in the Greenland Sea. There is much greater uncertainty about input fluxes via ocean currents because of the absence of toxaphene data for seawater in the West Spitsbergen, Barents Sea, and Norwegian Coastal Currents. Sedimentation was estimated by assuming a surface sediment concentration for toxaphene of 0.1 ng/g, similar to concentrations found in some Arctic lakes (Muir and Lockhart 1994). There appears to be no data on toxaphene in marine sediments. The small flux to sediments predicted in the mass balance is consistent with the physical properties of toxaphene. Like other chlorinated aliphatics, such as HCH, most chlorobornanes are less hydrophobic than chlorinated aromatics of similar molecular weight, such as PCBs, and are thus probably not scavenged to the same extent by sinking particles. Hydrolysis and biotransformation of toxaphene in seawater were assumed to be negligible. Both these processes could be important for removal of toxaphene, but values for this OC are largely unknown in seawater.

The quantity of loadings of toxaphene from rivers is also largely unknown. A tentative value of 0.15 ng/L was used, based on mean concentrations of recent measurements of lake surface waters in NWT and Yukon, Canada (Table 6·12). A toxaphene level of 0.1 ng/L was used for Eurasian rivers based on Kucklick *et al.* (1994) who found mean concentrations of 0.064 ng/L in Lake Baikal surface waters which ultimately drain into the Arctic Ocean via the Yenisey. Overall riverine inputs of toxaphene are low, representing about 10% of all inputs, and are outranked by inputs from gas deposition and ocean currents.

Although the modeling results are highly uncertain, they suggest that inputs and outputs of toxaphene to the Arctic Ocean are roughly in balance. Further measurements, especially of toxaphene concentrations in EAO waters, are critical to making more accurate budgets.

#### 6.6.4.3.3. **PCB mass balance in the Arctic Ocean and regional seas**

Unlike the situation for toxaphene, there are numerous measurements of PCBs in surface waters, air, and sediments. However, mass balance estimates for ΣPCBs are confounded by lack of measurements of seawater concentrations at key locations and by order of magnitude differences in concentrations in river water SPM and in seawater between North American/Norwegian and Russian waters. There is very limited information on PCB congener profiles in seawater although substantial information is available on air concentrations of individual congeners. Thus, a total PCB approach

was used to conduct a preliminary mass balance. Average properties for tetra- and pentachlorobiphenyls, which predominate in air and seawater, were used for vapor pressure and Henry's Law constant (H). Extrapolations of physical property data (generally at 20-25°C) to Arctic temperatures (0°C for water) for subcooled liquid vapor pressure versus temperature were made using slopes for toxaphene and for Henry's Law constant (logarithm H = 11.48 – 3416/T) based on the work for PCBs by Tateya *et al.* (1988).

Based on monthly ΣPCB averages for 1993 for air at Alert (NAAO) and Ny-Ålesund (EAO), and water concentrations of 10 pg/L, F values range from 0.1-4, with lowest levels during the summer months when air concentrations are generally lower. Prevailing PCB concentrations in air are well documented (although not collected simultaneously nor at the same location as water sampling), thus, uncertainty in the direction of flux is related to water concentrations. Much higher F values would result if PCB results from the Russian seas were used (Annex Table 6·A10). However, these values are difficult to reconcile with much lower results given in Chernyak *et al.* (1995) for the Barents Sea (38 pg/L), Hargrave *et al.* (1988) at the Canadian Ice Island (7 pg/L), or Iwata *et al.* (1993) in the Chukchi Sea (6.6-9.3 pg/L). Therefore, we used 10 pg/L as an ocean-wide average and 25 pg/L for inflowing waters of the EAO (Table 6·16). Overall atmospheric deposition of PCBs via dryfall, gas exchange, and precipitation was estimated to equal volatilization. Outgassing of PCBs would be expected to be especially important in the Russian seas, assuming relatively high concentrations there.

The importance of riverine inputs and sedimentation of PCBs differs from HCH and toxaphene. Using average concentrations of 5 ng/L PCBs in Eurasian rivers (derived from concentrations on river SPM, Annex Table 6·A7), fluxes of 15 tonnes/y are estimated. This represents 24% of all inputs estimated for PCBs. Although the 5 ng/L concentration is highly uncertain, levels one-tenth as large (that is, equivalent to North American rivers) would still give rise to significant inputs. Thus, knowledge of riverine inputs from Eurasian sources is critical for accurate estimates of loadings to the Arctic Ocean. By comparison, ocean current inputs are less significant because of low surface water concentrations.

Removal by sinking particles is much more significant for PCBs than it is for HCH and toxaphene because of their much greater hydrophobicity, and this gives rise to the low seawater concentrations observed by Hargrave *et al.* (1988) and Iwata *et al.* (1993). Sedimentation and burial of PCBs is not predicted to be a major removal process compared to outflows via ocean currents, because of low overall sedimentation rates, however, it is much greater than in the case of HCH and toxaphene. Sedimentation may be particularly significant in the EAO which has large shelf areas that could, at least temporarily, sequester particle-bound PCBs. Sediment concentrations are generally higher in the EAO than in the NAAO, although measurements in the latter region are limited. For budget purposes, average concentrations of 0.5 ng/g dw were used for EAO sediments and 0.25 ng/g dw for the NAAO.

There is greater certainty in some of the budget results for PCBs than for toxaphene, but overall, the estimates are highly uncertain. The results suggest that the Arctic Ocean is not in steady state or in export mode with respect to PCBs. Inputs exceed outputs by a factor of 1.5. Further measurements, especially of seawater concentrations, as well as confirmation of high PCBs in Russian seas and rivers, are critical to more accurate budgets for PCB.

#### 6.6.4.4. **Marine invertebrates**

OCs have been measured in small numbers of benthic and pelagic marine invertebrates, including crustaceans, amphipods, zooplankton, annelid worms, and mollusks (Annex Table 6·A15). Results are available from Russia, Canada, Iceland, and Greenland. There are very limited data for any one species/genera of invertebrates which makes evaluation of circumpolar trends difficult. Furthermore, OC values in invertebrates are quite variable, depending upon the trophic position of the organism and its life span. Zooplankton and pelagic amphipods generally have a one to two year life cycle and, therefore, lower levels of OCs than more long-lived fish and benthic amphipod species. An exceptional example of the influence of trophic position on levels of OCs in invertebrates are the high concentrations of ΣPCB and toxaphene found in the large benthic lysianassid amphipod, *Eurythenes gryllus*, in the Canadian Arctic, which may scavenge marine mammal tissues. Food chain biomagnification of OCs in the marine food web is discussed further in section 6.6.4.10.

For most invertebrates analyzed, the dominant OCs found were PCBs, followed by DDT and HCH. HCB and ΣCHL were also present in all samples analyzed. With the exception of *Eurythenes*, concentrations of major OCs were relatively low (e.g., ΣPCBs are generally less than 10 ng/g ww) and were similar within a species group. Where toxaphene has been measured, mainly in samples from the Canadian Arctic and the Bering Sea (Rice *et al.* 1992), it has often been present at similar concentrations as ΣPCB. Various data are given in Annex Table 6·A15, and more detailed studies are described in section 6.6.4.10.

The blue mussel, *Mytilus edulis*, was included in the AMAP monitoring program because of its widespread distribution in Arctic waters. Levels of OCs in blue mussel are available from several locations in Russia, Iceland, Canada, and Greenland (Annex Table 6·A15). Since lipid amounts were fairly similar, the results can be compared on a wet weight basis. The dominant OCs in all blue mussel samples were ΣPCBs (sum of ten congeners), followed by HCH, ΣDDT, ΣCHL, and chlorobenzenes. Values of HCB, ΣDDT, and ΣPCB were similar for Iceland and Greenland. In Greenland, values of ΣDDT and ΣPCB were higher at Uummannaq than at Nanortalik, which in turn were higher than at Disko. ΣPCB (sum of seven congeners), ΣDDT, and total chlorobenzenes were higher in the two samples from the Ob and Baydaratskaya Gulfs (southern Kara Sea) than in samples from Iceland or Greenland. Slightly elevated PCBs and DDT were observed in surface sediments from this region, especially the Ob Gulf (Annex Table 6·A14). Levels in blue mussels from the Ob and Baydaratskaya Gulfs were similar to those in blue mussels from Cambridge Bay, Canada, where levels are believed to be affected by the community garbage dump. The Russian samples had similar levels of HCH and ΣCHL to the Greenland samples, but higher levels of HCB and ΣDDT than all other samples.

OCs were determined in zooplankton (larger than 150 μm) at 12 locations in the central Arctic Ocean during the Russia/Canada transpolar cruise in 1994. ΣPCBs and toxaphene were the prominent OCs, with concentrations in the range of 35-67 ng/g ww and 15-31 ng/g ww, respectively (Figure 6·31). Higher ΣPCB levels were observed in samples from the highest latitudes, that is, at stations 24-26 in the Canada/Eurasian Basins, while toxaphene concentrations showed less of a spatial trend. The results for toxaphene in zooplankton did not follow the trend in seawater, which showed higher levels in samples near the pole (Figure 6·24). Zooplankton from four sampling locations in the Bering Sea

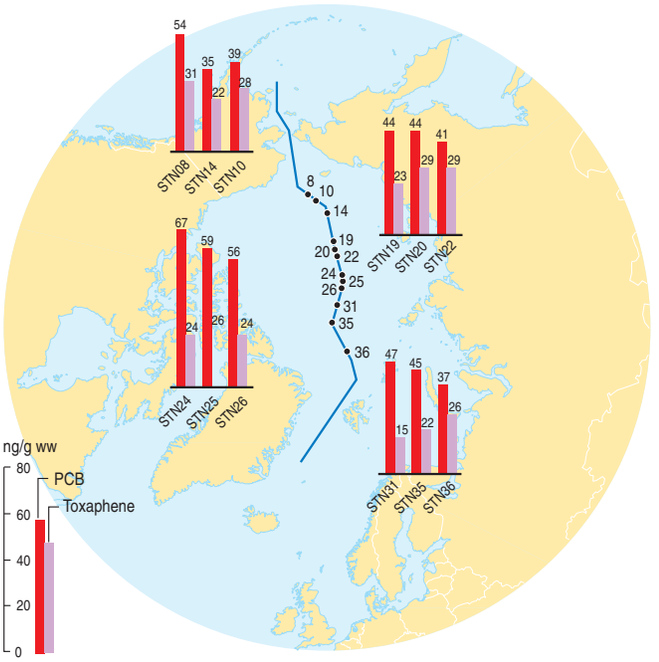



Figure 6·31. PCB and toxaphene (ng/g ww) in zooplankton from the transpolar cruise of the Arctic Ocean, July-September 1994.

(summer 1988 BERPAC cruise) had lower concentrations of most OCs than those from the 1994 transpolar cruise when compared on a wet weight basis (Rice *et al.* 1992, Annex Table 6·A15). However, adjusting for the low lipid content (1.3%) in the Bering Sea samples, levels of all OC groups were found to be higher than in the transpolar cruise samples.

#### 6.6.4.4.1. **Tributyltin**

TBT has been detected in snails from the Norwegian and Alaskan coasts including some collected from remote regions. Imposex has been documented in snails in harbors of northern Norway (*Nucella lapillus*), Spitsbergen and Svalbard (*Buccinum undatum*), Iceland (*Nucella lapillus*), and Alaska (*Nucella lima*), although not always accompanied by detectable TBT concentrations (Short *et al.* 1989, Harding *et al.* 1992, Brick and Bolte 1994, Berge 1995, Skarphédinsdóttir *et al.* 1996).

### 6.6.4.5. **Marine and anadromous fish**

A review of the information available up to 1990 on levels of POPs in marine fish from the Arctic found that few marine or anadromous species, other than Arctic char, had been analyzed for OCs (Muir *et al.* 1992b). A far larger data set is now available on OCs for Arctic marine and anadromous fish. Four species of marine fish and two anadromous species were included in the AMAP monitoring program for the marine environment: Arctic cod (polar cod, *Boreogadus saida*), Atlantic cod (*Gadus morhua*), sculpin (*Myoxocephalus* spp.), long rough dab (*Hippoglossoides platessoides*), Arctic char, and whitefish/cisco (*Coregonus* spp.). Numerous other locally important marine species have also been sampled, including redfish (*Sebastes marinus, S. mentella*), Greenland halibut (*Reinhardtius hippoglossoides*), Atlantic herring (*Clupea harengus*), navaga (*Eleginus navaga*), tusk (*Brosme brosme*), capelin (*Malotus villosus*), and Greenland cod (*Gadus ogac*).

This newer data generally meets the quality assurance criteria set by AMAP. Congener-specific results for ΣPCBs have been determined in all samples, along with HCB, major HCH, chlordane and DDT-related compounds. Toxaphene has been determined in only a limited number of fish, mainly from Canada and Greenland. Various numbers of congeners, from seven to about 90, have been used to calculate ΣPCB levels in fish. However, almost all studies have determined the congeners recommended by ICES (CB 28/31, 52, 101, 105, 118, 138, 153, 156, 170, and 180) which account for 40-60% of total ΣPCB congeners in Arctic char and Greenland halibut (Muir unpubl. data 1996). PCB results in Annex Table 6·A17 are presented only as ΣPCB, as received from the authors of various studies, because it was not possible in all cases to correct for possible differences due to congener numbers. Readers should be aware that ΣPCB results can, therefore, differ by a factor of 1.5-2.0, and this has been noted where relevant to the assessment.

OCs have been measured in the above-mentioned species in either liver and/or muscle or in whole fish. Species in the Gadidae family, such as Atlantic and Arctic cod, have high lipid content in the liver (ca. 50%) and lean muscle tissue (< 1% lipid). Species in the Pleuronectidae family, such as Greenland halibut and log rough dab, have more variable lipid distribution in liver and muscle, with less pronounced differences in lipid content between these tissues. These differences must be taken into account when comparing OC levels among species, both within and among different areas of the Arctic. As with many other fauna, lipid-normalized data are most informative.

The tendency for older individuals to have higher levels of OCs than young of the same species is also true for marine fish. Age and size information on individual fish is not always reported, but these parameters are included in Annex Table 6·A17 when available. In the Barents and Norwegian Seas, cod of 2-11 years and redfish of 7-26 years are represented in the samples (Stange and Klungsøyr 1997, Stange *et al.* 1996). Cod older than 4-5 years are seldom encountered. Ideally, fish of the same species in different areas should be of similar age and size when sampled or compared.

#### 6.6.4.5.1. **OC levels in anadromous fish**

Many stocks of anadromous fish species, including whitefish, Arctic char, and trout in both North America and Eurasia, feed in the sea in summer after reaching a certain size, and then return to rivers and lakes in winter to avoid low seawater temperatures. While at sea, these species feed in nearshore habitats. They feed mainly on crustaceans and small fish species available in that habitat, for example, Arctic cod or Pacific herring (*Clupea harengus*). In the most extreme conditions, for example, in the Canadian High Arctic, these species may feed sparsely or not at all during the winter, and even lose weight. Female Arctic char may not migrate to sea in a year during which they will spawn. The large age-at-size variability common to both landlocked and anadromous char is discussed in section 6.6.3.3.1.

Levels of major OC groups in both landlocked (Annex Table 6·A9) and anadromous char (Annex Table 6·A17), are compared on a circumpolar basis in Figure 6·16. As previously noted for landlocked char, toxaphene is the major OC contaminant in anadromous char at three of four locations in Greenland and at many Canadian sampling sites. Anadromous char from the Kola Peninsula also had higher toxaphene levels than ΣPCB or other OC pesticides. ΣPCB was present at levels 1.2 to three times lower than toxaphene at most locations. ΣDDT and ΣCHL levels were generally 1.5 to three times lower than those of ΣPCB. Levels of ΣDDT and ΣPCB show no clear spatial trends in anadromous char

because there are large variations in mean levels within regions (Figure 6·16). For example, mean ΣPCB levels in char from two lakes on Svalbard, Diesetvannet and Rickardvannet, differ by nine times (Skotvold 1996). This difference cannot be explained by fish maturity, because char from both lakes were in non-spawning condition, nor by lipid levels. However, char from Rickardvannet were older (13 years) compared to those from Diesetvannet (9 years) and there may be differences in trophic position of the char depending on the food chains in each lake.

Results for OC contaminants in anadromous char from Russian rivers are very limited and lipid content is not available (Annex Table 6·A17). Single samples of char from the Khatanga and Ob Rivers had ΣPCB levels of 7.6-8.4 ng/g ww (sum of seven congeners), which are lower than those found for the Kola Peninsula and Rickardvannet on Svalbard. The levels in the Ob and Khatanga River char were within the range of mean ΣPCB (wet weight) levels found in all anadromous char of 2.5-54 ng/g.

Levels of OC contaminants in whitefish (*Coregonus* spp.) muscle are generally lower than in char (means and single samples ranging from 0.82-27 ng/g ww). *Coregonus* spp. are generally first-order carnivores, feeding on phytoplankton and zooplankton at early life stages, then progressing to pelagic and benthic invertebrates in later life stages. In broad whitefish from Canadian rivers, toxaphene was the major OC, however, in three samples from the Ob River in Russia, PCBs predominated (Annex Table 6·A17). The PCB levels in broad whitefish from the Ob (Muir and Lockhart 1994) were similar to levels found in the survey by the 'Regional Center Monitoring of the Arctic' (Melnikov *et al.* 1995). The latter study found highest levels of PCBs in cisco (*C. autumnalis*) in the Ob River (25 ng/g ww). A single sample of inconnu (*C. leucichthys*), a second-order carnivorous fish, from the Yenisey River had higher ΣDDT and ΣPCB levels than all other samples of *Coregonus* species surveyed. However, sample numbers from this survey are too small to make general conclusions about spatial variations of OC levels in anadromous fish from Russian waters.

Koistinen *et al.* (1993) detected low levels of 26 polychlorinated diphenyl ether (PCDE) congeners in salmon from the Tenojoki River in northern Finland. Concentrations of individual PCDE congeners ranged from 0.01 to 0.047 ng/g ww with PCDE 99 (2,2',4,4',5-PeCDE) predominating. About ten-fold higher levels of PCDEs were found in salmon from the Simojoki River in southern Finland.

### 6.6.4.5.2. OC levels in marine fish

In general, the major OCs found in marine fish were toxaphene (measured only in Greenland halibut and Arctic cod) followed by ΣPCB, ΣDDT, ΣCHL, ΣHCH, chlorobenzenes, and dieldrin. The geographic coverage of OC measurements for any one marine species is limited. Perhaps the best spatial coverage is found for Arctic cod (*Boreogadus saida*) because samples from the Canadian Arctic, the mid-Arctic Ocean, the Barents Sea, and the Pechora Sea have been analyzed.

Arctic cod stay within the cold water masses of the Arctic region and have a circumpolar distribution. Arctic cod feed on plankton along the ice edge. Young fish feed on phytoplankton, while the diet of older fish consists of copepods and amphipods (Sameoto 1984). It is a key species in many Arctic food webs, consumed by fish, including char (Moore and Moore 1974) and plaice; birds such as murres, guillemots, and kittiwakes; harp and ringed seals; and whales (narwhal and beluga) (Bradstreet and Cross 1982).

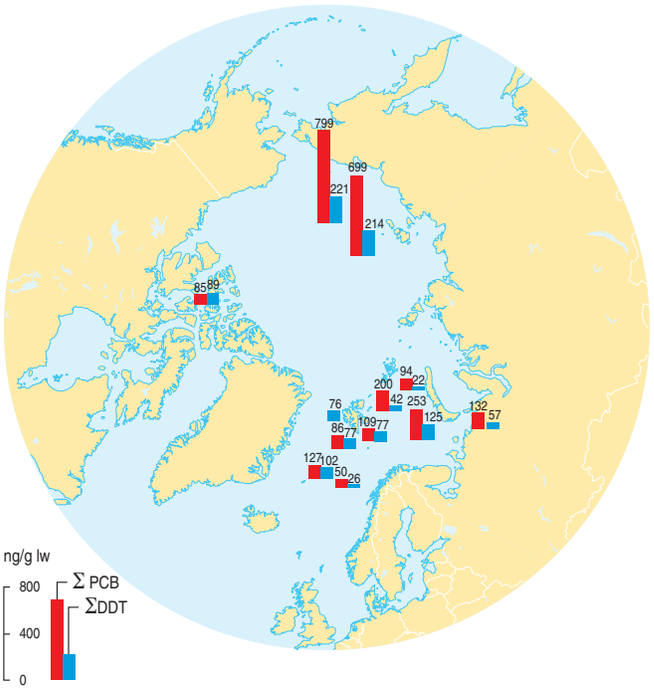



Figure 6·32. ΣPCB and ΣDDT (ng/g lw) in Arctic cod liver, 1994-95. Data are from Annex Table 6·A17.

Movements of individual Arctic cod have not been monitored, however, it is believed that they move depending on feeding opportunities. In the autumn, Arctic cod tend to move to coastal areas, this behavior believed to be a temperature response to spawning. Arctic cod show extensive migration patterns in the Soviet Arctic, believed to be in response to feeding and spawning behavior. They have been reported from along the coast to ranging well out to sea, and from the surface, where they can occur in pockets in sea ice, to as deep as 450 m (Lowry and Frost 1981) and 900 m (Walters 1955).

Lipid weight ΣPCB and ΣDDT concentrations in Arctic cod liver are compared in Figure 6·32. Results for samples from the Barents/Norwegian Seas are based on liver samples, while those from the central Arctic Ocean and Canada Archipelago are whole fish. Based on results of Killie and Dahle (1996a), who analyzed both muscle and liver of Arctic cod (Annex Table 6·A17), lipid normalized concentrations in the two tissues are similar. Highest concentrations of ΣPCB were found in fish from the central Arctic Ocean (Muir and Macdonald unpubl. data) and those from the southern Novaya Zemlya area compared to Arctic cod from the Greenland Sea. Killie and Dahle (1996a) found ΣPCB (sum of ten congeners) in cod liver from the Pechora Sea ranged from 59 to 110 ng/g ww.

Atlantic cod (*Gadus morhua*) inhabit cool-temperate to subarctic waters from offshore regions to the edge of the continental shelf (Scott and Scott 1988). It is found in most coastal areas in the North Atlantic Ocean and in the Baltic Sea. Commercially, it is one of the most important fish species in the Atlantic, however, it is not always considered to be an Arctic species. Due to its wide distribution, Atlantic cod is a species given priority in many national and international marine monitoring programs within the OSPARCOM and HELCOM areas.

Atlantic cod is an opportunistic feeder, and the diet will vary considerably from year-to-year based on availability of prey species. As fry, they feed on copepods, amphipods, and crustaceans, including crabs, and as adults, on redfish, capelin, and herring.

There are several different stocks of Atlantic cod within the Northeast Atlantic. The two main European stocks migrate annually from southern locations in summer to northern areas in winter. Cod that spawn around Greenland are part of the Iceland-Greenland migration. The Arcto-Norwegian stock in the Barents Sea migrates down the Norwegian coast for spawning (Harden-Jones 1968). The Canadian stocks also migrate annually. One tagged fish was recorded to move from the Grand Banks, off Newfoundland, to the North Sea, a distance of 3228 km (Gulland and Williamson 1962). The stock of Labrador cod move north and south along the coast of Labrador.

In the Barents Sea, OC concentrations in Atlantic cod liver (expressed as wet weight) were two to ten times higher than in Arctic cod and long rough dab (Stange and Klungsøyr 1997). Atlantic cod feed on a variety of prey, including other fish species, and have a higher trophic status than plankton-eating Arctic cod and benthic-feeding long rough dab. The levels of OCs in Atlantic cod liver from the Barents Sea (ΣPCB 94-685 ng/g ww, ΣDDT 67-344 ng/g ww) were lower than the levels found in cod from Haltenbanken in the Norwegian Sea (ΣPCB 232-519 ng/g ww; ΣDDT 129-452 ng/g ww; Stange *et al.* 1996) and cod from the northern parts of the North Sea (ΣPCB 506 ng/g ww, ΣDDT 350 ng/g ww; de Boer 1988). The lowest OC concentrations (ΣPCB and ΣDDT < 100 ng/g) were found in livers of cod from different stocks in Icelandic and Faeroese waters (Jóhannesson *et al.* 1995, Stange *et al.* 1996).

Toxaphene was a prominent OC in Atlantic cod liver from Tanafjord, northern Norway, and was present at levels similar to those found in the same species from the Baltic Sea collected in the late 1980s (Paasivirta and Rantio 1991). Few other measurements of toxaphene are available for Atlantic cod (liver) from Arctic waters.

OCs have been measured in liver of sculpins (*Myoxocephalus* sp.) in the Canadian Archipelago and at four locations in Greenland waters (Annex Table 6·A17). These species are bottom feeders, preying on benthic fauna such as bivalves, sea urchins, polychaetes, and amphipods. Sculpins are, therefore, good potential indicators of sediment contamination. In Cambridge Bay (Victoria Island, Canada), where PCB contamination of sediments has been identified, sculpins had higher levels of ΣPCB than those from reference areas (Bright *et al.* 1995c). Cleeman *et al.* (1996c) found higher ΣPCB concentrations in sculpin from East Greenland (Scoresbysund) than locations on the western side of Greenland or in the Canadian Archipelago. The reason for these differences is unknown and could not be explained by lipid content.

Greenland halibut (*Reinhardtius hippoglossoides*) is a bottom-feeding predatory fish with high lipid content in the muscle tissue and in the liver (≈15%). The species has a circumpolar distribution and is found on the east and west coasts of Greenland (Muus *et al.* 1982). It is common at depths of 200-1000 m. Tagging studies of Greenland halibut have shown that at least some individuals migrate over 1000 km (Sigurdsson 1981, Bowering 1984). Bowering and Brodie (1991) suggest that Greenland halibut from off the coast of Labrador and eastern Newfoundland migrate to deep-water spawning areas, then after spawning move back to summer feeding areas (Chumakov 1969, Sigurdsson 1981).

Relatively high levels of OCs were found in Greenland halibut liver from the eastern Canadian Arctic (ΣPCB = 136 ng/g ww) and West Greenland (ΣPCB = 119 ng/g ww) (Berg *et al.* 1996). ΣPCB levels in Greenland halibut muscle from the Beaufort Sea were very similar to those from western Davis Strait (Cumberland Sound) (means of 202 and 165 ng/g ww, respectively; Muir and Lockhart 1996). These levels are three to five times higher than wet weight levels in sea-run char muscle and 15-20 times higher than Arctic cod (whole fish). However, ΣPCB levels in Greenland halibut liver from the Norwegian and Barents Sea were lower, 24 and 18 ng/g ww, respectively (sum of 13 congeners; K. Stange unpubl. data). Toxaphene was the major OC found in Greenland halibut muscle and liver. Mirex was detectable in halibut tissues, at mean concentrations of 1 ng/g in muscle and 2.8 ng/g in liver (Muir and Lockhart 1996). Higher ΣHCH levels were found in Greenland halibut from the Beaufort Sea and Cumberland Sound (Baffin Island) than in halibut from the eastern Davis Strait (Annex Table 6·A17).

Stange and Klungsøyr (1997) found ΣPCB concentrations in long rough dab (American plaice, *Hippoglossoides platessoides*) from the Barents and Greenland Seas ranging (means) from 15 to 57 ng/g ww. Levels in long rough dab were similar to those reported from Iceland by Jóhannesson *et al.* (1995). Dab can be found on both sides of the northern North Atlantic. It is the most abundant flatfish in the Barents Sea (Albert *et al.* 1994). Long rough dab live near the bottom and prey mainly upon various benthic organisms such as Ophiuroidea and Polychaeta (Pálsson 1981). Small long rough dab also feed on planktonic prey groups.

PCB levels in liver of redfish (*Sebastes* spp.) from the Greenland Sea and the North Atlantic Ocean ranged from 13 to 203 ng/g ww, and ΣDDT levels from 13-177 ng/g ww (Stange *et al.* 1996). Similar concentrations were found in redfish liver from West Greenland (ΣPCB, 127 ng/g ww; ΣDDT, 77 ng/g ww) (Berg *et al.* 1996). Redfish are found at depths down to 500 m in the North Atlantic and the Barents Sea. They are thought to feed on planktonic crustaceans and small fish (Pedersen and Riget 1992).

Berg *et al.* (1996) studied OC levels in livers of eight different deep-sea fish species from the Davis Strait. The highest levels of PCBs were found in blue hake (*Antimora rostrata*, 615 ng/g ww) and tusk (*Brosme brosme*, 522 ng/g ww). Lowest levels were found in jelly wolffish (*Anarhichas denticulatus*, 38 ng/g) and Greenland halibut (119 ng/g ww). OC levels in liver of these deep-sea Arctic species were similar to or slightly higher than concentrations in Atlantic cod caught at the same depth in the northwest Atlantic (off Newfoundland/Labrador) (Hellou *et al.* 1993). Berg *et al.* (1996) did not observe any clear relationship between food preference or feeding depth and OC levels in deep-sea species. They noted that some differences between species may be related to lipid distribution. Greenland halibut and jelly wolffish have significant lipid storage in muscle, whereas liver is the major organ for other species, for example, gadiformes, blue hake, tusk, and roughhead grenadier. The deep sea predatory fish were distinguished from shallow depth pelagic feeders, such as Arctic cod, by lower levels of more water soluble OCs, such as HCH, and higher proportions of highly chlorinated PCB congeners. This reflects the fact that more highly chlorinated ΣPCBs are preferentially sorbed to sinking particles, whereas HCH is mainly in the dissolved phase with much lower dissolved concentrations at depth.

#### 6.6.4.5.3. **PCDD/Fs and planar PCBs in fish and invertebrates**

Information on levels of PCDD/Fs and non-*ortho*-substituted PCBs (nPCBs) in anadromous and marine fish is very limited. Arctic char from the Canadian Arctic were found to have low pg/g ww concentrations of nPCBs and PCDD/Fs (Annex Table 6·A19, Ford *et al.* 1993). CB 77 was the major non-*ortho*-substituted congener in char (means of 6.9-63

pg/g ww). TCDD TEQs in char muscle were attributable mainly to PCDD/Fs when these and nPCBs were determined in the same samples.

Bright *et al.* (1995c) detected nPCBs (CB 77 and 126) in sea urchins and sculpin from Cambridge Bay (Victoria Island), but levels in mussels and soft shell clams were below detection limits (< 10 pg/g ww). Arctic cod from Lancaster Sound in the Canadian Arctic Archipelago had low concentrations of nPCBs, mainly CB 77 (5 pg/g ww). Schlabach and Skotvold (1996a, 1996b) found low pg/g levels of PCDD/Fs in samples of the filter-feeding mussel (*Modiolus modiolus*) in the vicinity of a smelter near Kirkenes in northern Norway. TCDD TEQ concentrations in mussels ranged from 0.61 pg/g ww near the smelter (Jakobsnes) to 0.2 pg/g at the reference site (Heikenesset). Atlantic cod caught at Prestøyskjær, located 1.5-2.3 km northeast of the source of emissions, and at a reference area close to Russeviknesset, approximately 12.7 km north of the smelter works, also had low TCDD TEQs in muscle (0.02-0.04 pg/g ww).

#### 6.6.4.6. **Seabirds**

There are approximately 50 species of seabirds in Arctic marine waters, feeding primarily on fish, crustaceans, amphipods, and other marine invertebrates (chapter 4). Auks, guillemots, razorbills, loons, and mergansers of the family Alcidae are the characteristic fish-eating birds of the Arctic. Alcids usually breed on cliffs and islands, but feed at sea. Skuas and jaegers winter at sea, but feed almost entirely onshore on insects and small mammals during the breeding season. Loons (divers) and several species of ducks switch from marine foraging in winter to feeding and breeding on the tundra wetland in summer. Also, three species of petrel nest on land and feed on zooplankton at sea. Many prey species are often first- or second-level carnivores, therefore the possibility for biomagnification of contaminants exists.

Some gull species are opportunistic surface feeders as well, with diets including carrion, bird eggs and chicks, and also garbage. Some individuals of glaucous gulls (*Larus hyperboreus*) and herring gulls (*Larus argentatus*) feed on seabird nestlings and adults.

Surveys of OC levels in eggs and tissues of Arctic seabirds conducted in the mid-1970s showed that seabirds breeding in the High Arctic were contaminated with a similar suite of organic contaminants as those breeding in temperate regions (Noble and Elliott 1986). Since that time, Arctic seabird species have been monitored for contaminants, with various species having been selected to represent different geographic areas, trophic levels, feeding strategies, and overwintering habits. Data on contaminant levels in eggs sampled from Canada and northern Eurasia were available for several species, thus, comparison of levels on a geographic basis is possible. Samples were also analyzed from chicks and from various tissues in adults (liver, brain, fat, muscle) in a number of species, mainly in samples from Norway and Russia. Levels of OCs in Arctic seabirds from several studies are given in Annex Table 6·A16.

In a recent Canadian study, lipid weight levels of OCs in chicks were lower than levels in eggs from the same species at the same site for common guillemot, black-legged kittiwake (*Rissa tridactyla*), glaucous gull, and northern fulmar (*Fulmaris glacialis*) (Braune unpubl. data, Annex Table 6·A16). OC levels were similar in chicks and eggs of thick-billed murre (Brünnich's guillemot, *Uria lomvia*) from the same sites. Where several tissue types were analyzed in adult birds (mainly from Norway and Russia) lipid weight levels of OCs were highest in liver and fat tissues and lowest in brain tissue. ΣPCB was the dominant OC contaminant in egg samples followed by ΣDDT, except for king eider, which had higher ΣCHL levels. HCB, ΣCHL, ΣHCH, and dieldrin were also present. Where congener-specific PCB analyses were done, the dominating CBs in samples were those with six chlorines. No data on levels of toxaphene, PCDD/Fs, or nPCB congener levels were available.

Lipid weight levels of ΣPCB in eggs from various species, based on two studies (B. Braune unpubl. data, Barrett *et al.* 1996) are shown in Figure 6·33. In general, spatial trends in levels of ΣPCB were correlated with trends in other OCs. Glaucous gull, herring gull, black-legged kittiwake, cormorant, and puffin had the highest lipid weight OC levels. DDE concentrations were highest in glaucous gulls from the Canadian High Arctic (1580 ng/g ww), herring gull from West Finnmark, Lofoten, and Tromsø (440, 530, and 1000 ng/g ww, respectively), and Brünnich's guillemot from Svalbard (Figure 6·33). Both gull species examined are fish-eating opportunistic species. The northern fulmar and black-legged kittiwake are both offshore surface feeders, but the fulmar is also a scavenger that includes carrion in its diet. Shag (*Phalacrororax aristotelis*), a piscivorous species, also had relatively high OC levels. Lowest lipid weight levels of most OCs were found in common eider (*Somateria mollissima*) and king eider (*Somateria spectabilis*). These species are molluscivores.

Differences in levels of OCs in different species related to feeding were evident in data from Prince Leopold Island in the Canadian High Arctic (B. Braune pers. comm., Annex Table 6·A16). Analyses of variance on residue levels found in eggs of black guillemot (*Cepphus grylle*), glaucous gull, black-legged kittiwake, northern fulmar, and thick-billed murre from Prince Leopold Island, indicated that the five species of seabird had significantly different levels of HCB, ΣCHL, DDE, and ΣPCB ($p < 0.001$). OC residue levels were four to ten times higher in glaucous gulls than in the other species. Similarly, in the Barents Sea, gulls had the highest OC levels, and levels in common eider were the lowest, while kittiwakes had intermediate levels.

Levels of persistent OCs in seabird eggs are particularly low in the western Canadian Low Arctic (western NWT). Analyses of variance of OC levels in eggs of thick-billed murres collected from two High Arctic colonies (Prince Leopold Island, Coburg Island) and two eastern Canadian Low Arctic colonies (Coats Island, Digges Island) showed that there were significantly different levels among colonies for HCB and ΣPCB at the two locations ($p < 0.001$ and $p = 0.05$, respectively) (B. Braune pers. comm.). The eggs from Digges Island contained higher residue levels of OCs than the eggs from the High Arctic sites. This may be partly due to differences in the overwintering habitats. Similar patterns existed for black guillemots. All of the species except for the black guillemot disperse widely after breeding. Large numbers of the northern fulmar, thick-billed murre, and glaucous gulls from the eastern Arctic overwinter in the northwest Atlantic. The black guillemot is relatively sedentary, overwintering at the edge of the winter pack ice (Godfrey 1986). This same trend of higher OC levels in eastern Canada is also evident in the waterfowl (section 6.6.2.2.2).

In the Barents Sea, levels of persistent OCs in seabird tissues differed in the four areas examined, namely Franz Josef Land, Ny-Ålesund, Bjørnøya (Bear Island), and Hornøya, although these could not be shown to be significantly different statistically (Savinova *et al.* 1995b). Hepatic ΣPCB levels, expressed as wet weight values, in juvenile common eider from Franz Josef Land were 25% of levels from Ny-Ålesund. On a lipid weight basis, the levels at Franz Josef Land were 4-

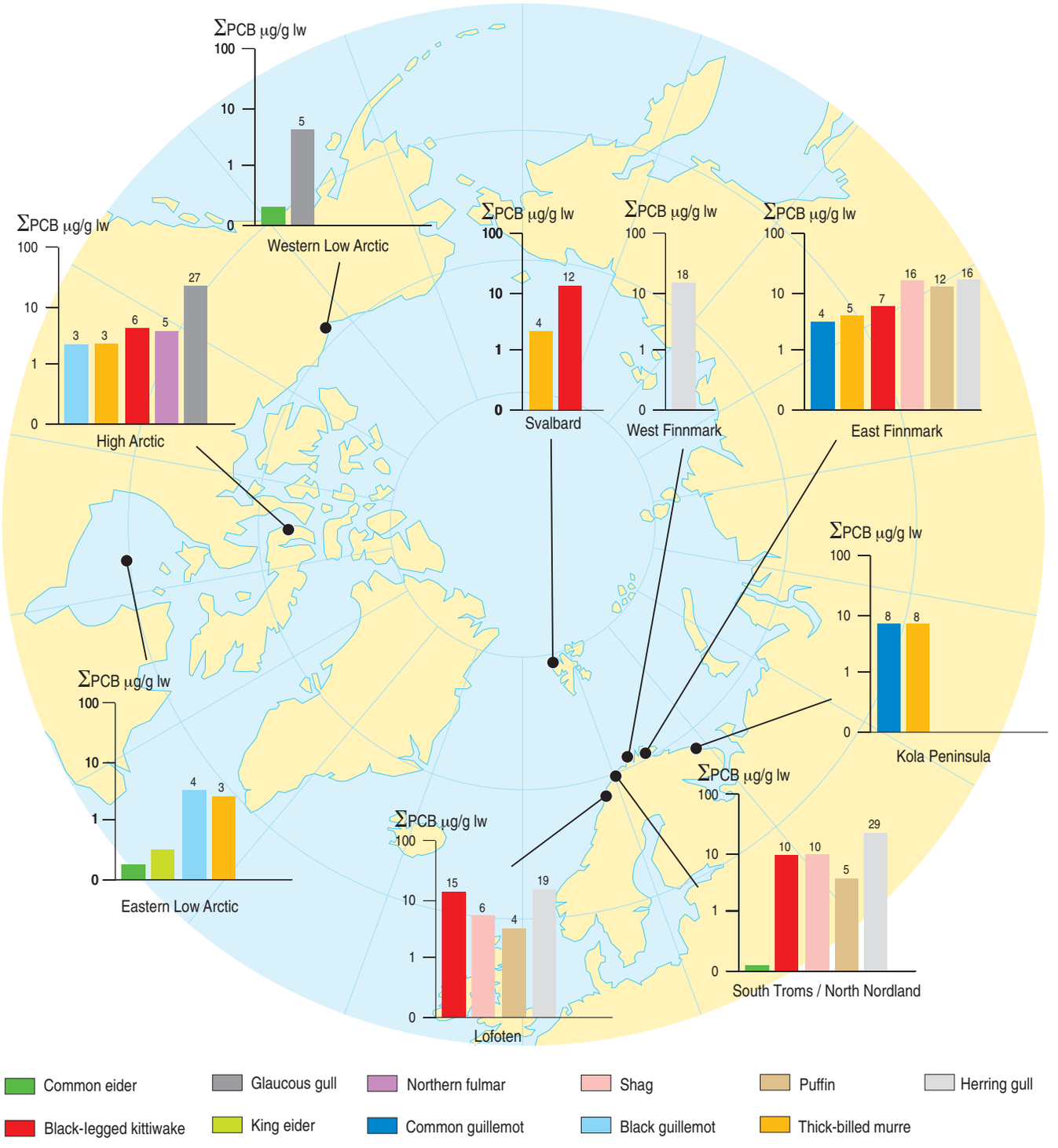



Figure 6·33. Levels of PCBs in seabird eggs (µg/g lw) (Braune unpubl. data, Barrett *et al.* 1996). Values above bars are actual mean concentrations. Wet weight means are given in Annex Table 6·A16.

5% of those on Svalbard, which is an even larger difference. Since these juveniles had not yet migrated, these differences reflect primarily local contamination. Wet weight hepatic OC levels in adult common eider from Ny-Ålesund were approximately two times higher than the corresponding levels in juveniles from the same area. On a lipid weight basis, the juveniles had higher PCB levels (2500 versus 1600 ng/g lw). Mean hepatic levels of OCs found in adult kittiwake, the most common species in all three areas, were approximately five times as high as levels in common eider. On a lipid weight basis the levels were 7-34 times higher. The comparison should probably only be made for Ny-Ålesund, as this is the only site where data for adults of both species are available, in which case the levels in kittiwake are about seven times higher than in common eider. Also, mean hepatic levels of OCs in kittiwakes increased from Ny-Ålesund to Bjørnøya. Glaucous gulls from Bjørnøya had significantly higher levels than those from Ny-Ålesund on a wet weight basis. On a lipid weight basis, the levels were higher in the samples from Ny-Ålesund.

Barrett *et al.* (1985) had previously concluded that there were few differences in OC levels in seabirds on the Norwegian mainland, the Kola Peninsula, and Svalbard. This may be due to similar overwintering areas and similar environmental contamination throughout the Barents Sea.

Lipid weight levels of most OCs in black-legged kittiwake eggs from Svalbard are higher than in eggs from the Canadian High Arctic (Figure 6·33). ΣDDT and ΣPCB levels in

thick-billed murre eggs are higher in samples from Svalbard, Hornøya, and the Kola Peninsula when compared to samples from the High Arctic and eastern Low Arctic of Canada. However, ΣHCH levels are higher in the Canadian thick-billed murre eggs than in the Norwegian site and HCB and chlordane levels are similar at the Norwegian and Canadian sites. For glaucous gull, wet weight hepatic levels of ΣPCB and ΣDDT, in particular, seem to be high in the one sample from the Taimyr Peninsula, Russia, when compared to samples from Svalbard and Bjørnøya. ΣPCB levels, as well as other OCs, are also higher in a liver sample from herring gull from the Taimyr Peninsula, when compared to liver samples from the same species from eastern Taimyr and the Yamal Peninsula. The lipid content of these samples was not available, thus, it is not possible to determine whether these higher levels were simply due to higher lipid contents. However, the picture that emerges from the results of OC analyses in Arctic seabirds is that the Barents Sea may be more contaminated with ΣDDT and ΣPCB than the Canadian High Arctic.

Surveys in 1994-1995 by Roshydromet (Melnikov *et al.* 1995, Melnikov *et al.* 1996a) have provided the only data available on OC contaminants in seabirds/shorebirds in the Kara and Laptev Sea region (Annex Table 6·A16). However, sample numbers are small and could be unrepresentative of populations as a whole. Some samples have occasional high ΣDDT and ΣPCB levels compared to other Arctic regions. In Steller's eider (*Polysticta stelleri)*, the liver levels of most OCs are not different from common eider liver except for ΣDDT and ΣPCB, which are higher. The same seems to be true of the king eider sample for ΣPCB. The northern fulmar and thick-billed murre from the Laptev Sea have relatively high ΣPCB levels. OC levels in both gull species were similar to the range of mean concentrations found in gulls from Canada and Norway (Annex Table 6·A16).

Gabrielsen *et al.* (1995) obtained data on glaucous gull from Svalbard, from seabirds that were found dead of unknown causes. The wet weight concentrations of various OCs are not notably different from those observed for live-sampled tissues. However, these animals were emaciated and stressed before death, with lipid levels in all tissues except brain much lower than those observed in whole eggs or chicks in Canadian samples. Thus, the lipid-normalized concentrations are high. The authors believe that PCBs cannot be discounted as contributing to the death of the gulls (Gabrielsen *et al.* 1995). OC levels in eggs of two of the gulls' prey species (both guillemot species) are similar to, or lower than those observed in Canadian samples.

There are several population groups of eiders throughout Arctic Canada and Hudson Bay, some of which overwinter in the north and others which migrate to more temperate latitudes (Barry 1986, Reed and Erskine 1986). The main migration routes are shown in Figure 6·34. Only the eiders from Ungava Bay, Hudson Bay, and Hudson Strait area (eastern Low Arctic) migrate south to overwinter off Newfoundland and in the Gulf of St. Lawrence. The Gulf of St. Lawrence receives the effluent flow from the Great Lakes and St. Lawrence River, waters known to be contaminated (Gagnon *et al.* 1990, Comba *et al.* 1993). The western High Arctic population overwinters in less contaminated northern waters. These differences in overwintering areas are reflected in the tissue residue concentrations of the various groups of eiders (Figure 6·34).

Although no biomagnification studies have been carried out in Arctic seabirds, lipid- normalized OC levels can be compared between seabirds and their major prey to determine if biomagnification is occurring. Fish-eating Arctic

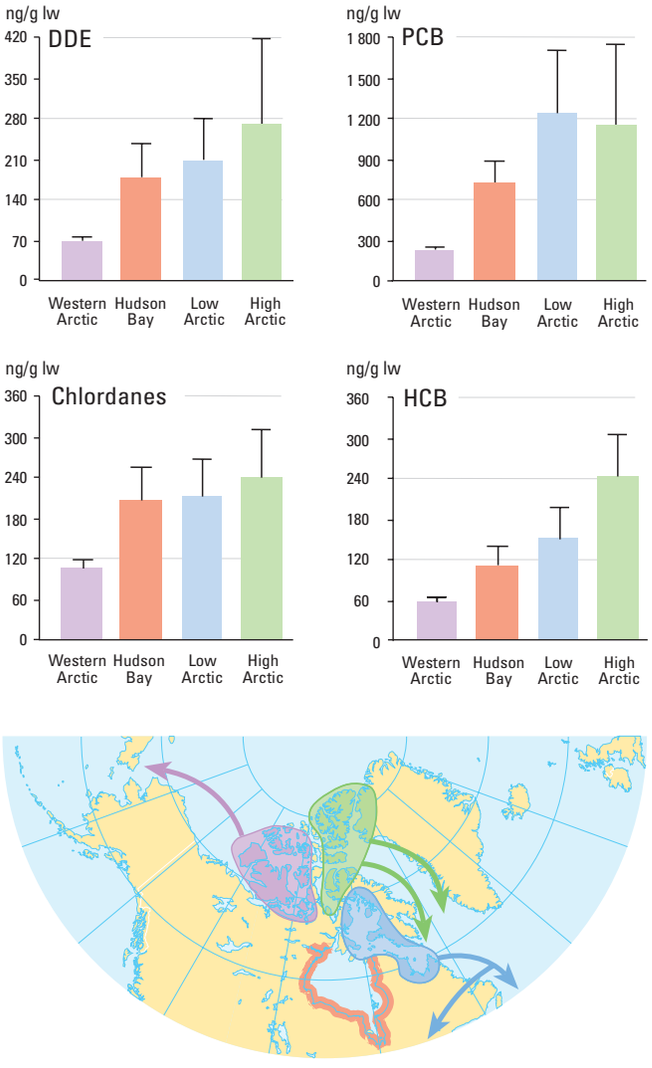



Figure 6·34. Migration patterns of four main stocks of eider ducks in the Canadian Arctic. Organochlorines in the same four stocks. Values in the western Arctic are based on 11 birds in 2 pools; in Hudson Bay, 28 birds in seven pools; in the High Arctic, 18 birds in three pools; and in the Low Arctic, 55 birds in eight pools (Braune unpubl. data).

seabirds have higher lipid weight OC levels than those found in the Arctic marine fish species studied (Annex Table 6·A17). Seabirds that prey on mollusks also have higher lipid weight OC levels than those found in blue mussels from the Arctic. OC levels in glaucous gull are also much higher than those found in guillemot eggs, which is a food source in many areas. These observations indicate that biomagnification of OCs occurs in Arctic seabirds.

### 6.6.4.7. **Pinnipeds and cetaceans**

There is an extensive series of measurements of OCs, especially of PCBs and DDT-related compounds, in blubber samples from pinnipeds and cetaceans. Work carried out prior to about 1990 has been reviewed and tabulated by Muir *et al.* (1992b). Recent results for OCs in pinniped and cetacean tissues (mainly blubber) are presented in Annex Table 6·A18. Annex Table 6·A19 presents results for PCDD/Fs and nPCBs in pinnipeds, cetaceans, and polar bears. This review of spatial trends will focus mainly on work conducted on samples collected between the late 1980s and the present.

In general, all OCs, as well as brominated organics, that have been detected in tissues of marine biota in temperate waters (e.g., Baltic Sea, North Sea, Gulf of St. Lawrence, Mediterranean Sea) have also been detected in Arctic pin-

nipeds and cetaceans. By far the most frequently determined OCs are PCBs (usually as congeners) and DDT-related compounds. Less frequently measured are the hexachlorocyclohexanes, chlorobenzenes, chlordane-related compounds (*cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane), chlorobornanes (toxaphene components), and cyclodienes (dieldrin, endrin). Measurements of chlordane and toxaphene-related compounds are much less frequent than might be expected considering their relatively high proportional contribution to total OCs determined in marine mammal tissues. For example, there is very little data on toxaphene in Arctic marine mammals from Greenland, Norwegian, or Russian waters.

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) have been determined, along with non-*ortho*-substituted PCBs (nPCBs), in seals from Arctic Canada, northeastern Greenland, and Svalbard, but information on their spatial trends is limited. Brominated diphenyl ethers and brominated biphenyls were also identified in ringed seal blubber from Svalbard (Jansson *et al.* 1993), but have not been measured at any other location.

It must be emphasized that spatial trends of persistent OCs discussed below are qualitative because they are based on evaluation of means and ranges of concentrations. Intercomparability of the laboratories involved in the analyses is reasonably good because all have been involved in ICES intercomparisons (de Boer *et al.* 1992). However, in the case of PCBs and chlordane, some laboratories have included more congeners or components than others in ΣPCB and ΣCHL results. Rigorous comparisons between locations also require information on age, sex, blubber thickness, nutritional status, collection season, and reproductive status of the animals. This information has been collected for most locations (Canada, Greenland, Norway, Russia), but has been used only qualitatively in this assessment of spatial trends.

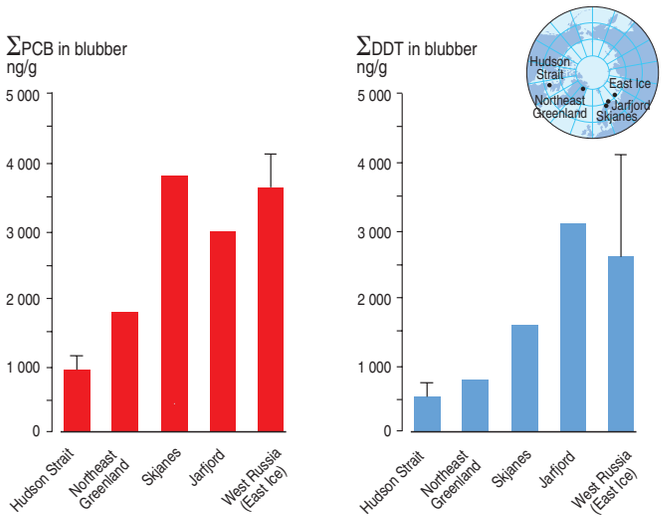



Figure 6·35. Geographical trends of PCB and DDT in harp seals from Arctic waters. Vertical bars represent arithmetic means ± SD of combined results for males and females. The SD has been estimated from the range in some cases.

### 6.6.4.7.1. **Pinnipeds**

• *Harp seals*

Harp seals (*Pagophilus groenlandicus*) are important Atlantic species, inhabiting Arctic and subarctic waters. They feed primarily on small marine fish and secondarily on crustacean macroplankton. Pups feed on euphasid shrimps, krill, and prawn. There are three separate harp seal populations in the Arctic, all three of which migrate annually between southerly breeding sites and northern feeding grounds, both sites at the edge of the pack ice (Lavigne and Kovacs 1988). Herds that breed off 'The Front' near the Magdalen Islands and in the Gulf of St. Lawrence migrate north to Hudson Bay, Davis Strait, and Baffin Bay. Animals that reach the edges of the range may migrate as far as 5000 km. The breeding population that congregates in the White Sea off the coast of Russia and the population that pups mainly between Jan Mayen and Svalbard move to ice patches north of the breeding areas, which include the northern Barents and Kara Seas north of Svalbard, Franz Josef Land, and Severnaya Zemlya. The migrations involve nearly all of the adults and some of the immatures. Many young animals, immature adults, and some pregnant females do not migrate.

An extensive series of measurements of OCs in harp seals from the Norwegian Arctic, northeastern Greenland, and the western Russian Arctic have recently been completed (Skaare 1996, Oehme *et al.* 1995d, Kleivane *et al.* 1996) (Annex Table 6·A18). Intra-species variation was large. Nevertheless, there seems to be a west to east increase in OC levels in harp seal blubber (Figure 6·35). Concentrations of ΣPCBs and ΣDDT in harp seals from northeastern Greenland (west ice north of Jan Mayen Island) were found to be about two- to three-fold lower than those from northern Norway (Jarfjord, Skjånes) and western Russian waters ('east ice'). ΣPCB levels in female harp seals collected at Salluit in Hudson Strait (Beck *et al.* 1994) are similar to levels in females from northeastern Greenland, but 2.5-fold lower than mean levels in females from the southern Barents Sea in western Russia (Kleivane *et al.* 1996). Apart from the work of Beck *et al.* (1994), there are no other recent studies of OCs in harp seals from the eastern Canadian Arctic or western Greenland. Female harp seals from the east coast of Newfoundland have similar ΣPCB concentrations in blubber (mean of 1 μg/g in samples from the early 1990s) to those from Salluit and northeast Greenland (Muir 1996 unpubl. data).

Harp seals show major seasonal changes in OC concentrations in blubber, which makes it difficult to interpret spatial or temporal trends from different studies. Kleivane *et al.* (1995, 1996) found that highest levels of all OCs occurred in April/May when animals were at their leanest, and lowest levels in September when blubber layers were thicker.

• *Ringed seals*

The ringed seal (*Phoca hispida*) is by far the most abundant and widely distributed resident Arctic pinniped, with a widespread circumpolar distribution. Ringed seals have a preference for annual, landfast ice, but are also found in multiyear ice. Their diet consists of fish, mainly Arctic cod, and crustaceans (amphipods, mysids, and euphausids). A summer diet of about 54% cod has been estimated for ringed seal in Barrow Strait using $^{15}N$ isotope enrichment analysis (Hobson and Welch 1992).

Ringed seals are not generally considered to be a highly mobile species. Based on site tenacity and territoriality, Smith and Hammill (1981) estimated that male ringed seals may occupy the same small under-ice habitat for as much as nine months of the year. Nevertheless, high rates of immigration and emigration have been reported. For example, ringed seals tagged at Point Parry, NWT (70°N, 125°W), were killed at Holman and Sachs Harbour in the NWT, Point Barrow in Alaska, and East Cape in Siberia (66°N, 170°W) (Smith 1987). A yearling ringed seal tagged at Resolute Bay, NWT (75°N, 95°W) was killed near Paamiut near the southern tip of Greenland (S. Innes pers. comm.).

OC levels in blubber of ringed seals collected prior to 1988 from ten locations in the Canadian Arctic (including

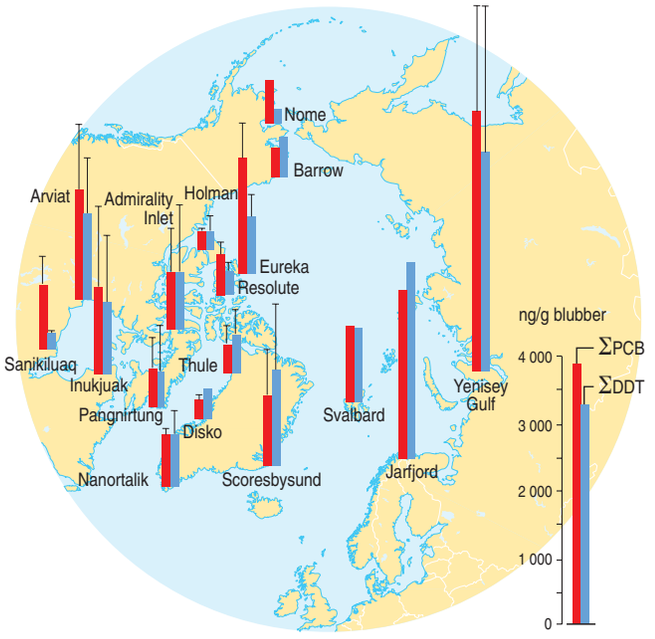



Figure 6·36. ΣPCB and ΣDDT in ringed seal blubber (ng/g). Vertical lines denote standard deviation.

the western and central High Arctic, northwest and northeast Hudson Bay, Baffin Bay, Ungava Bay) and at Svalbard have been summarized in Muir *et al.* (1992a). That review showed that PCBs, chlordanes, and DDTs were the most prominent contaminants, while toxaphene (or chlorobornanes), HCHs, and chlorobenzenes were present at lower concentrations. Concentrations of PCBs and ΣDDT were higher at Svalbard than in the Canadian Arctic. Within the Canadian Arctic, concentrations of ΣPCBs and ΣDDT are quite similar in males and females from all locations, especially when age and sex of the animals are taken into account using analysis of covariance (Weis and Muir 1996). More recent (samples collected post-1988) analyses of seal blubber samples are given in Annex Table 6·A18. Spatial trends for ΣPCBs and ΣDDT as means of combined results for males and females at 17 locations from the eastern Bering Sea (Nome) to the southern Kara Sea (Yenisey Gulf) are summarized in Figure 6·36. These mean concentrations are influenced by age and sex differences among sites. Geographical comparisons of OCs in seal blubber are best made with females because they show less variation with age than males, however, data from females only were not available from all sites so the combined results were used instead.

Two trends are discernible in Figure 6·36. Ringed seals from Hudson Bay (Arviat, Inukjuak, Sanikiluaq) have higher concentrations of ΣPCB and ΣDDT than those in the central Canadian Archipelago (Resolute, Admiralty Inlet) and western Greenland (Thule, Disko). Secondly, ringed seals from eastern Greenland (Scoresbysund), Svalbard (Luckas *et al.* 1990, Daelemans *et al.* 1993), and northern Norway (Skaare 1996) had similar levels to those in Hudson Bay, but generally higher levels than found in the central Canadian Archipelago or western Greenland. Highest levels of ΣPCBs were found in ringed seal blubber samples from the Yenisey Gulf (Nakata *et al.* 1996). ΣPCB concentrations in ringed seals from Nome (Alaska) on the eastern Bering Sea (Schantz *et al.* 1993) were similar to those from the western and central Canadian Arctic. Ringed seals from Scoresbysund in East Greenland had higher mean concentrations of PCBs and DDT than those from western Greenland (Cleeman *et al.* 1996c, 1996d). Ringed seals from Jarfjord in northern Norway had about two-fold higher concentrations of ΣPCBs than those at Svalbard, but lower mean levels than a large sample analyzed by Nakata *et al.* (1996) from the Yenisey Gulf/south Kara Sea.

Higher ΣHCH levels were found in ringed seals from the Canadian Arctic than in the same species from Greenland, Svalbard, and the Yenisey Gulf. The elevated levels of ΣHCH are consistent with higher ΣHCH in seawater in the Canadian Archipelago and Beaufort Sea (section 6.6.4.1).

There is little information on levels of other POPs in ringed seals. Low concentrations of PBDEs were found in ringed seal blubber from Svalbard (Sellström *et al.* 1993). Tetrabromo-DE averaged 47 ng/g lw, while total pentabromo-isomers were present at about ten-fold lower levels. Jansson *et al.* (1993) detected low ng/g levels of chlorinated paraffins (60-70% chlorinated $C_{10}$-$C_{13}$ alkanes) in blubber of Svalbard ringed seals. There are no data for these compounds in ringed seals from the North American or Russian Arctic.

• *Harbour and grey seals*

Skaare (1996) has compared concentrations of ΣPCB (Figure 6·37), ΣDDT, and total chlordane (ΣCHL) in two coastal species, harbour seal (*Phoca vitulina*) and grey seal (*Halichoerus grypus*), from Bjørnøya, Svalbard, the Kola Peninsula, and Finnmark. Significantly lower ($p < 0.05$) concentrations of all three OC groups were found in males (both species combined) from the Kola Peninsula. Among females, mean levels of ΣDDT and ΣCHL were also lower in animals from the Finnmark/Kola regions than from Svalbard. Harbour seals from northern Norway (Vesterålen, Jarfjord) had significantly lower levels (by about two- to three-fold) than those from three locations in southern Norway (Oslofjord, Kristiansand, and Bergen areas). Harbour seals from Iceland (Luckas *et al.* 1990) have comparable levels of PCBs and DDT to those from the southern Barents Sea ('East Ice') (Annex Table 6·A18). There are no data for OC levels in harbour seals from the Canadian Arctic.

Combining the data for the Norwegian Arctic with results from the Wadden Sea (Luckas *et al.* 1990) and the coast of England (Law *et al.* 1990), a decreasing PCB concentration gradient with increasing latitude is observed for harbour seal (Figure 6·37). Concentrations may be slightly higher in Jarfjord near the Russian border (Skaare 1996).

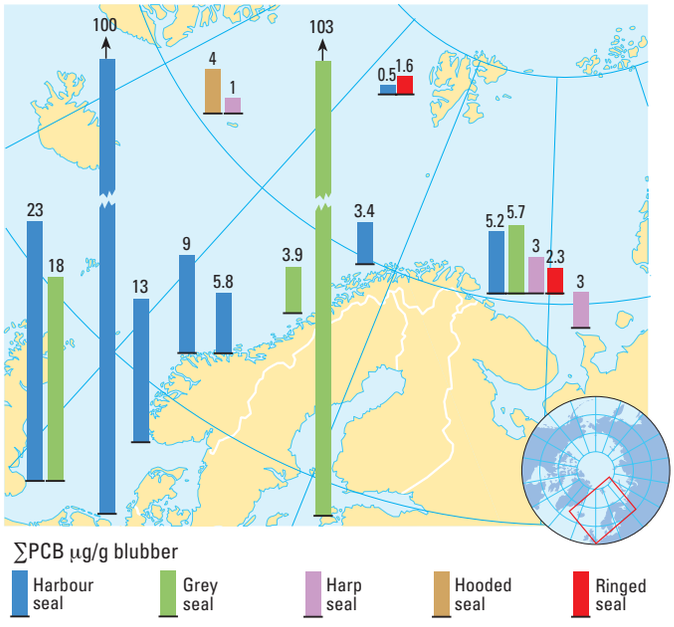



Figure 6·37. ΣPCB levels in pinnipeds in Norway (Skaare *et al.* 1990, Bernhoft and Skaare 1994, Jenssen *et al.* 1995a, Kleivane *et al.* 1995, Skaare 1996, Espeland and Skaare unpubl. results).

Grey seals from northern Norway have about 25-fold lower ΣPCB concentrations than are found in this species in the Baltic Sea (Blomkvist *et al.* 1992) and about four-fold lower than in eastern England (Law *et al.* 1990 ) (Figure 6·37).

The overall trend for PCBs and DDT in seals, which are the only contaminant groups for which anything approaching a circumpolar survey has been done, is that levels seem to increase from west to east (Annex Table 6·A18). However, additional samples are needed from the Russian Arctic to confirm this trend. Norwegian and Canadian data suggest that levels of PCBs decline from south to north in blubber of ringed seals and harbour seals. These two species are relatively sedentary and, therefore, better indicators of spatial trends within the Arctic than migratory species such as harp seal. The large data set on OCs in ringed seal blubber offers the possibility of more rigorously establishing circumpolar geographical trends of OCs after adjusting for age, sex, blubber thickness, and other factors.

• *Walrus*

Walrus (*Odobaenus rosmarus*) blubber samples from eastern Hudson Bay (Inukjuak and Akulivik), Foxe Basin (Igloolik and Hall Beach), and eastern Baffin Island (Loks Land) were found to have higher than expected levels of ΣPCB congeners (*ortho*- and non-*ortho*-substituted) and other persistent OCs (DDT, toxaphene, chlordanes, dieldrin, mirex) when compared with previous studies of walrus from Greenland and Alaska (Muir *et al.* 1995c). Samples from 19 of 53 individuals had concentrations of ΣPCBs greater than 1000 ng/g ww; the remaining individuals had much lower concentrations (50-600 ng/g ww). Local contamination was ruled out because levels of all OCs were elevated in each animal from Inukjuak, and elevated levels were also found in animals from other areas of the eastern Canadian Arctic. Skaare *et al.* (1994b) also found a wide range of concentrations of ΣPCB, ΣDDT, and chlordane-related compounds in walrus skin/blubber biopsy samples from Svalbard. Concentrations of ΣPCB (mean 11.5 μg/g; median 8.8 μg/g lipid) in Svalbard walrus were similar to those from eastern Hudson Bay, but much higher than Foxe Basin animals (Annex Table 6·A18). The results suggest that the walrus with elevated OCs are feeding at a higher trophic level than those with low levels and are probably utilizing ringed seals for a portion of their diet.

Walrus from the Thule region of western Greenland had low levels of ΣPCBs and other persistent OCs similar to those in Foxe Basin animals (Annex Table 6·A18) (Muir and Born 1996).

6.6.4.7.2. **Cetaceans**

• *Beluga* (*white whale*) *and narwhal*

Belugas (*Delphinapterus leucas*) have a nearly continuous distribution across the Russian Arctic coast, but are limited in the Atlantic to the north coast of Norway and in the Pacific to the Okhotsk Sea (Kleinenberg *et al.* 1964). They are present along the east and west coasts of Greenland and in North America, extending from Alaska across the Canadian western Arctic to a large population in Hudson Bay and among islands in the eastern Canadian Arctic. They feed in shallow estuaries on a variety of foods, including capelin, herring, cisco, sculpin, Atlantic and Arctic cod, flounder, and char, as well as invertebrates such as octopuses, squid, shrimp, and paddleworms (Banfield 1974).

The beluga may be the most mobile large mammal in the Arctic. Movement of pods is seasonal and predictable. They come into the coastal waters and estuaries in mid-summer, and winter offshore in pack ice and polynyas (Brodie 1989). These migrations are responses to offshore feeding opportunities, coastal ice formation, and the need for estuarine conditions during the summer calving period. Some general facts of the migration of major stocks are known. Tagging experiments of belugas in Hudson Bay suggest that the migratory range is 800 km, from southwest to northwest Hudson Bay (Sargeant and Brodie 1969). The Canadian High Arctic stock essentially migrates along the south coast of Devon Island. A number of extralimital observations suggest that some individuals, probably males, will wander great distances. The results of some recent tagging studies, in which individuals migrated hundreds of kilometers in unexpected directions, have emphasized that dispersal and migration are not well understood (P. Richard pers. comm., Norris 1994, Stewart and Burt 1994). Nevertheless, OC profiles have proven to be a valuable tool for stock discrimination (S. Innes pers. comm.).

Few statistically significant differences in the mean concentrations of the major OCs were found in the five Canadian Arctic beluga stocks sampled in the period 1983-1989 (Muir *et al.* 1990b). Toxaphene levels showed no geographical trends, however, ΣPCBs and ΣDDT were significantly higher ($p < 0.05$) in male belugas from Cumberland Sound than in samples from Hudson Bay or the Beaufort Sea. No significant differences were found among mean levels in females within the five stocks. Differences between stocks are best determined with male belugas because they show little variation of OC levels with age relative to females (the opposite is observed in seals). Additional samples of beluga blubber from western Greenland, the Bering/Chukchi Seas, and the western Canadian Arctic (south Beaufort Sea) have confirmed the slightly higher PCB and toxaphene levels in the Baffin Bay and southeast Baffin animals (Stern *et al.* 1994, Becker *et al.* 1995) (Annex Table 6·A18). A single beluga blubber sample from the White Sea had the highest levels of all persistent OCs found in Arctic beluga. North-south comparisons can also be made with beluga because of the presence of isolated populations in the St. Lawrence estuary and Cook Inlet (Alaska). In the eastern Canadian Arctic, ΣPCB concentrations, in samples collected from 1992-1994, averaged about 6000 ng/g ww in male belugas, and 4000 ng/g ww in blubber of females; these levels are about 12 times lower (in males) than in blubber of dead belugas from the St. Lawrence estuary sampled in 1993-1994 (Muir *et al.* 1996a). Levels of ΣPCB in belugas from Cook Inlet on the Pacific coast of Alaska ($2\,600 \pm 1140$ ng/g in males, $n = 9$) are significantly lower than in the Beaufort Sea population where mean levels exceed 5000 ng/g (Muir *et al.* 1996a).

Narwhal (*Monodon monoceros*), which are found mainly in the eastern Canadian Arctic and in western Greenland waters, have similar concentrations of PCBs and toxaphene in blubber as belugas from the same area (Annex Table 6·A18).

• *Harbour porpoise and minke whale*

Skaare (1996) has reviewed results for OCs in cetaceans sampled along the coast of Norway since 1988, and results for harbour porpoise (*Phocoena phocoena*) and minke whale (*Balaenoptera acutorostrata*) are summarized in Annex Table 6·A18. Minke whales had low levels of OCs compared with odontocetes (toothed whales) such as the beluga and harbour porpoise. This is due to their feeding primarily on invertebrates. The harbour porpoise from northern Norway had the highest levels of ΣPCBs and ΣDDT of any cetacean in Arctic waters. Like other odontocetes in the Canadian Arctic (Muir *et al.* 1990b, 1992b), the porpoises had higher proportions of lower-chlorinated PCBs, and higher concen-

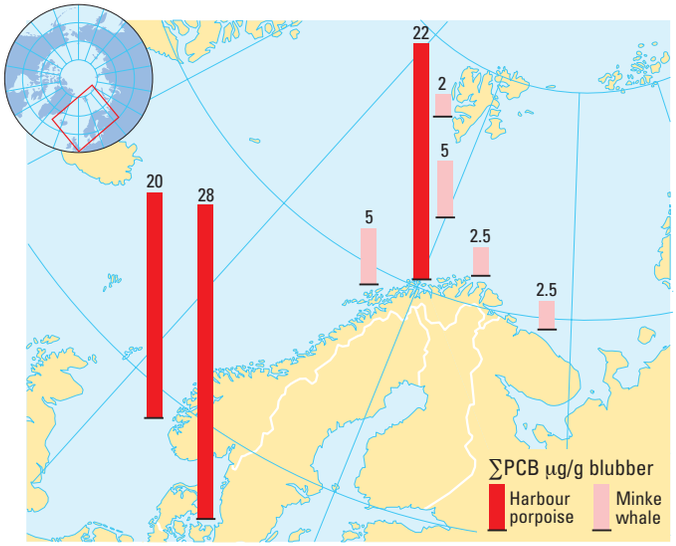



Figure 6·38. ΣPCB levels in cetaceans around Norway (Kleivane *et al.* 1995, Skaare 1996, Espeland and Skaare unpubl. data).

trations of chlordane and dieldrin than animals from southern Norway. PCB concentrations in harbour porpoise from northern Norway are similar to those found in animals from the west coast and southern Norway (Kleivane *et al.* 1995) (Figure 6·38).

### 6.6.4.7.3. Persistent OCs in other marine mammal tissues

Fewer analyses of kidney, liver, and muktuk of marine mammals have been conducted than of blubber. OC concentrations are uniformly lower than in blubber due to their lower lipid content. Low ng/g ww levels of PCBs and OC pesticides were found in liver, kidney and muscle samples of beluga and narwhal (Muir *et al.* 1992b, Muir 1995, Stern *et al.* 1994). On a lipid weight basis (beluga liver and muscle averaged 3.4 and 1.3% extractable lipid, respectively), levels of OCs were comparable to or higher than those found in blubber.

Muktuk from Beaufort Sea belugas contained low ng/g ww levels of all major classes of OC contaminants (PCBs, chlorobenzenes, OC pesticides) (Annex Table 6·A18). The lipid content of muktuk ranged from 3 to 7% which indicates that it was relatively free of the underlying blubber layer. Toxaphene was the major OC contaminant, with mean concentrations of 400 ng/g. As observed for blubber samples, PCB concentrations in muktuk are higher in males than females. Muktuk from western Greenland belugas had similar concentrations of OCs compared to Beaufort Sea samples, and concentrations were the same in males and females (Stern *et al.* 1994, Annex Table 6·A18). All animals analyzed were under 4.5 years of age so that differences between males and females would not be anticipated.

### 6.6.4.7.4. Effects of age and sex on organochlorine levels in marine mammals

As noted above, age and sex are important factors to be taken into account when comparing geographical and temporal trends in marine mammals. Most OCs, except HCH and mirex, are lower in adult female cetaceans and pinnipeds than in males because of elimination of these lipophilic compounds via lactation (Addison and Smith 1974). However, trends with age vary with species and sex. Stern *et al.* (1994) found no significant trend of OC levels with age in male beluga (70 animals were analyzed) and a substantial decrease in females after five or six years of age, corresponding to the time of first parturition. Kleivane *et al.* (1996) found ΣPCBs increased with age in harbour porpoise. In seals, PCB concentrations increase with age in males but not in females. This has been observed previously (Addison and Smith 1974), but the large number of samples from the western Hudson Bay (Arviat) (30 males, 21 females) has enabled a thorough test of the age/concentration relationship (Figure 6·39). Geographical comparisons are, thus, most easily done with adult female ringed seals and on a case by case basis for cetaceans. A major consideration, evident from the studies on ringed seals (Cameron *et al.* 1997) and harp seals (Kleivane *et al.* 1995) is the need to standardize the time of year and nutritional/reproductive status (moulting or lactating) of the seals or cetaceans.

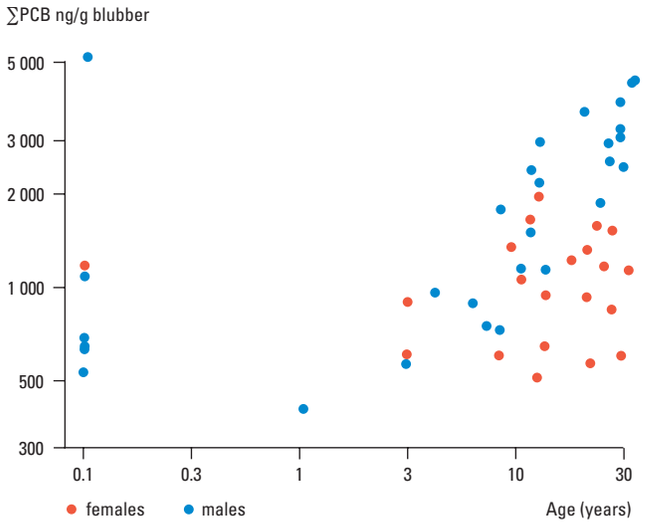



Figure 6·39. Variation of PCB with age in blubber of ringed seals (western Hudson Bay, Canada). A total of 21 females and 30 males were analyzed. The correlation with age applies to males one year and older only. Similar correlations were observed for ΣDDT and ΣCHL. Log ΣPCB vs. log age: $r^2 = 0.88$.

Modeling and measurements of mother-calf pairs has shown that in the early life of marine mammals, significant quantities of lipid-soluble contaminants may be transferred to the young via lactation (Addison and Brodie 1987, Kingsley and Hickie 1994, Espeland *et al.* 1996). Females have accumulated large burdens early in their reproductive life, and marine mammal milk is high in fat. This initial dose is further concentrated as the newborn burns fat to provide energy, before it starts to feed itself and dilute the initial intake by less contaminated food. For the female, this represents a loss of contaminants, evidenced by the fact that actively reproducing females have notably lower concentrations than mature males.

### 6.6.4.7.5. Spatial trends in non-*ortho* PCBs and PCDD/Fs in marine mammals

Information on levels of PCDD/Fs and nPCBs in Arctic marine biota is limited in comparison with other OCs (including mono-*ortho* and *ortho*-substituted PCBs). The greater complexity and higher cost of analysis of these compounds has limited the number of samples analyzed to date. All published results up to early 1995 are summarized in Annex Table 6·A19. For PCDD/Fs there is evidence of higher concentrations in Barents Sea animals, a similar geographic trend to that observed for PCBs and other OCs.

The PCDD/F profile in marine mammal blubber differs between the Canadian Arctic and Barents/Greenland Sea

animals. Ringed seals, polar bear, and walrus in the Canadian Arctic have relatively high 2,3,7,8-TCDD and low PeCDD and PeCDF levels. In Barents and Greenland Sea seals, 2,3,7,8-TCDF and PeCDF tend to be at equal or higher concentrations than TCDD and PeCDD (Oehme *et al.* 1988, 1995d, Bignert *et al.* 1989). Concentrations of 2,3,7,8-TCDF in Canadian ringed seals ranged from <2 to 7 pg/g ww which was lower than values reported for Svalbard ringed seals (9 to 13 pg/g) (Oehme *et al.* 1988). Concentrations of 2,3,7,8-TCDD in ringed seal blubber from Svalbard ranged from < 2 to 8.2 pg/g ww (Oehme *et al.* 1988) and from 2 to 37 pg/g in pooled blubber samples from various locations in the Canadian Arctic (Norstrom *et al.* 1990). Beluga and narwhal blubber had uniformly low levels of PCDD/Fs (Annex Table 6·A19), reflecting the ability of cetaceans to metabolize planar aromatics due to higher cytochrome P450 1A1 activity in comparison with seals and birds (Tanabe *et al.* 1988, Norstrom *et al.* 1992). In general, concentrations of PCDD/Fs in Arctic seals are lower than in animals from the Baltic Sea and the North Sea (Oehme *et al.* 1988, Bignert *et al.* 1989), but higher than found in Antarctic seals (Oehme *et al.* 1995c).

Unlike the PCDD/Fs, levels of nPCBs in blubber of ringed and harp seals from the Canadian Arctic, the Greenland Sea, and Svalbard are similar (Annex Table 6·A19). These comparisons are mainly for males and are therefore confounded by the effects of age. There are insufficient data at present with which to compare geographic variations of nPCBs in female seals. Daelemans *et al.* (1993) reported a mean total nPCB of 293 pg/g ww in blubber of ringed seals from Svalbard, which is very similar to results for the eastern Hudson Bay ringed seals, but lower than levels found in Danish harbour seals (Storr-Hansen and Spliid 1993). Levels of nPCBs in Arctic cetaceans, narwhal, and beluga are generally higher than in seals from the same location. Males have higher levels than females. Condition of the animals may be important in the relative proportion of the three toxic nPCBs (CBs 77, 126, and 169). Ice-entrapped belugas from the southern Beaufort Sea stock were emaciated when their blubber was collected. These animals had much higher proportions of CB 169 than 'normal' belugas in the eastern Canadian Arctic (Annex Table 6·A19). Oehme *et al.* (1995d) also found unexplained high levels of CB 169 in some harp seal blubber samples from the Greenland Sea. Highest nPCB levels in Arctic marine mammals have been found in walrus from Inukjuak (eastern Hudson Bay) which also had high ΣPCB levels. Levels in these walrus were within the range observed for harbour seal in the Danish Wadden Sea.

Estimates of toxic equivalents of 2,3,7,8-TCDD (TCDD TEQs) in Arctic marine mammals indicated that CB 126 (3,3'4,4',5-pentachlorobiphenyl) contributes 60-70% of the total TEQ in narwhal (Ford *et al.* 1993) and 30-50% in ringed seals using recent toxic equivalent factors for nPCBs and mono-*ortho* PCBs (Ahlborg *et al.* 1994). Results for TCDD and nPCBs in ringed seals from Svalbard and harp seals from the Greenland Sea are similar (Bignert *et al.* 1989, Oehme *et al.* 1995d). Asplund *et al.* (1991) also found low ng/g levels of chlorinated naphthalenes in seal blubber from Svalbard which could also contribute to 'dioxin equivalents'. There have been no other reports of chlorinated naphthalenes in Arctic animals.

### 6.6.4.8. **Polar bear**

The polar bear (*Ursus maritimus*) has received particular attention in relation to contaminants in the Arctic. As a fourth-level carnivore, biomagnification of persistent OCs, such as PCBs and chlordane-related compounds, leads to higher concentrations than in most other Arctic mammalian species.

Polar bear are distributed widely throughout the Arctic and subarctic polar regions. The ecology of polar bears is closely tied to that of ringed seals (Stirling and Archibald 1977). Densities of seal vary in response to the overall productivity of the ecosystem in different areas, and these changes also cause changes in the productivity of bears (Stirling and Øritsland 1995). Polar bears preferentially consume ringed seal blubber and skin. In addition to ringed seal, they may eat lesser amounts of bearded seal (*Erignathus barbatus*) (Smith 1980, Stirling and Archibald 1977), and occasionally prey on beluga and walrus (Lowry *et al.* 1987, Calvert and Stirling 1990). In late spring, polar bears become highly active in response to readily available young and moulting seals. The bears acquire most of their annual nutrient reserves during this period (Ramsay and Stirling 1988). Using $^{13}C$ stable isotope analysis, it has been shown that polar bears do not eat significant amounts of terrestrial food even when forced onto land (Ramsay and Hobson 1991).

Polar bears occur in low densities throughout the polar basin and are circumpolar in their distribution. Polar bears are distributed into relatively discrete populations (Bethke *et al.* 1996, Taylor and Lee 1995), although individuals can wander over several thousand kilometers in a year (Ramsay and Stirling 1986, Messier *et al.* 1992). In the Beaufort Sea, movements have been estimated to range over 10 000-23 000 $km^2$ (Amstrup *et al.* 1986); in the archipelagic habitats of Canada, they range between 2500 and 23 000 $km^2$ (Schweinsburg and Lee 1982, Schweinsburg *et al.* 1984); on Svalbard, the movement is much more variable, 69 000 ± 80 000 $km^2$ or a range of 1000 to 325 000 $km^2$ (Wiig 1995); and, in the Bering and Chukchi Seas, the movements are extensive, in the range 150 000-350 000 $km^2$ (Garner *et al.* 1994).

Polar bears mate in the spring (Wiig *et al.* 1992). Fertilized eggs do not implant before September-October, about the same time that the pregnant female enters the den (Ramsay and Stirling 1988). According to Sandell (1990), species with delayed implantation appear to be especially susceptible to deleterious reproductive effects from OC pollution. Derocher (1991) suggested that OC-induced effects on reproduction may be a factor in the decline in reproductive performance of western Hudson Bay polar bears. Cubs are born around Christmas and the female emerges from the den usually with two cubs in March-April after fasting for six months. Polar bear milk has a high fat content (Arnoud and Ramsay 1994) and lactation seems to occur more or less until weaning at 2.5 years.

PCBs and *p,p'*-DDE were first discovered in polar bear tissues in the early 1970s in the Canadian Arctic by Bowes and Jonkel (1975), a few years after they were identified as environmental contaminants (Jensen *et al.* 1969). At later dates, ΣPCBs and persistent OC pesticides were found in bears from other Arctic areas (Norstrom *et al.* 1988, 1997, Norheim *et al.* 1992).

Annex Table 6·A20 summarizes levels of OCs measured in all major polar bear studies. This includes results from Norheim *et al.* (1992), who analyzed polar bear fat and liver collected from Svalbard during the period 1978-1989, recent Svalbard data (Bernhoft *et al.* 1996), early Canadian results, results from a 1982-1984 systematic survey (Norstrom *et al.* 1988) in the Northwest Territories of Canada, and recent circumpolar data, summarized by Norstrom *et al.* (1997).

The Norwegian Polar Institute has studied the biology of polar bears in the Svalbard area since the late 1960s. The exceptionally high PCB levels in polar bears from this area were first found by Norheim *et al.* (1992), who analyzed

Table 6·16. Geographical regions used to describe OC levels among polar bear populations, relationship to Canadian management zones, and sampling dates for each region (Taylor and Lee 1995).

| Sampling region | Sampling period | Zone | Geographical location |
|---|---|---|---|
| R1 | April 1993 | – | Wrangel Island (Russia), Chukchi Sea |
| R2 | March 1988-March 1990 | – | Bering Sea and Bering Strait south of the Arctic Circle, Chukchi Sea and Goodhope Bay, Alaskan coast to 155°W |
| R3 | April 1989-May 1993 | NB (part) | McLure Strait and the adjacent Arctic Ocean |
| R4 | December 1989-May 1990 | NB (part) | Amundsen Gulf and Beaufort Sea to 135°W |
| R5 | April 1989-May 1990 | VM PC (part) | Viscount Melville Sound west of 100°W |
| R6 | December 1989-May 1990 | MC | Queen Maud Gulf and Larsen Sound |
| R7 | January 1990-May 1990 | PC (part) | Barrow Strait and Cornwallis Island |
| R8 | December 1989-May 1990 | GB | Gulf of Boothia |
| R9 | April 1989-June 1990 | PC (part)<br>BB (part) | Baffin Bay north of 72°N, Lancaster Sound,<br>Jones Sound, Kane Basin, Thule, and Ellesmere Island |
| R10 | December 1989-January 1990 | BB (part) | Southern Baffin Bay and northern Davis Strait (between the Arctic Circle and 72°N) |
| R11 | October 1989-April 1990 | FB | Foxe Basin and Hudson Strait west of 72.5°W |
| R12 | August 1989-September 1990 | WH | Western Hudson Bay (Cape Churchill area) |
| R13 | January 1990-April 1991 | SH | Eastern Hudson Bay (Belcher Islands) |
| R14 | December 1989-March 1991 | DS (part) | Davis Strait below the Arctic Circle and Hudson Strait east of 72.5°W |
| R15 | January 1990-July 1990 | – | East coast of Greenland near Scoresbysund |
| R16 | March 1990-April 1990 | – | Svalbard (Norway) |

samples from 24 bears shot or found dead during the period 1978-1989. Since 1990, samples from anaesthetized, free-ranging bears at Svalbard have been analyzed yearly for OCs at the Norwegian College of Veterinary Medicine. Including samples collected in the 1996 season, 150 bears have been sampled (fat biopsies and blood) in the Svalbard area during the month of April. Additional data on each bear sampled included length, girth, reproductive status, nutritional status, sex, and age. Initially, the research was focused on studying occurrence and levels of OCs, and the high PCB levels found earlier (Norheim *et al.* 1992) were confirmed (Annex Table 6·A20). The influence of age, sex, reproduction, and nutritional status on the OC variation in bears was also investigated. More recently, possible effects of the very high PCB levels on reproduction and survival of the cubs have been investigated. The reproductive success of Svalbard polar bears is discussed in more detail in section 6.8.3.6. Information on reproductive success, revealed by satellite telemetry, has been coupled with physiological parameters, such as thyroid hormones and Vitamin A in blood, and plasma OC levels (Bernhoft *et al.* 1996) (section 6.8.3.6). Studies on possible effects on the immune system from very high PCB exposure have also been initiated.

In a study of 85 polar bears, Skaare *et al.* (1994a) and Bernhoft *et al.* (1996) found that ΣCHL and ΣPCBs were the major OC contaminants in subcutaneous fat, similar to previous observations by Norstrom *et al.* (1988) in polar bears from the Canadian Arctic. Levels of all OCs in females were generally lower than in males (Annex Table 6·A20). In young (1-2 years old) and subadult (3-6 years old) bears, there were no differences in OC levels between sexes, except for significantly higher ΣCHL levels in subadult females than in subadult males. The highest ΣCHL levels were found in young (3.38 μg/g ww in fat) and subadult bears (3.44 μg/g ww). Levels of ΣCHL were significantly lower in adult (7-15 years) and old males (16-22 years) than in subadults. Oxychlordane constituted 72% of ΣCHL in all polar bear fat samples that were analyzed.

Mean concentrations of ΣPCB in subcutaneous fat samples from various age groups of polar bear at Svalbard ranged from 11.4 μg/g ww in juveniles to 28.1 μg/g in adult males. Extremely high PCB levels (up to 80 μg/g in fat of males and up to 36.7 μg/g in females) were found in some adults (Bernhoft *et al.* 1996). ΣPCB levels found in adult males were significantly higher than in young and adult female bears. The highly recalcitrant PCB congeners 153 and 180 constituted 62% of ΣPCB found in the bears. Nine of 14 PCB congeners detected in bear fat showed significantly higher levels in adult males than in one or more of the other groups. All PCB congeners, except CBs 99 and 138, increased with age in males. The increase was more prominent with increased chlorination degree. All congeners, except CBs 105 and 118, tended to reach a plateau or decline in old males. In females, the individual congeners tended to follow the levels in males until adulthood. After that age, usually 7-11 years, lower levels of PCB congeners 153, 156, 157, 170, and 180 were found in females than in males.

Levels of ΣHCHs in adult male polar bear fat (385 ng/g) were significantly higher than in subadults (197 ng/g) and adult females (163 ng/g). β-HCH, the recalcitrant HCH isomer, constituted 81% of ΣHCH in the 85 samples. In males, ΣHCH increased with age until about 12 years, while in females no change with age was evident.

DDE (*p,p'*-isomer) and HCB were also major individual OC contaminants in bear fat from Svalbard. DDE was the only DDT-related compound present above detection limits in bear fat. For DDE and HCB, no statistically significant differences with sex or age were observed.

Bernhoft *et al.* (1996) also analyzed OCs in maternal milk, subcutaneous fat, plasma, and blood cells of female polar bears and in subcutaneous fat, plasma, and blood cells of yearlings. Considerable amounts of OCs are transferred to the offspring via milk. Correlations of OCs in subcutaneous fat with levels in plasma and milk were significant for most OC compounds. Correlations of OC levels in plasma lipids with levels in milk were also significant, but only for lower chlorinated PCBs (especially penta- and hexachlorobiphenyls). These results indicate a more efficient OC transfer between subcutaneous lipid depot and circulatory lipids than from the circulatory system to milk, particularly for the most lipophilic compounds. The OC pattern in suckling yearlings reflects the low transfer of the higher chlorinated (hepta- and octachloro) PCBs into maternal milk. The levels of most other OCs were higher in subcutaneous depot lipid of yearlings than in that of their mothers.

OC concentrations were determined in 586 samples of subcutaneous adipose tissue from hunted polar bears and tissue biopsies from animals tranquilized for research purposes between the spring of 1989 and the spring of 1993, as part of a large collaborative circumpolar study (Norstrom *et al.* 1997). Samples were analyzed from Wrangel Island in the East Siberian Sea, eastward through the polar bears' complete range in North America to East Greenland and Svalbard. Samples were assigned to 16 regions based on knowl-

edge of the subpopulation range (Table 6·16, Norstrom *et al.* 1997). Additional information on each bear sampled included its age, sex, length, girth, and reproductive status.

Linear models (mixed analysis of variance and covariance) were used to describe and account for the effects of measured covariates, and geographic comparisons were then made with data corrected for age and sex. The linear models showed that there were significant relationships between residue levels and *Region* (16 regions, Table 6·16 and Annex Table 6·A20), *Sex* (three categories – females, females with cubs, and males), *Age* as a continuous variable, and *Ageclass* (two categories; subadults, 0-4 years, and adults, 5-31 years). However, model $R^2$ values were still low ($R^2$ being the fraction of variation in the data explained by the linear model), ranging from 0.27 for ΣCHL to 0.57 for ΣPCB, suggesting that many factors besides the ones considered in the study were important in determining OC levels. Some of the unexplained variability is believed to be due to seasonal effects (fasting, lactation), dietary factors (individual preferences, food web structure), and physical condition.

To compare OC levels among regions for the circumpolar study (Norstrom *et al.* 1998 in press), differences in the age and sex of bears sampled had to be taken into account. To do this, only the 345 adult bears were considered, and values of most OCs were standardized to represent levels in mature males, using coefficients from the linear models. For ΣCHL, data were age-standardized to males of age of 11. Actual means and standardized geometric means for ΣHCH, ΣCHL, ΣDDT, ΣPCB, and dieldrin are summarized in Annex Table 6·A20. Median (range) levels for the whole data set were as follows: ΣPCB, 7119 (1228-70 421) ng/g lw; ΣCHL, 1988 (207-15 013) ng/g; DDE, 190 (24-2821) ng/g; and dieldrin, 149 (7-835) ng/g.

Levels of ΣPCB were 46% higher in males than females, and there was no trend with age. Levels of ΣCHL were 30% lower in males than females and there was a negative trend with age in both sexes: $log_e[\Sigma CHL] = -0.02\ Age + k$. The age effects described for ΣDDT are more complex. Although the overall age effect was not significant, the *Age* × *Region* effect was, suggesting that there were different age effects within regions, or that there were age effects in some regions and not in others. ΣCHL and ΣPCB levels decreased in subadults from about twice the levels in their mothers at birth to adult levels around age 4-5. The minimal age effect thereafter suggests that the whole-body half-life of these persistent OCs in non-pregnant females and males may be of the order of a year, based on the calculated time to approach greater than 90% steady state with the diet.

Levels of various OCs in polar bear are plotted by *Region* as bars in Figure 6·40 (next page). In general, OC levels were significantly different among regions ($P \leq 0.05$). One of the most important findings was the relatively uniform distribution of the levels of some OCs over much of the study area. This is a clear indication of extensive transport and deposition of OCs to all areas of the Arctic and subarctic.

The geographical distribution of ΣPCB is notable for higher levels occurring at McLure Strait and the adjacent Arctic Ocean (R3), the east coast of Greenland near Scoresbysund (R15), and Svalbard (R16) (Annex Table 6·A20, Figure 6·40). The mean ΣPCB levels found by Norstrom *et al.* (1997) in 14 fat samples from Svalbard (22.7 μg/g lw) were similar to the mean ΣPCB of 21.9 μg/g lw for combined adult males and females found by Bernhoft *et al.* (1996).

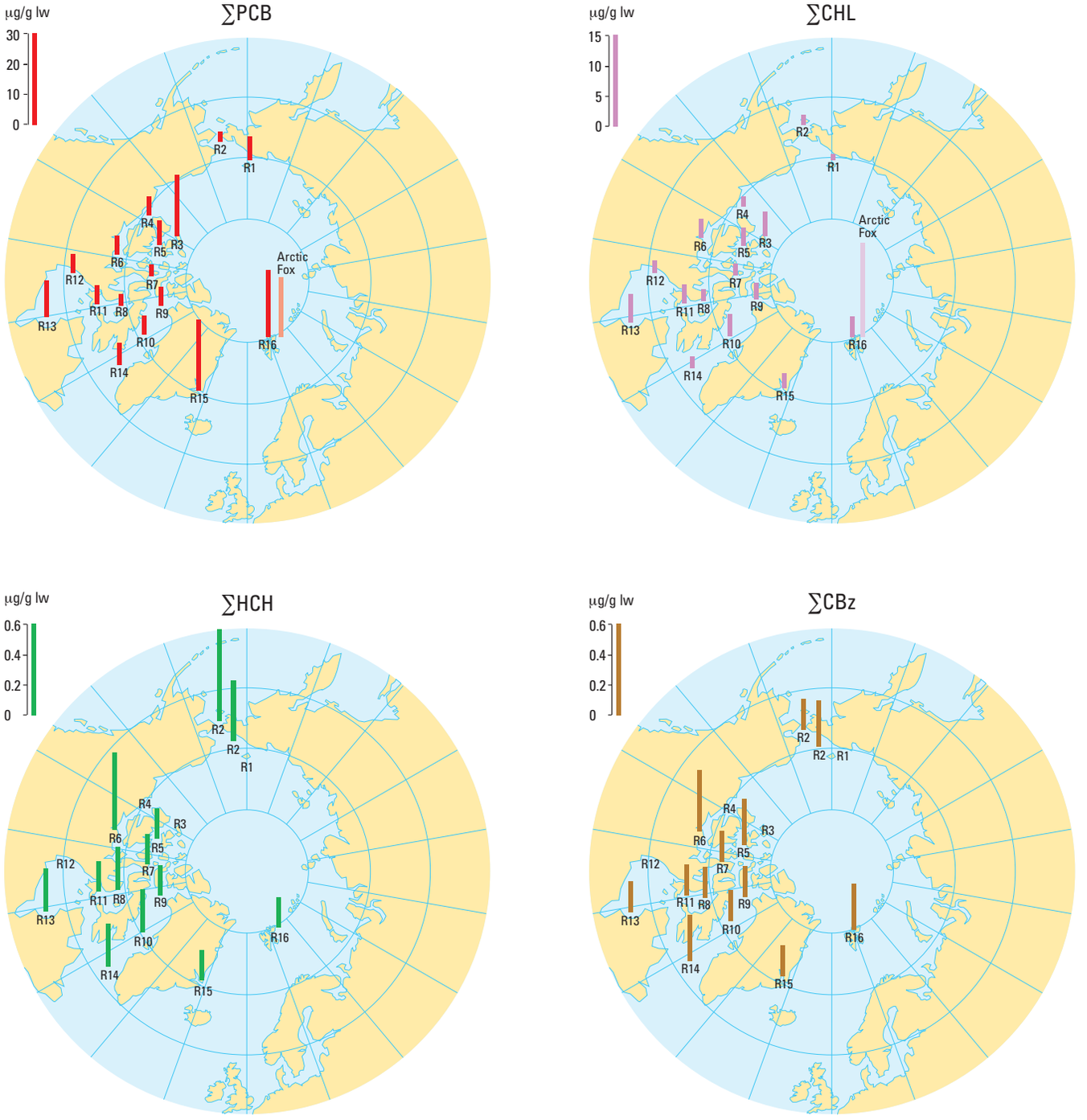
The higher ΣPCB levels at Wrangel Island in the East Siberian Sea (R1) compared to other samples in the same region may have been because this group was almost entirely composed of females with cubs just emerging from dens. Polischuk (pers. comm.) has found that ΣPCB levels in fat of females emerging from dens after six months of fasting are two times higher than in the previous autumn. The net effect of this distribution is to create a band of lower values, spreading east from Alaska along the continental coast into the Canadian Arctic Archipelago, Baffin Bay, and western Hudson Bay, surrounded by higher values in areas in the north and south, eastern Greenland, and Svalbard (Figure 6·40). ΣPCB levels were similar (20.3-24.3 μg/g lw) in bears from Svalbard (R16), East Greenland (R15), and the Arctic Ocean near Prince Patrick Island in Canada (R3), and significantly higher than most other areas. There were few other significant differences in ΣPCB levels apart from some between R5 and R13 and the *Regions* with the lowest mean levels: R2, R6, R7 and R8. There were no significant trends in geographical distribution of ΣPCB levels within the band of *Regions* across the North American coast and the Canadian Archipelago which had the lowest levels of ΣPCB (R2, R4, R6-R12, R14; Figure 6·40). Nevertheless, average levels in this band were higher in Baffin Bay and Hudson Bay ($6.27 \pm 0.631$ μg/g lw; R9-12, R14), than in the western Regions ($3.97 \pm 1.03$ μg/g lw; R2, R4, R6-8), indicating a slightly increasing west-east gradient.

The high ΣPCB levels in Arctic Ocean (R3) bears may be influenced by a different food web structure in a permanent ice environment, or by some as yet unknown feature of atmospheric or oceanic transport. There is anecdotal information that bearded seal may be more abundant in this area, and there is a possibility that the base of the ringed seal food web is more epontic than pelagic compared to areas farther east (Welch *et al.* 1992).

Significant differences in ΣCHL occurred mainly between southern Hudson Bay (R13, highest) and Wrangel Island, East Siberian Sea (R1, lowest) and other *Regions* (Figure 6·40). Levels were about 50% higher than average in the three *Regions* which had highest ΣPCB concentrations, but the highest level was found in southeastern Hudson Bay (R13), which also had the highest DDE and dieldrin levels among the *Regions*. ΣCHL was the most uniformly distributed of the OCs. Mean ΣCHL concentrations in bears from Svalbard (3.29 μg/g lw) were similar to mean concentrations in subadult and adult female bears observed by Bernhoft *et al.* (1996).

DDE levels in R2, R4, R6, and R8 were significantly lower than most other *Regions* (Annex Table 6·A20). DDE levels were higher in the eastern *Regions*, with peaks in R1 and R3 in the west and R13 in the east. As suggested for ΣPCB, the high DDE in R1 may be because this group of samples was biased towards fasting females. The high DDE in R10 is probably spurious, because it is driven by one bear out of the five sampled from this area. Dieldrin levels displayed a more pronounced west-east increasing gradient than the other OCs (Annex Table 6·A20). Levels in the Bering/Chukchi *Regions* (R1 and R2) were significantly lower than for most other *Regions*. The dieldrin distribution, as well as that of ΣCHL and DDE, strongly suggests an influence from North American sources, although ecological factors cannot be ruled out.

Total chlorobenzene levels were very uniform across the entire study area, with the exception of a peak in R6 (Figure 6·40). HCH levels were also similar among areas, but, unlike all other OCs, the highest levels were found in the western regions, especially the Bering and Chukchi Seas (R2) (Figure 6·40). This is a clear indication of ongoing contributions from Asian and southeast Asian sources (Barrie *et al.* 1992). The relatively high level of both ΣHCH and total chlorobenzenes in R8 compared to nearby regions was also noted in 1984 (Norstrom *et al.* 1988).




| | |
|---|---|
| R1 | Wrangel Island (Russia), Chukchi Sea |
| R2 | Bering Sea and Bering Strait south of the Arctic Circle, Chukchi Sea and Goodhope Bay, Alaska Coast to 155° W |
| R3 | McLure Strait and the adjacent Arctic Ocean |
| R4 | Amundsen Gulf and Beaufort Sea to 135° W |
| R5 | Viscount Melville Sound west of 100° W |
| R6 | Queen Maud Gulf and Larsen Sound |
| R7 | Barrow Strait and Cornwallis Island |
| R8 | Gulf of Boothia |
| R9 | Baffin Bay north of 72° N, Lancaster Sound, Jones Sound, Kane Basin, Thule and Ellesmere Island |
| R10 | Southern Baffin Bay and Northern Davis Strait (between the Arctic Circle and 72° N) |
| R11 | Foxe Basin and Hudson Strait west of 72.5° W |
| R12 | Western Hudson Bay (Cape Churchill area) |
| R13 | Eastern Hudson Bay (Belcher Islands) |
| R14 | Davis Strait (below the Arctic Circle) and Hudson Strait east of 72.5° W |
| R15 | East coast of Greenland near Scoresbysund |
| R16 | Svalbard (Norway) |

Figure 6·40. Organochlorine levels in polar bear adipose tissues (µg/g lw), adjusted to levels expected in 11-year-old males (after correction for age and sex), for ΣPCB, ΣCHL, ΣHCH, and ΣCBz, and in Arctic fox from Svalbard for ΣPCB and ΣCHL. Data are given in Annex Table 6·A20 and the 16 regions are described in Table 6·16.

Methylsulfone-PCB and -DDE metabolites were found in polar bear fat at levels similar to those of ΣHCH, total chlorobenzenes, DDE, and dieldrin (Bergman *et al.* 1994b). Letcher *et al.* (1994) determined $\Sigma MeSO_2$-PCB levels and PCBs in the same composite polar bear fat samples analyzed for ΣHCH and total chlorobenzenes. They found the geographical distribution to be very similar to that of ΣPCBs (Figure 6·40). Concentrations of $\Sigma MeSO_2$-PCB were 4-8% of ΣPCB. The ratio of $\Sigma MeSO_2$-PCB to ΣPCB decreased from west to east, due to an increase in the proportion of

higher chlorinated PCB congeners. Most of the PCBs which form methylsulfone metabolites have 4-6 chlorines.

In addition to the OC residues listed above, several others have been identified in polar bear tissues, but there is much more limited information on their geographical distribution, and none on temporal trends. These OCs include PCDD/Fs (Norstrom *et al.* 1990), tris(*p*-chlorophenyl)methanol (Jarman *et al.* 1992), photoheptachlor (Zhu *et al.* 1994), and toxaphene (Zhu and Norstrom 1994).

PCDD/F levels in subcutaneous fat from polar bears in the Canadian Arctic sampled in 1983-1984 ranged from 2-23 pg TEQ/g ww (Norstrom *et al.* 1990). Newer data are available on PCDD/F, nPCB, and mono-*ortho* PCB levels in polar bear liver collected in 1992-1994 from Canada (Letcher 1996, Letcher *et al.* 1996). Expressed as TCDD TEQs, levels of PCDD/Fs were 27 pg/g lw, nPCBs were 29 pg/g lw, and mono-*ortho* PCBs were 172 pg/g lw. Data on mono-*ortho* PCBs (CB 105, 118, 156, 157) in subcutaneous fat are available for polar bears from Svalbard (Bernhoft *et al.* 1996). TEQs ranged from 82-256 pg/g lw. Oehme *et al.* (1995a) determined PCDD/Fs and nPCBs in polar bear milk from Svalbard. OCDD and OCDF were the major congeners with concentrations ranging from 4-149 pg/g and 0.2-5.7 pg/g lipid, respectively (Annex Table 6·A19). Low (<1 pg/g) concentrations of 2,3,7,8-substituted tetra- to hepta-CDD/Fs were also detected. TCDD TEQs ranged from 1.0 to 3.5 pg/g lw. nPCBs were present at higher levels than the tetra- to hepta-CDD/Fs. TCDD TEQs attributable to nPCBs ranged from 3.0-9.2 pg/g lw.

### 6.6.4.9. **Arctic fox (Svalbard)**

The Arctic fox does not migrate in the true sense of the word, however, some populations feed in very different habitats at different times of the year. The coastal populations primarily scavenge the leavings from other predators on the ice in the winter, and feed on small mammals, mainly lemmings, on land in the summer.

Arctic fox from Svalbard are mainly carnivorous, feeding on leftover kill from polar bears in winter, and on small mammals and birds in summer. Very high concentrations of ΣPCB are observed in Arctic fox liver (0.5-15 μg/g ww), among the highest concentrations observed in liver of any Arctic mammal (Wang-Andersen *et al.* 1993). ΣCHL levels are also very high (Annex Table 6·A20). If lipid weight concentrations are estimated from the liver wet weight concentrations in Annex Table 6·A20 (Skaare unpubl.), by assuming that liver lipid content is 10% or lower, then the PCB levels (8.6-208 μg/g lipid) are in the same ranges as for polar bear, and comparable or higher than other marine mammals.

Wang-Andersen *et al.* (1993) reported that CB 153 and CB 180 dominated in samples, just as in polar bears. Significantly higher levels of PCBs were found in older animals (older than three years) with less than 2 cm of fat on the rump, compared to young animals (1-2 years) with more than 2 cm of fat. No data are available on OCs in Arctic fox from other circumpolar regions.

### 6.6.4.10. **Specific examples of food web transfer – marine environment**

A series of studies undertaken since 1986 (Hargrave *et al.* 1989a, 1989b, 1993, Hargrave 1994), have investigated the transfer of persistent OCs in the marine food web. The studies were conducted using facilities on the Canadian Ice Island (79-81°N) and more recently at Resolute Bay (≈75°N, 95°W). OCs were measured in epontic (under-ice) particulate matter, tissues of planktonic and benthic invertebrates, and abyssal and coastal marine fish. The Resolute site also permitted an assessment of transfer of contaminants to seabirds, ringed seals, and belugas which were sampled in nearby Lancaster Sound/Barrow Strait (Annex Tables 6·A16 and 6·A18). These measurements were also complemented with information on likely predator-prey linkages higher in the food web (see Figure 6·2).

As has been discussed in sections 6.6.4.4 and 6.6.4.5, toxaphene, PCBs, and DDT-related compounds, especially DDE, were the predominant OCs measured in epontic particulate matter, in all size classes and taxa of pelagic and benthic zooplankton and amphipods, and in tissues from Arctic cod and anadromous char (Annex Tables 6·A15 and 6·A17). In fish, chlordane-related compounds were present in slightly lower amounts than toxaphene and PCBs, while concentrations of HCH isomers and HCB were lowest. The most abundant compounds in air and seawater samples, ΣHCH and HCB, are the least prominent in Arctic marine biota from all trophic levels.

Figure 6·41 (next page) illustrates the bioaccumulation of the major OC groups at different trophic levels in the marine food webs of Lancaster Sound and Svalbard. The trophic levels were assigned based on the nitrogen ($^{15}N/^{14}N$) stable isotope ratio results of Hobson and Welch (1992). These data were not available for the Svalbard animals, therefore, they were assumed to be at the same trophic levels as those from the Canadian Arctic. A few organisms for which stable isotope data were not available, e.g., benthic amphipods, Greenland halibut and redfish, were assigned a trophic level value consistent with their known feeding relationships. Significant correlations were found between ΣPCB and trophic level (TL) at both Lancaster Sound ($\log \Sigma PCB = -0.64 + 0.99TL$; $r^2 = 0.70$; $p < 0.001$) and Svalbard ($\log \Sigma PCB = -0.62 + 1.06\,TL$; $r^2 = 0.62$; $p < 0.001$). Correlations for ΣCHL and TL were also significant for both food webs. Slopes for ΣPCB and ΣCHL versus TL of approximately 1.0 and 0.72, respectively, indicate a ten-fold increase for ΣPCB and a five-fold increase for ΣCHL at each level of the food web. ΣDDT concentrations at Svalbard and Lancaster Sound were also significantly correlated with trophic level if polar bear was omitted. DDT does not biomagnify between seals and polar bears.

A more detailed analysis of this data would show that strong relationships are observed for other individual recalcitrant components of each OC group, such as CB 153 (2,2',4,4',5,5'-hexachlorobiphenyl) (Norstrom 1996) and nonachlors, but other components, for example, trichlorobiphenyls, would show no trend. Biomagnification of various OCs thus depends on structural properties of individual components. For example, in the case of PCBs, marine mammals, especially pinnipeds and polar bears, are capable of metabolizing congeners with vicinal unsubstituted *meta-*, *para-* positions, as well as *ortho-*, *meta*-positions (Boon *et al.* 1994). In general, the more highly chlorinated, less water soluble compounds are preferentially accumulated because of a lower degree of metabolism. Similarly, for the complex mixture of toxaphene, which contains compounds with 6-10 chlorine molecules, the heavier $Cl_8$ and $Cl_9$ bornanes accumulate in Arctic zooplankton and benthic amphipods when compared to those present in seawater (Bidleman *et al.* 1993).

Biological factors are also important in modifying observed biomagnification factors (BMF) values. For example, in marine mammals, there is a positive correlation between tissue concentrations and age. Comparisons with lower trophic levels and fish also show that more short-lived species may have lower concentrations (Hargrave *et al.* 1992, 1993).

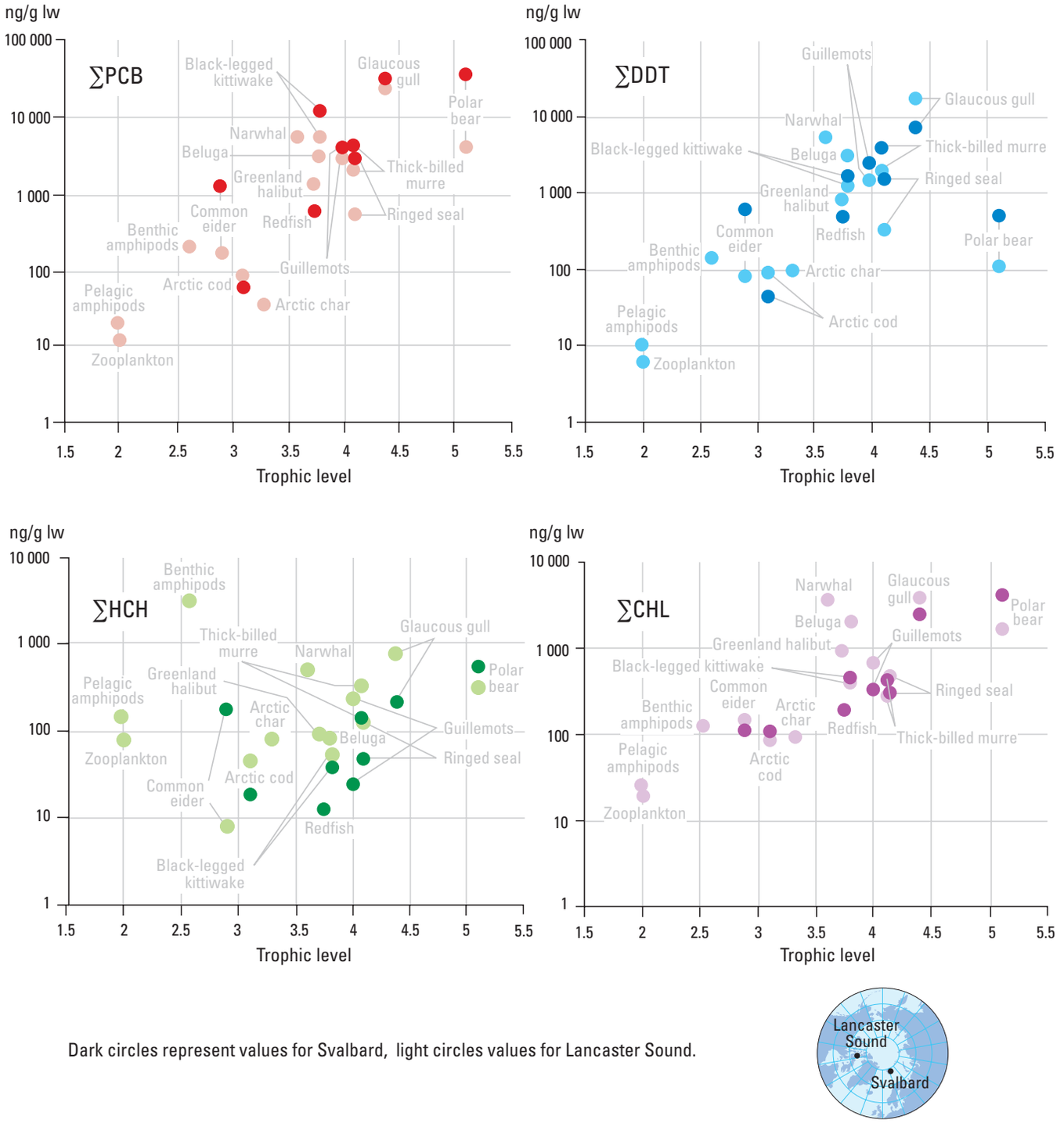



Figure 6·41. Correlations between concentrations (lw) of major persistent OCs and trophic level in the marine food web for Svalbard and Lancaster Sound. Trophic level data were assigned based on Hobson and Welch (1992). Trophic level of Greenland halibut and redfish were assumed to be 3.5, that is, higher than cod, but lower than seals. Data for invertebrates are from Hargrave (1994). Other results are from Annexes 6§A3, 6§A15, 6§A17, and 6§A20.

Zooplankton and pelagic amphipods generally have a one- to two-year long life cycle and these species, feeding on microalgae and suspended particulate matter, accumulate less of the most abundant OCs than do more long-lived fish and benthic amphipod species.

The combined importance of life span and food source in affecting OC body burdens was evident in data for the large benthic lysianassid amphipod *Eurythenes gryllus*, which can contain concentrations that equal or exceed those in marine mammals. The life span of these organisms is not well known, but it may exceed ten years, which is much greater than annual values for pelagic crustaceans (Hargrave *et al.* 1992). The food of these scavengers is thought to be tissues of dead animals, and the high levels of OCs could be explained if carcasses of marine mammals are a major component of their diet. In this case, lysianassid amphipods function as top predators in the Arctic marine food web. In shallow water, smaller-sized amphipods in pelagic and epontic populations are an important food resource for fish and seals. *E. gryllus* is restricted in its distribution to deep water where there is a refuge from predation due to the low abundance of fish.

BMFs were calculated for the marine food webs of Svalbard and Lancaster Sound (Canadian Archipelago) using information on likely predator-prey linkages in the Arctic marine food web combined with OC concentrations (Table 6·17). The data for OCs and fraction lipid in biota used for these calculations are presented in the Annexes. BMFs for

Table 6·17. Biomagnification of persistent organochlorines in Arctic marine food chains.

| | Biomagnification factor (lipid/lipid basis for biota) | | | | | | |
|---|---|---|---|---|---|---|---|
| Compound/ location | Zooplankton/ water | Arctic cod/ water | Seal/ Arctic cod | Gull (eggs)/ Arctic cod | Kittwake(eggs)/ Arctic cod | Beluga/ Arctic cod | Polar bear/ seal |
| *Svalbard* [a] | | | | | | | |
| ΣCBz | – [b] | $5.0\times10^4$ | 3.3 | 32 | 17 | – | 3.6 |
| ΣHCH | – | $5.8\times10^5$ | 7.8 | 12 | 2.1 | – | 4.1 |
| ΣCHL | – | $3.6\times10^7$ | 3.9 | 24 | 4.5 | – | 10 |
| ΣDDT | – | $1.2\times10^6$ | 36 | 172 | 37.1 | – | 0.3 |
| ΣPCB | – | $4.3\times10^6$ | 48 | 525 | 214 | – | 14 |
| *Lancaster Sound* [c] | | | | | | | |
| ΣCBz | $1.4\times10^6$ | $2.7\times10^6$ | 1.7 | 40 | 8.8 | 18 | 2.2 |
| ΣHCH | $2.6\times10^4$ | $1.0\times10^4$ | 7.6 | 17 | 1.2 | 1.9 | 0.9 |
| ΣCHL | $2.3\times10^6$ | $6.2\times10^6$ | 5.8 | 49 | 5.7 | 25 | 3.7 |
| ΣDDT | $1.0\times10^7$ | $8.9\times10^7$ | 3.9 | 208 | 15 | 36 | 0.3 |
| ΣPCB | $5.5\times10^5$ | $8.5\times10^6$ | 7.0 | 313 | 68 | 38 | 7.2 |
| Toxaphene | $7.7\times10^6$ | $1.1\times10^7$ | 0.3 | – | – | 15 | – |

a. Results for Svalbard: Bernhoft *et al.* 1996 (polar bear); Oehme *et al.* 1988 (ringed seal, except for ΣCBz and ΣCHL results taken from 'west ice' harp seals (Skaare 1996)); Savinova *et al.* 1995b (seabirds); Killie and Dahle 1996a (Arctic cod); Chernyak *et al.* 1995 (seawater).
b. No results for zooplankton available in the Svalbard area.
c. Results for seawater from Bidleman *et al.* (1995b) and Falconer *et al.* (1995a); Lancaster Sound invertebrate data from Hargrave (1994); marine mammal data from Annex Table 6·A18; polar bear data from Norstrom *et al.* (1997 in press); seabird data from Annex Table 6·A16.

various pairs of biota at Svalbard and Lancaster Sound are quite similar in most cases. For example, BMFs for Arctic cod to glaucous gull (eggs) were 525 for ΣPCBs at Svalbard and 313 at Lancaster Sound. BMFs for PCBs and ΣDDT between beluga/narwhal and their prey, are similar to those reported for cetaceans in the North Sea (Duinker and Hillebrand 1983) and in the northwest Pacific Ocean (Tanabe *et al.* 1984).

The seal/bear BMF for HCH is lower at Lancaster Sound than Svalbard, consistent with higher ΣHCH concentrations in seal blubber at the former site. In general, the BMFs for HCH were much lower than for the other OC groups. ΣHCH concentrations also showed no correlation with trophic level. The lower bioaccumulation of HCH isomers is consistent with the more rapid rate of elimination and lower bioconcentration factors reported for these compounds in laboratory studies with freshwater fish (Niimi 1987). Tanabe *et al.* (1984) observed a BMF of $3.7\times10^4$ for ΣHCH from water to striped dolphin in the northwestern Pacific Ocean, which is close to the BMF calculated for water to beluga ($6.3\times10^4$). Low seal-to-bear BMFs for ΣDDT at both locations reflect the capacity of polar bears to metabolize DDT-related compounds.

There are large uncertainties in the BMFs between lower food web organisms, such as zooplankton and Arctic cod. The BMFs are based on mean concentrations and, therefore, have uncertainties of at least a factor of 2, but less than a factor of ten. With data presently available, however, the observed patterns of biomagnification between organisms and for various compounds are sufficient to account for high OC levels observed in marine fish and mammals.

### 6.6.4.11. **Summary and conclusions: Marine environment**

#### 6.6.4.11.1. **Abiotic environment**

The information now available on POPs in seawater, ice, snow, suspended sediment, and bottom sediments is now far superior to that which was available for previous assessments of marine pollution in the Arctic (Muir *et al.* 1992b). Recent data confirm that the relative abundance in pelagic Arctic seawater is α-HCH > HCB > γ-HCH ≈ toxaphene > chlordanes ≈ PCBs > DDTs, as described by Bidleman *et al.* (1990). An exception seems to be the Russian Arctic seas, where the order is reversed, ΣPCBs > ΣDDT > ΣHCH > ΣCHL > chlorobenzenes. ΣPCB concentrations in some seawater samples are so high, for example, 15 ng/L (Annex Table 6·A10), that they are difficult to reconcile with data from other studies (e.g., Hargrave *et al.* 1988, Iwata *et al.* 1993), however, they are consistent with measurements based on suspended particulate matter in seawater and with reports of elevated DDT and PCBs in suspended sediments of some Russian rivers (e.g., the Ob River). If confirmed by future measurements, this would imply major inputs of DDT and PCB to the Arctic Ocean from Russian sources. The PCB levels in seawater are elevated when examined from the perspective of environmental quality guidelines. For example, PCB concentrations exceed the guideline limit for protection of freshwater aquatic life of 17 pg/L (USEPA 1995) and guidelines of 1 ng/L (OMEE 1993) for total PCB concentrations in surface waters.

Highest levels of ΣHCH in the world's oceans are found in the Arctic Ocean, especially in the Beaufort Sea and Canadian Arctic Archipelago. ΣHCH levels measured in the late 1980s to early 1990s appear to increase in a smooth gradient with latitude from the tropical western Pacific Ocean to the Arctic Ocean. Wania and Mackay (1996) have suggested that this is evidence of the 'cold-condensation' effect. Other less volatile OCs (e.g., chlordanes, PCB, DDT) were present at lower concentrations in the Bering/Chukchi Seas than at more temperate latitudes.

The transport of these contaminants by sea ice either in ice and overlying snow or associated with sediment particles embedded in sea ice could result in their release in marginal ice areas. Pfirman *et al.* (1995) make an interesting case that the release of these particulates following melting in the marginal ice areas in the Greenland and Barents Seas may be an important mechanism for focusing contaminants from a wide area of the Arctic into these regions. Higher ΣPCB levels east of Greenland and especially in the Svalbard area in polar bears and several marine mammal species (e.g., ringed seals, harp seals) may be due to a combined influence of long-range atmospheric transport from North America and Europe plus the melting of ice transported from the Laptev/Kara Seas. There is insufficient evidence to confirm this and clearly it should be a priority for future work.

While evidence for transfer of OCs via ice may be lacking, there is evidence that the elevated concentrations of ΣHCH, ΣDDT, and ΣPCB in Russian rivers (on SPM or in whole water) and nearshore seawaters has given rise to higher concentrations of these POPs in surficial sediments, particularly in the Baydaratskaya, Ob, Pechora, and Khatanga Gulfs. Circumpolar coverage of OCs in surface grab samples of

marine sediments is relatively good in Norwegian and Russian waters, especially in the Barents Sea. In general, concentrations of all OCs in marine sediments are extremely low in comparison with freshwater sediments. Most sites have ΣPCB concentrations less than 1 ng/g dw. These sediment concentrations are generally low from an environmental quality point of view. They do not exceed sediment ERL values for PCBs (Table 6·14) associated with low probability of effects (Long *et al.* 1995).

A distinct difference between offshore and nearshore sediments is evident at locations along the coast of Norway between Bergen and Tromsø and along the Russian coast. Relatively high levels of ΣHCH, ΣPCB, and HCB were found at some sites on the Norwegian coast compared to open ocean sites in the Norwegian Sea. Data are lacking on OCs in marine sediments from the Canadian and Alaskan Arctic, except for a limited number of samples from the southern Beaufort Sea, the Cambridge Bay area, and the Bering/Chukchi Seas.

Levels of PCDD/Fs have been determined in marine sediments from northern Norway (near Kirkenes), in the Mackenzie River Delta area, and in the Barents Sea. PCDD/Fs in the Barents Sea were 10 to 20 times lower than those in the northern North Sea. There was no evidence of direct contamination of nearshore Barents Sea sediments by PCDD/Fs emitted from a smelter on Bøkfjord near Kirkenes in northern Norway. PCDD/F isomer patterns were very similar for both the Barents Sea and North Sea samples and indicative of combustion sources. PCDD/F homologue patterns in estuarine sediments from the Mackenzie River Delta region of Canada differed from those in freshwater sediments upstream and also from typical North American patterns in which OCDD predominates. The sources of PCDD/Fs at the Mackenzie Delta location are unknown. TCDD TEQ levels in most marine or estuarine sediments exceeded Canadian environmental quality guidelines for protection of aquatic life of 0.09 pg/g dw, but did not exceed the Norwegian Environmental Authority 'background' levels of 30 pg/g TEQs.

Measurements made in coastal regions of northern Norway show sediment contamination by TBT in several harbors (Berge 1995). TBT concentrations are highly variable, as they are elsewhere, and are probably related to shipping activities and local rates of flushing and sedimentation.

#### 6.6.4.11.2. **Modeling**

Results of mass balance modeling of HCH suggest that the Arctic Ocean is in steady state with respect to γ-HCH inputs and is exporting α-HCH. Inputs and outputs of HCHs are dominated by ocean current advection, with about a 15% contribution by atmospheric processes, largely air-sea gas exchange (Figure 6·30). In the future, we can expect to see a decline in the large pool of α-HCH in the Canada Basin due to drainage out through the Canadian Archipelago and to a lesser extent by sea-to-air volatilization. Outgassing will also result in a slow decline of both HCH isomers in waters of the Eurasian Basin and the regional seas, assuming that atmospheric concentrations continue their downward trend.

The fugacity ratios for HCH measured in the Canadian Archipelago (1992-1994), the Bering/Chukchi Seas, and on the transpolar cruise in 1994 were greater than 1.0 at most stations. This implies that northern waters are now oversaturated and outgassing HCHs to the atmosphere. Toxaphene and HCHs which are trapped under the polar cap are thus 'ghosts of the past' and will be slowly drained, largely through the Canadian Archipelago, on a time scale of decades.

Although the modeling results for toxaphene are highly uncertain, they suggest that inputs and outputs of the compounds to the Arctic Ocean are roughly in balance. There are numerous uncertainties in the modeling of toxaphene. For example, there appear to be no data on toxaphene in marine sediments and inputs from rivers are also largely unknown. Other factors, such as limitations in icecover data, compound the uncertainty in the toxaphene loading calculations, therefore, at present, it is unlikely that atmospheric loadings of toxaphene can be estimated to better than an order of magnitude. Further measurements, especially of concentrations in the European Arctic Ocean waters, are critical to making more accurate budgets for toxaphene.

There are also large uncertainties in the inputs and outputs of PCBs to the Arctic Ocean. However, it is clear that riverine inputs and sedimentation are much more important input and removal processes, respectively, for PCBs than for HCH and toxaphene. Using average concentrations of 5 ng/L PCBs in Eurasian rivers, fluxes of 15 tonnes/y are estimated. These inputs represent 24% of the total inputs estimated for PCBs. Although the 5 ng/L concentration is highly uncertain, levels one-tenth as large (that is, equivalent to North American rivers) would still give rise to significant inputs. Thus, knowledge of riverine inputs from Eurasian sources is critical for accurate estimates of PCB loadings to the Arctic Ocean. By comparison, ocean current inputs are less significant because of low surface water PCB concentrations.

#### 6.6.4.11.3. **Biota**

Recent studies of contaminants in Arctic anadromous and marine fish, seabirds, and marine mammals have greatly expanded the information available on levels of persistent OCs. Geographic coverage of important species identified in the AMAP monitoring plan, such as Arctic cod, ringed seal, and polar bear, is good in the western hemispheric Arctic (Alaska, Canadian Archipelago, West and East Greenland, and the Norwegian and Barents Seas). There is limited information on OCs in marine biota from the Kara and Laptev Seas.

Most of the persistent organic pollutants originally identified for monitoring in biota in the AMAP plan have been determined. Typically, data are available for HCB, HCH isomers, major chlordane and DDT-related compounds, and at least seven PCB congeners. Information on toxaphene, especially in biota from the Eurasian Arctic Ocean, is much more limited. Toxaphene was found to be the major OC contaminant in muscle of Arctic cod from the Canadian Arctic. Very limited information from analyses of anadromous char and whitefish from Russian locations suggests that toxaphene may also be an important contaminant in these areas.

Data on nPCBs and PCDD/Fs is very limited compared to that for OC pesticides and *ortho*-substituted PCBs. Where non-*ortho*-substituted PCBs and PCDD/Fs have been measured in the same sample of marine biota, TCDD TEQs are mainly due to the PCBs. This is the case for fish, marine mammals, and polar bears. No PCDD/F or nPCB data were available for seabirds in the Arctic.

In general, lipid weight PCB levels in freshwater fish are very similar to levels in anadromous and marine fish, with the major differences being related to trophic level. For example, predatory fish, such as lake trout and burbot (freshwater) and marine gadiformes (cod-like fish), have higher OC levels than other fish.

Although circumpolar coverage for any one species is poor, and sample numbers very limited in many key locations, the results for OCs in marine invertebrates and fish indicate generally higher levels of PCBs and ΣDDT in biota from Russian waters. Evidence for this includes higher ΣPCB, ΣDDT, and chlorobenzene levels in bivalve mussels from the Ob and

Baydaratskaya Gulfs (southern Kara Sea) than in samples from Iceland or Greenland; higher ΣPCB levels in zooplankton in waters of the European Arctic than at the highest latitudes in the Canada/Eurasian Basins; highest levels of PCBs in *Coregonus* sp. from Russian rivers compared to Canadian rivers; and, higher ΣPCBs in Arctic cod from the southern Novaya Zemlya area compared to Arctic cod from the Greenland Sea. While sample numbers from Russian waters are generally too small, especially for anadromous species, to make general conclusions about spatial variations of OC levels in Russian waters, the results are consistent with higher levels of PCBs and ΣDDT reported in surface sediments and seawater from this region.

Although bivalve mussels are widely used for monitoring POPs in marine environments, there are insufficient data in Arctic waters with which to discern spatial trends. This is particularly the case with TBT.

The levels of OCs in Atlantic cod liver from the Barents Sea were lower than the levels found in cod from Haltenbanken in the Norwegian Sea and cod from the northern parts of the North Sea. The lowest OC concentrations were found in livers of cod from different stocks in Icelandic and Faeroese waters.

Relatively high levels of OCs were found in Greenland halibut liver and muscle. Levels in muscle are three to five times higher than wet weight levels in sea run char muscle and 15-20 times higher than Arctic cod (whole fish). Lower ΣPCB levels were found in Greenland halibut muscle from the Norwegian and Barents Sea. ΣPCB concentrations in Greenland halibut muscle do not exceed guidelines of 2 μg/g for import of fish in the European community.

Berg *et al.* (1996) provided the first data on OCs in a number of important deep sea fish. OC levels in liver of these deep sea Arctic species were similar to or slightly higher than concentrations in Atlantic cod caught at the same depth in the northwest Atlantic (off Newfoundland/Labrador). They concluded that predatory fish were distinguished from shallow depth pelagic feeders, such as Arctic cod, by lower levels of more water soluble OCs, such as HCH, and higher proportions of highly chlorinated PCB congeners. This reflects the fact that more highly chlorinated PCBs are preferentially sorbed to sinking particles, whereas HCH in seawater is mainly in the dissolved phase with much lower dissolved concentrations at depth.

Surveys of OC levels in eggs and tissues of Arctic seabirds show that seabirds breeding in the High Arctic are contaminated with a similar suite of organic contaminants as those breeding in temperate regions. Glaucous gull, herring gull, black-legged kittiwake, cormorant, and puffin generally have the highest lipid weight OC levels. DDE concentrations were highest in glaucous gulls from the Canadian High Arctic (1580 ng/g ww) and herring gull from western Finnmark, Lofoten, and Tromsø (440, 530, and 1000 ng/g ww, respectively) and Brünnich's guillemot from Svalbard. These levels are higher, on a lipid weight basis, than concentrations in seals from the same areas.

Although circumpolar coverage is incomplete, the picture that emerges from the results of OC analyses in Arctic seabirds is that the Barents Sea may be more contaminated with PCB and DDT than the Canadian High Arctic. There is no information on OC contaminant levels in seabirds from the Alaskan Arctic or the Chukchi/East Siberian Seas with which to assess the geographic extent of this trend. Lipid weight levels of most OCs in black-legged kittiwake eggs from Svalbard are higher than in eggs from a site in the High Arctic in Canada. DDT and PCB levels in thick-billed murre eggs are also higher in samples from Svalbard, Hornøya, and the Kola Peninsula compared to samples from the High Arctic and eastern lower Arctic sites of Canada. However, HCH levels are higher in thick-billed murre from Canada than from Norwegian sites.

All POPs, organochlorines as well as brominated organics, that have been detected in tissues of marine biota in temperate waters (that is, the Baltic Sea, the North Sea, the Gulf of St. Lawrence, the Mediterranean) have also been detected in Arctic pinnipeds and cetaceans. Data are available for concentrations of major PCB congeners (all studies have determined CB 28/31, 52, 101, 118, 153, 138, 180, and most have determined more congeners), DDT and chlordane-related compounds, HCH isomers, and HCB. Less frequently measured are the chlorobornanes (toxaphene components) and cyclodienes (dieldrin, endrin). There are very few data on toxaphene in Arctic marine mammals from Greenland, Norwegian, or Russian waters. PCDD/Fs have been determined, along with non-*ortho*-substituted PCBs, in seals from Arctic Canada, northeast Greenland and Svalbard, but information on their spatial trends is limited. Brominated diphenyl ethers and brominated biphenyls were also identified in ringed seal blubber from Svalbard, but no comparable data are available for other locations.

A west to east increase in some OC levels, from North America to western Russia, is indicated in blubber of harp seals and possibly in ringed seals. ΣPCBs and ΣDDT levels in harp seals from northeastern Greenland ('west ice' north of Jan Mayen Island) were found to be about two- to three-fold lower than those from northern Norway (Jarfjord, Skjånes) and western Russian waters ('east ice'). Highest levels of ΣPCBs and ΣDDT in Arctic ringed seals were found in samples from the Yenisey Gulf. ΣPCB levels in (female) harp seals collected at Salluit in Hudson Strait were similar to levels in females from northeastern Greenland, but 2.5-fold lower than mean levels in females from the southern Barents Sea in western Russia.

The large number of ringed seal populations studied has revealed the large degree of complexity of geographic trends for this species. Ringed seals from Hudson Bay had higher concentrations of ΣPCB and ΣDDT than those in the central Canadian Archipelago and western Greenland. However, ringed seals from eastern Greenland (Scoresbysund), Svalbard, and northern Norway had similar ΣPCB levels to those in Hudson Bay. The high levels in seals from Russian waters, compared with other Arctic locations, are consistent with observations of higher PCB levels in seabirds, marine fish, river water SPM, and some nearshore marine sediments in the western and central Russian Arctic. Additional samples are needed from the Russian Arctic to confirm this trend.

An exception to this geographic trend in most OCs is the higher level of ΣHCH in Canadian Arctic ringed seals than in the same species from Greenland and Svalbard. In Arctic cod, samples from the Canadian Arctic Archipelago had about two to three-fold higher levels of ΣHCH (46 ng/g lw) than Arctic cod from the Barents Sea (14-36 ng/g lw). A similar situation is seen for thick-billed murre. The elevated levels of ΣHCH are consistent with higher ΣHCH in seawater in the Canadian Archipelago and Beaufort Sea.

Norwegian and Canadian data suggest that levels of PCBs in blubber of ringed seals and harbour seals decline from south to north. The two seal species are relatively sedentary and, therefore, better indicators than migratory species such as harp seal. The large data set on OCs in ringed seal blubber offers the possibility of more rigorously establishing circumpolar geographical trends of OCs after adjusting for age, sex, blubber thickness, and other factors.

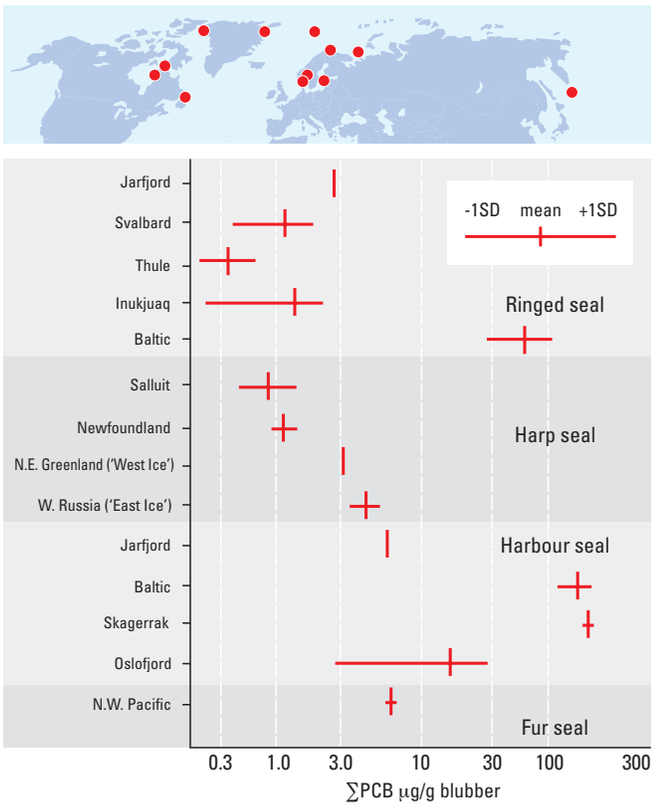



Figure 6·42. Comparison of ΣPCB concentrations in seal blubber from various Arctic regions with results from the same species in temperate waters. Vertical bars represent means and the horizontal bars the standard deviation, where available. Results are generally for females except where a combined result for males and females was reported. Data are from Annex 6§A18.

North-south differences in levels of ΣPCB in seals are summarized in Figure 6·42. Female adult ringed seals from the Baltic Sea have about 40 times higher levels of PCBs than ringed seals from Svalbard and 100 times higher than seals from Thule in western Greenland. Harbour seals from Jarfjord have about 20 times lower ΣPCB levels than the same species in the Baltic or Skagerrak Seas. Female fur seals from the northwest Pacific coast of Japan (Tanabe *et al.* 1994a, 1994b) have similar ΣPCB levels to harbour seals in Jarfjord and harp seals in the southern Barents Sea.

Slightly higher PCB and toxaphene levels are observed in the Baffin Bay and southeast Baffin beluga compared to those from the Chukchi/Bering Seas and from the western Canadian Arctic (southern Beaufort Sea). A single beluga blubber sample from the White Sea had the highest levels of all persistent OCs found in Arctic beluga. Unfortunately, no other samples of beluga from Russia have been analyzed.

North-south comparisons can also be made with beluga because of the presence of isolated populations in the St. Lawrence estuary and Cook Inlet (Alaska). ΣPCB concentrations, in beluga blubber from the eastern Canadian Arctic (about 6 µg/g lw in male belugas, and 4 µg/g in females) were about 12-times lower (in males) than in blubber of dead belugas from the St. Lawrence estuary. Levels of ΣPCB in belugas from Cook Inlet on the Pacific coast of Alaska were about two-fold lower than in the Beaufort Sea population.

Harbour porpoise from northern Norway had the highest levels of ΣPCBs and ΣDDT of any cetacean in Arctic waters. Like other odontocetes in the Canadian Arctic, the porpoises had higher proportions of lower chlorinated PCBs, and higher concentrations of chlordane and dieldrin than animals from southern Norway. PCB concentrations in blubber of harbour porpoise from northern Norway are lower than levels in animals from the west coast and southern coast of Norway.

Decreasing PCB concentrations with increasing north latitude are also observed for harbour seal when results for the Norwegian Arctic are combined with data for seals from the Wadden Sea and the coast of England. PCB concentrations in harbour and grey seal blubber may be slightly higher in Jarfjord near the Russian border than along the Norwegian Sea coast of Norway.

Fewer analyses of kidney, liver, and muktuk than of blubber of marine mammals have been conducted. This reflects the fact that OC concentrations are uniformly lower in these tissues than in blubber, corresponding to their lower lipid content. However, from the point of view of dietary exposure of indigenous peoples, there is a need for information, especially for muktuk (skin plus some fat), which is consumed as part of the traditional whale hunt. There are apparently no data on OC levels in muktuk from Greenland.

Levels of persistent OCs in polar bear tissues have been the focus of detailed studies at Svalbard and in the Canadian Arctic. Extremely high PCB levels (up to 80 µg/g in fat of males and up to 36.7 µg/g in females) were found in some adults from Svalbard. ΣPCB levels found in adult males were significantly higher than in young and adult female bears.

The highest ΣCHL levels were found in young (3.38 µg/g in fat) and subadult bears (3.44 µg/g). Levels of ΣCHL were significantly lower in adult (7-15 year) and old males (16-22 years) than in subadults, which suggests an increased capacity with age to metabolize chlordane-related compounds. Indeed, the metabolite oxychlordane constituted 72% of ΣCHL in all polar bear fat samples that were analyzed.

Considerable amounts of OCs are transferred to polar bear offspring via milk. Correlations of OCs in subcutaneous fat with levels in plasma and milk were significant for most OC compounds, indicating a more efficient OC transfer between subcutaneous lipid depot and circulatory lipids, than from the circulatory system to milk, particularly for the most lipophilic compounds.

PCDD/F levels in Canadian polar bear fat sampled in 1983-1984 ranged from 2-23 pg TEQ/g ww (Norstrom *et al.* 1990). More recent Canadian data from 1992-1994 gave PCDD/F, nPCB, and mono-*ortho* PCB levels in liver of 27 pg TEQ/g lw, 29 pg/g lw, and 172 pg/g lw, respectively (Letcher 1996, Letcher *et al.* 1996). TEQs from mono-*ortho* PCBs (CB 105, 118, 156, 157) in subcutaneous fat from Svalbard polar bears ranged from 82 to 256 pg/g lw (Bernhoft *et al.* 1996), which is in the same range as for the Canadian polar bears. Oehme *et al.* (1995a) determined PCDD/Fs and nPCBs in polar bear milk from Svalbard. TEQs from PCDD/F were 1.0-3.5 pg/g lw, and for nPCBs 3.0-9.2 pg/g lw.

The circumpolar study of OCs in polar bear fat by Norstrom *et al.* (1998 in press) shows generally increasing OC concentrations from west to east, similar to that observed in several other marine species. Higher ΣPCB levels were found in bears from eastern Greenland and Svalbard, in agreement with results of Bernhoft *et al.* (1996) from the Svalbard population. This trend, which is also seen in ringed seals and harp seals, as well as in seabirds such as glaucous gull, may be due to the combined influence of long-range atmospheric transport from North America and Europe. Another possible factor is transport of contaminants in sea ice and overlying snow or associated with sediment particles embedded in sea ice derived from the Russian continental shelf. Pfirman *et al.* (1995) have suggested that release of these particulates, following melting in the marginal ice areas in the Greenland and Barents Seas, may be an important mechanism of focusing contaminants from wide areas of the Arctic into these regions.

ΣCHL and HCB in polar bears were more uniformly distributed over the study area. This result is consistent with the

finding of lower geographical variation of ΣCHL and HCB in air and seawater in the northern latitudes than in tropical areas (Iwata *et al.* 1993). The lower ΣCHL levels in polar bears from Wrangel Island and the Alaskan Beaufort Sea area indicate that ΣCHL loading is less in the Chukchi and Bering Seas than in the rest of the Arctic. This is consistent with results in seawater, for which high ΣCHL levels are found in the Canadian Archipelago and Barents Sea compared with the Chukchi Sea.

Concentrations of ΣPCB and ΣCHL observed in Arctic fox liver are among the highest observed in liver of any Arctic mammal. On a lipid weight basis, assuming that liver lipid content is 10% or lower, the PCB levels (8.6-208 μg/g lw) are in the same range as observed in polar bear, and comparable or higher than in other marine mammals.

BMFs estimated for various pairs of biota in the marine food webs at Svalbard and Lancaster Sound are quite similar in most cases. Highest BMFs were found between glaucous gull/kittiwake (eggs) and Arctic cod. BMFs of some OCs were lower at higher trophic levels, reflecting increased biotransformation in top predators. For example, the BMFs for ΣDDT from ringed seal to polar bear and from Arctic cod to ringed seal were less than one, as was the BMF for toxaphene for Arctic cod to ringed seal.

Significant correlations were found between ΣPCB and ΣCHL concentrations and trophic level for the marine food webs of Svalbard and Lancaster Sound. The slopes of the relationships were very similar implying a similarity in the pathways of transfer of these persistent contaminants in both regions. The results indicate an increase of ten-fold for ΣPCB and five-fold for ΣCHL at each level of the food web. ΣDDT also increased about seven-fold with each level of the food web, except between seals and bears.

## 6.7. Temporal variation in POP levels

The accurate monitoring of temporal trends in both the abiotic and biotic environments is necessary in order to determine the correlation between contaminant inputs and risk to biota in an area. It must be determined whether remedial actions reduce inputs of contaminants into the environment. Temporal trends measured in biota can also serve as warning signals if contamination levels increase, or if levels do not change over time in response to remedial actions. In addition, the verification of many models depends on informative time trend measurements.

A critical question in the assessment of persistent organic pollutants in the Arctic is, therefore whether concentrations in both abiotic and biotic media are increasing or decreasing. Consequently, we have considered temporal trends separately from the discussion of prevailing concentrations and spatial trends. With a few exceptions, assessment of temporal trends of OCs in the Arctic is very difficult because most measurements are recent. The Arctic was long regarded as a clean, remote area, removed from the pollution of the urbanized parts of the world. For this reason, studies in which contaminants were measured seldom lasted more than a few years; many involved sampling in only a single year and samples were not archived. Also, up until the early 1980s, analytical methods for OCs used packed column GC and different methods of quantification, for example, ΣPCBs as Aroclor or Phenochlor equivalents compared with current congener-specific analyses. Adjustments can be made for the different methodologies, however, some comparisons still have a high degree of uncertainty. Archived samples have provided some information about temporal trends in contaminant levels, but samples are limited.

The high degree of variability of contaminant levels in tissues makes it difficult to identify and statistically confirm temporal trends. As discussed previously (section 6.6), numerous factors including an organism's size, age, fitness, reproductive status, individual food preferences or habits, and many environmental factors influence OC levels in biota. Environmental factors that differ between years, particularly temperature, unusual climatic conditions, and productivity, are a major cause of variation, making it even more difficult to detect temporal trends.

Ice cap records are potentially very useful for elucidating long-term temporal trends in POP deposition. Unlike lakes, the ice cap receives snow directly, and there are no terrestrial processes that must be considered when interpreting the record. Information about short-term (less than 50 year) temporal trends of OCs are also available from lake sediment cores and from biota in the terrestrial, freshwater, and marine environments.

Observations about temporal trends in contaminant levels in both the abiotic and biotic Arctic often coincide with temporal trends in biological effects. These trends are discussed in section 6.8.

### 6.7.1. Air and precipitation

#### 6.7.1.1. Temporal trends of OCs in air

Concentrations of ΣHCH in Arctic air have been measured in the Bering/Chukchi Seas and at several locations in the Canadian Arctic Archipelago since 1979, generally in the summer months. Declining concentrations of ΣHCH were observed, from more than 900 pg/m$^3$ in the early 1980s to 100 pg/m$^3$ in the early 1990s (Figure 6·43) (Jantunen and Bidleman 1995, Bidleman *et al.* 1995b). The results suggest

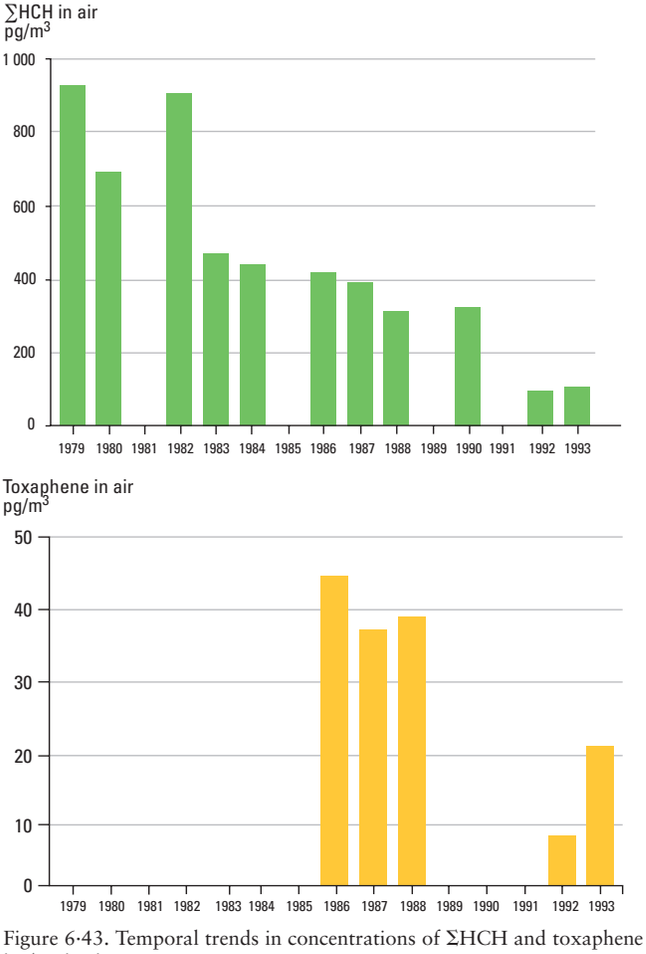



Figure 6·43. Temporal trends in concentrations of ΣHCH and toxaphene in Arctic air.

a nine-fold decline over 14 years. This decline in airborne concentrations has been paralleled by a significant, but much smaller decline in ΣHCH in surface seawater of about 3% per year (Bidleman *et al.* 1995b).

Air monitoring from 1984 to 1992 on Svalbard also showed a consistent decline in ΣHCH. Oehme *et al.* (1995b) found lower mean concentrations for α-HCH in spring 1992 at Ny-Ålesund compared to results from the same months in 1984. When concentrations for 1984 and 1992-1994 are considered, the decline may be as much as two-fold. Oehme *et al.* (1995b) also observed declines in α-HCH at two sites in mainland Norway between 1984 and 1992. However, γ-HCH levels were higher in 1992 at all three Norwegian air-monitoring sites than in 1984, reflecting the increased use of lindane and declining use of α-HCH as use of technical HCH products ceased in Europe during the 1980s.

Although the number of samples from the 1980s was small, recent atmospheric measurements suggest that a decline in toxaphene has also taken place. A direct comparison between summertime measurements in 1986-1988 (Ice Island, Canada and the Bering/Chukchi Seas) and 1992-1993 (Resolute Bay, Canada and the Bering/Chukchi Seas) suggests that toxaphene levels decreased by a factor of 3 over this time period (Figure 6·43). In these studies, toxaphene was sampled by the same method (filter-polyurethane foam), the same air pumps and volume calibration procedures were used, and the analysis of all samples was done by GC-NIMS using the total peak area method of quantification. A second comparison can be made between the 1980s NIMS measurements and the 1992-1994 adjusted values from Tagish and Alert (Table 6·6). This comparison suggests that the recent toxaphene levels are 1.5-2.5 times lower.

#### 6.7.1.2. Long-term trends in OCs in precipitation – the Agassiz Ice Cap

Gregor *et al.* (1995) examined a 30-year record of ΣPCB residues in the Agassiz Ice Cap (80°49'50''N, 72°56'30''W) on Ellesmere Island in Canada. Winter is an important period for the atmospheric transport of contaminants into the north due to the strong Siberian anticyclone which causes winds to flow mainly from the Eurasian continent into the Arctic. Snow is an effective scavenger of contaminants, probably even of hydrophobic compounds at low temperature. In addition, the Agassiz Ice Cap is believed to provide a credible record of pollutant deposition for the following reasons: (i) average summer melt affects only about 3% of the winter snow layer, resulting in negligible redistribution of ions between layers (Barrie *et al.* 1985); and (ii) annual snow accumulation at this latitude and elevation represents, on the average, more than 75% of the total annual precipitation (Woo *et al.* 1983). The seasonal specific conductance record of the firn reflects the trace constituent composition of the atmospheric aerosol (Barrie *et al.* 1985), which undergoes a strong seasonal variation with maximum ion concentrations and therefore specific conductance during the Arctic winter (Rahn and Shaw 1982, Hoff and Barrie 1986).

The ΣPCB (60 congeners) flux (ng/m$^2$/y) profiles from samples representing 30 years of deposition are shown in Figure 6·44. ΣPCB concentrations over the period of the record ranged from 1.2 to 6.7 ng/L. Mean annual ΣPCB deposition (±1 standard deviation) to the ice cap over the 30 years was 406 (±187) ng/m$^2$/y. The general temporal pattern of deposition is a period of high deposition prior to 1968/69; in 1968/69 there was a significant decrease in deposition followed by a slow, consistent increase until 1979/80 when a further decrease in deposition occurred.

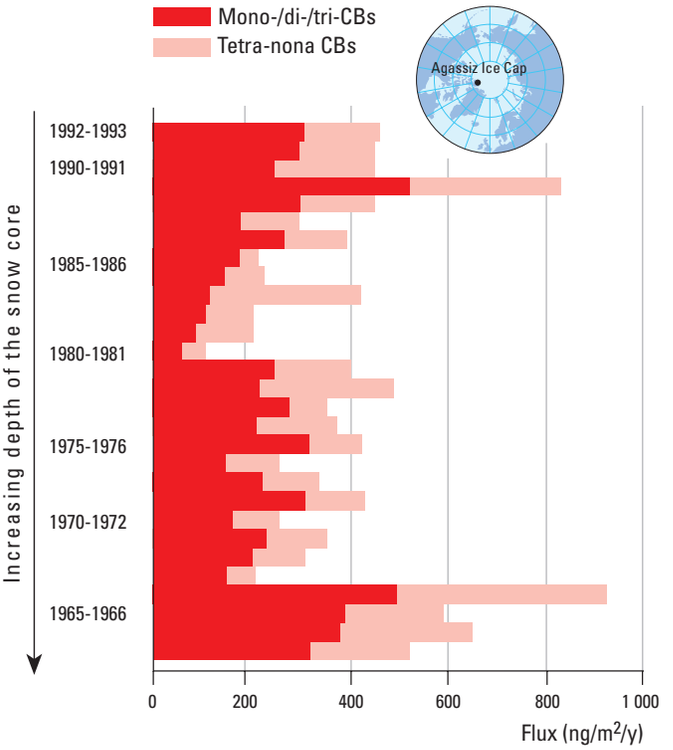



Figure 6·44. Deposition of PCBs, as mono/di/trichloro- and tetra-nonachlorobiphenyls) in snow cores from the Agassiz Ice Cap (northern Ellesmere Island, Canada) (Gregor *et al.* 1995). Combined light and dark shaded sections of each bar indicate ΣPCB flux.

The minimum for the period of record was 91 ng/m$^2$/y in 1980/81. The maximum annual deposition for the period of record was 930 ng/m$^2$/y in 1967/68. Subsequently, ΣPCB deposition rose again with a local maximum of 848 ng/m$^2$/y in 1989/90. The mean annual flux for the last three years of record (1990/91, 1991/92, and 1992/93) is 465 ng/m$^2$/y, which is not significantly different but moderately higher than the period of record mean flux (406 ng/m$^2$/y). This indicates the absence of any overall temporal trend for the time period from 1964/65 to 1992/93. The fluxes of lower chlorinated ΣPCB congeners dominated with only infrequent detections of congeners with eight or more chlorines (Gregor *et al.* 1995).

### 6.7.2. Terrestrial environment

#### 6.7.2.1. Temporal trends of PCBs in moss across Norway

Lead *et al.* (1996a) analyzed samples of the epigeic moss *Hylocomium splendens* collected in 1977, 1985, and 1990 from 46 remote sites across Norway for 37 PCB congeners (Annex Table 6·A3). ΣPCB concentrations decreased with time in all samples from all locations, reflecting the global decline in the manufacture and use of these compounds. In the north of Norway, mean ΣPCB concentrations in coastal areas fell from 21.2 ng/g dw in 1977 to 6.9 ng/g in 1990, and in inland areas from 27.7 ng/g to 7.9 ng/g. Over the same time period, ΣPCB in the south of the country decreased from 52.1 ng/g to 9.5 ng/g. As moss depends entirely on the atmosphere for delivery of nutrients and lacks both cuticle and internal transport mechanisms, this reduction is indicative of a decrease in atmospheric PCB concentrations.

A latitude-dependent change in the congener profile was noted. Although the relative importance of the higher chlorinated PCBs increased at all sites, it did so most in samples from the north of Norway (Annex Table 6·A3). Lead *et al.* (1996a) tentatively interpret this as evidence for differences

in PCB congener cycling in the environment according to volatility. They believe that this is consistent with the cold condensation hypothesis (Wania and Mackay 1993).

Lead *et al.* (1996b) also reported temporal trends for the ΣPCB content of United Kingdom soils between 1951 and 1993. They found increasing ΣPCB concentrations up to the late 1960s/early 1970s, after which there was a substantial decline. There was an increase in the relative importance of the heavier, more highly chlorinated, homologue groups over this time period. The authors believe that PCBs are being lost by volatilization from temperate soils. There is then an accumulation of the heavier compounds in colder regions. For the less chlorinated groups, however, temperatures seem to be high enough, even in the north of Norway, for revolatilization to occur. A latitudinal fractionation, driven mainly by lower summer temperatures, particularly in the Arctic regions of Norway, is thus thought to be occurring.

### 6.7.2.2. Temporal trends of OCs in reindeer in Sweden

A study at Abisko, Sweden, on a reindeer herd with a defined spatial range, provided information on temporal trends of POPs in the terrestrial environment. Muscle samples from three-year-old male reindeer (n = 10 per year) were collected annually before the rut (Odsjö *et al.* 1996 unpubl.). The study started in 1983 following measures taken in Europe during the 1970s to stop the use of DDT and PCB. Slopes for the rate of decline of levels of PCB congener 138, α-HCH, and HCB in the reindeer were negative and significantly different from zero at $p < 0.05$ (Figure 6·45). However, substantial between-year variation existed. The trends are similar to those seen in freshwater fish from Swedish Arctic lakes (section 6.7.3.2).

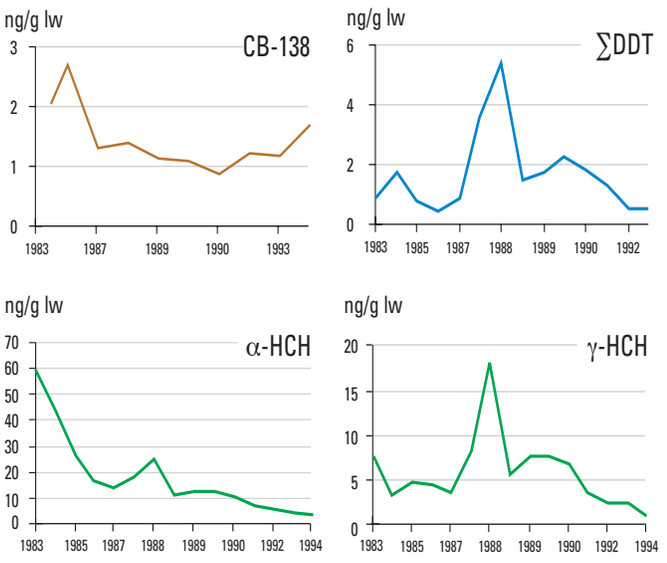



Figure 6·45. Temporal trends in OC levels (ng/g lw) in reindeer from Abisko, Sweden. Levels are usually based on a pool of ten samples (M. Olsson unpubl. data).

### 6.7.2.3. Temporal trends of OCs in birds of prey

The decline of populations of birds of prey in both temperate and Arctic regions of the world since the introduction of OC pesticides is well-documented (Ratcliffe 1967, Peakall 1976a, Newton 1979, Peakall *et al.* 1990). Their recovery after the ban of DDT and the reduced use of PCBs has also been reported. Examples of these studies are described below.

#### 6.7.2.3.1. The North American peregrine falcon

Johnstone (1994) reported that there has been an overall decrease of contaminant levels in plasma from peregrine falcons at Rankin Inlet since the early 1980s (Annex Table 6·A5). Mean *p,p'*-DDE levels in peregrine falcon eggs declined from 7.6 μg/g (ww) in the 1980s to 4.5 μg/g in the 1990s. ΣPCB (as Aroclor 1254 : 1260, 1 : 1) showed no significant change (means of 8.7 μg/g ww in 1981-1985 and 8.31 μg/g ww in 1991-94). Dieldrin, oxychlordane, and HCB also showed no change in concentration over the 10-13 year period.

A review by Peakall *et al.* (1990) of measurements of OCs in falcon eggs from northern Canada from the mid-1960s to 1987 suggests that ΣDDT levels in both F*alco peregrinus anatum* and *F. tundrius* eggs remained constant at levels near 12-13 μg/g ww (Annex Table 6·A5), over the period 1966 to 1987. Levels of heptachlor epoxide (heptachlor is a major component of technical chlordane) increased in all peregrine eggs from the Canadian north, coinciding with increased use of chlordane in the 1970s and 1980s (Peakall *et al.* 1990) (Annex Table 6·A5). These temporal trends generally matched those in gyrfalcon (*Falco rusticolus*) eggs from northern Canada except that one gyrfalcon egg sample from 1966 had more than 20 times as much DDE as samples obtained in the mid-1970s (Annex Table 6·A5). This may have been a reflection of use in southern Canada, where DDT was used for black fly and mosquito control until 1970.

#### 6.7.2.3.2. Eurasian birds of prey

The Fennoscandian population of peregrine falcon (*Falco peregrinus*) inhabits both Arctic and southern areas of Fennoscandia. The population declined from 2000-3500 pairs pre-1950 to about 65 known pairs in 1975 (Lindberg 1995a), however, in 1994, 360-410 pairs were found. Concentrations of DDE, PCB, and dieldrin have declined in both the northern and southern populations between 1984 and 1994 (Lindberg 1995a). Studies on the Swedish Arctic population of peregrine falcon revealed a decline in concentrations of DDE and PCB since 1972. Lindberg *et al.* (1985) and Lindberg (1995a) compared egg material (n = 201) collected during 1972-1981 and 1991-1994 (n = 30), and found a decrease in concentrations from 22.8 and 12 μg/g ww of ΣPCB and *p,p'*-DDE, respectively, to 14.5 and 2.9 μg/g ww. Concentrations of DDTs and PCB were not significantly different between eggs from the northern and the southern populations. Factors explaining this are the importance of the wintering areas on egg contaminant concentrations, the more aquatic-based diet of the northern population, as well as the fact that the prey of the northern population largely consists of migratory species (Lindberg *et al.* 1985, Lindberg 1995b).

#### 6.7.2.3.3. Gyrfalcon in Iceland

Òlafsdóttir *et al.* (1995) compared contaminant levels in gyrfalcon in early (1966-1973) and later (1979-1993) samples. Taking the effects of age into account, the levels of ΣPCB and ΣDDT appear very similar over the period of about 15 years. However, the sample size is small in view of the fact that sex, age, and annual variation are sources of variability in the data.

### 6.7.2.4. Temporal trends of OCs in mustelids in Sweden

Median values for ΣDDT and ΣPCB concentrations in 123 otter muscle samples from northern Sweden, including Arctic areas, collected during the 1970s, 1980s, and 1990s, were

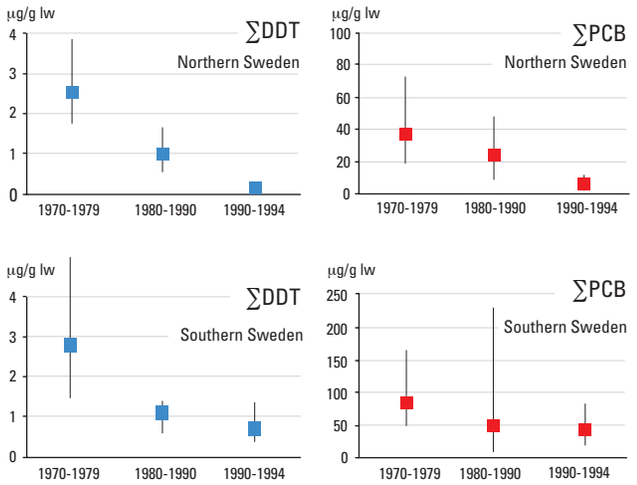



Figure 6·46. Temporal trends in PCB and DDT levels (µg/g lw ± 95% confidence limits) of otter muscle in Sweden. Levels are based on means of 26, 16, and 28 samples (northern Sweden) and 29, 9, and 15 samples (southern Sweden).

2.6, 1.0, and 0.14 µg/g lw for DDT and 38, 25, and 7.5 for PCB µg/g lw, respectively (Olsson *et al.* 1996a, 1996b). This decline is illustrated in Figure 6·46.

A study of the Norwegian coastal otter population, inhabiting predominantly the northern coastal areas of the Fennoscandian Peninsula, could not reveal any temporal trend when studying the time period 1978-1990. The material used comprised 38 juvenile otter (older than two years) (Christensen 1995a, 1995b, Christensen and Heggberget 1995).

### 6.7.3. Freshwater environment

Information about temporal trends of OCs in the freshwater environment comes from freshwater sediments and from fish data.

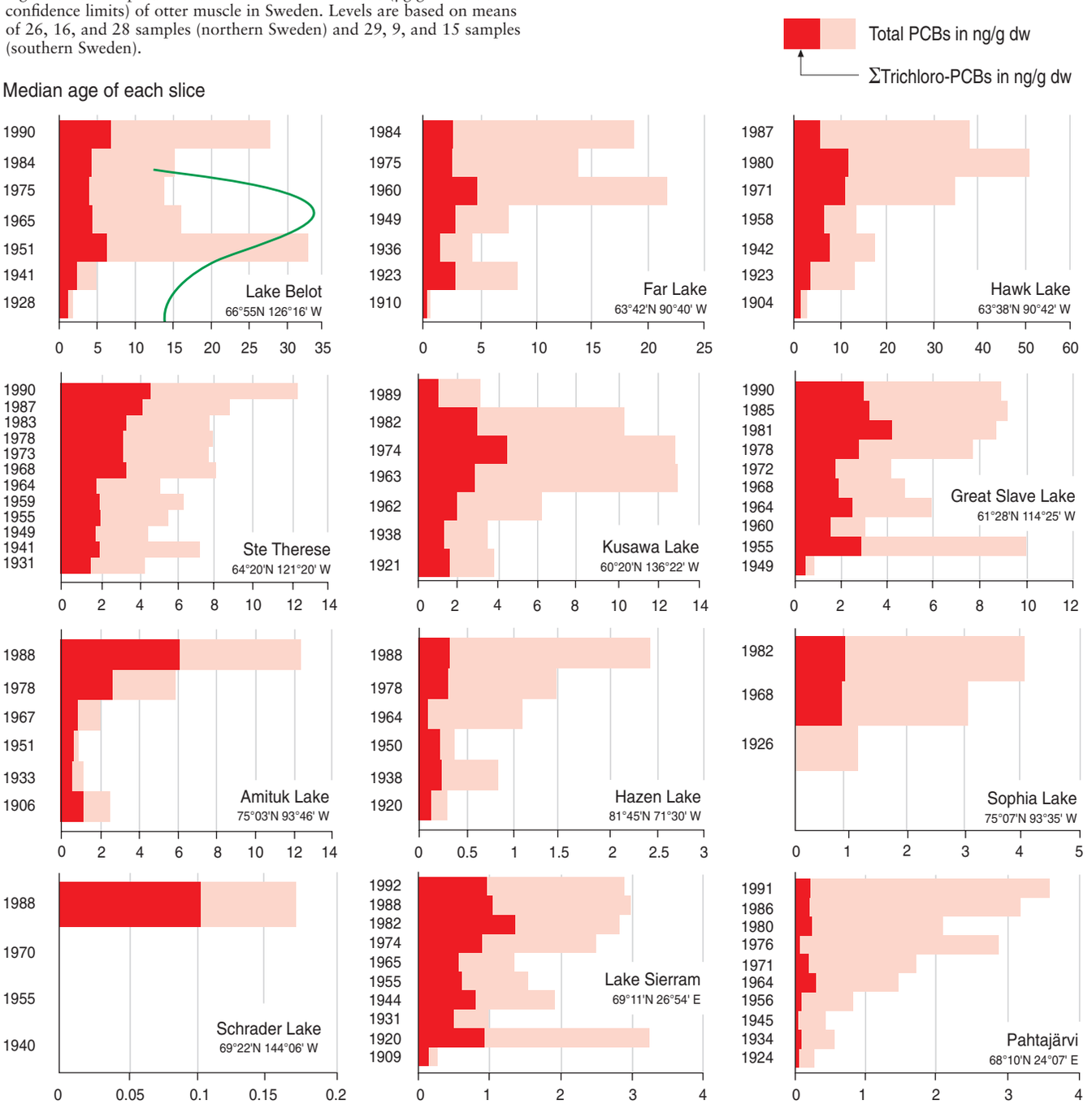



Figure 6·47. Historical profiles of PCBs (trichloro- and all other) in slices from dated sediment cores in Alaska, Canada, and Finland. Median age of each slice is given on the vertical axis. The green line indicates the source function of PCBs in the USA and in Europe, that is, the approximate duration and relative amount used (no units). Source function data is from Rapaport and Eisenreich (1988).

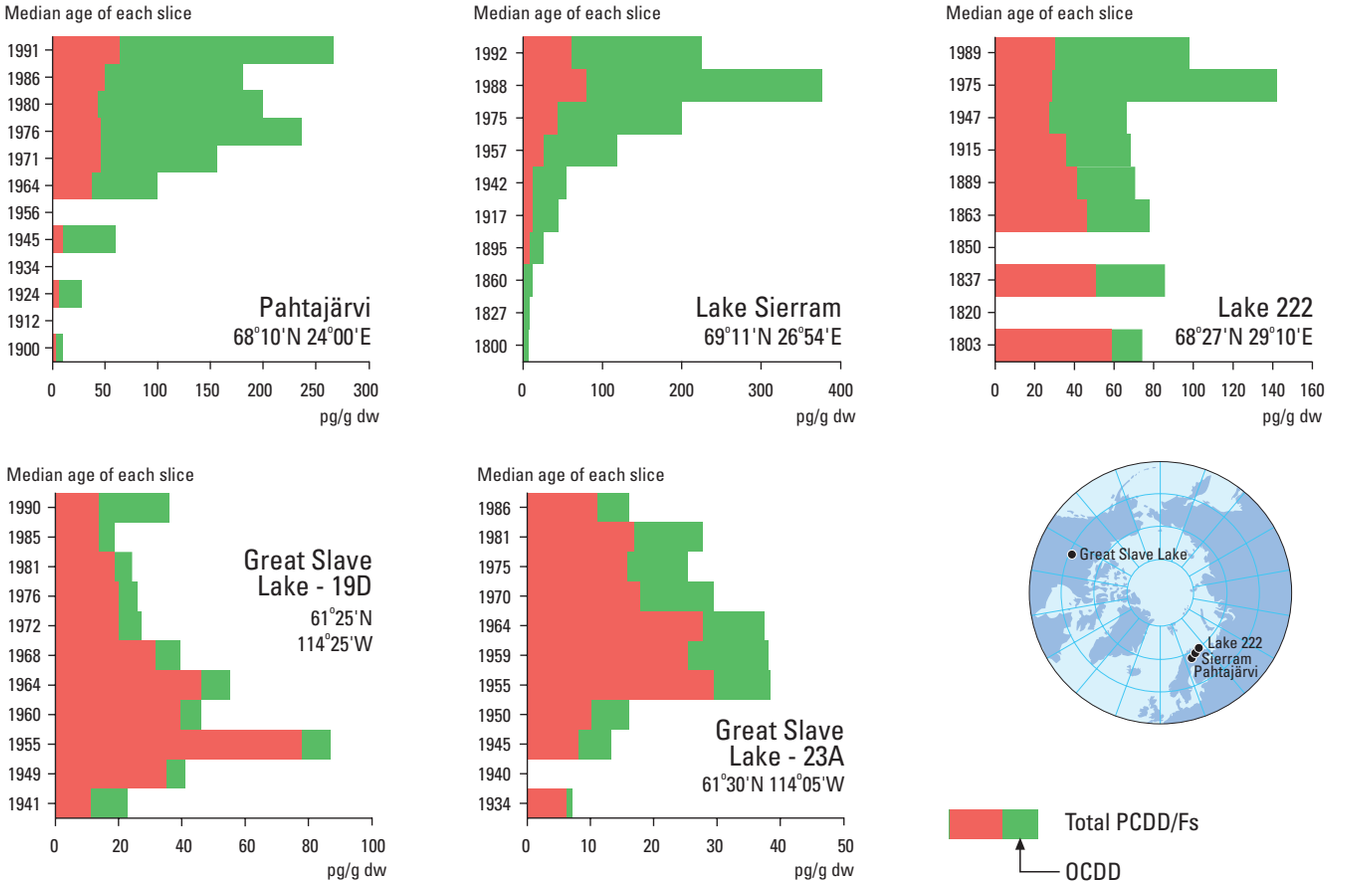



Figure 6·48. Concentration (pg/g dw) profiles for octachlorodioxin (OCDD) and total PCDD/Fs in dated sediment cores from Arctic Canada and Finland. ΣPCDD/Fs = sum of 2,3,7,8-substituted tetra- to octachloro-DD/Fs plus non-2,3,7,8-substituted congeners. Median age of each slice is given on the vertical axis.

### 6.7.3.1. **Lake sediments**

#### 6.7.3.1.1. **Historical profiles of PCBs in lake sediments**

Analysis of dated sediment cores has been used to infer the depositional history of ΣPCBs and current inputs (fluxes in ng/m$^2$/y) in the Great Lakes region of North America (Swackhamer *et al*. 1988, Eisenreich *et al*. 1989, Jeremiason *et al*. 1994) and in lakes in western Europe (Sanders *et al*. 1992, 1993, Vartiainen *et al*. 1997). The sediment records from Arctic lakes, therefore, could provide information on temporal trends of deposition of these hydrophobic contaminants in the Arctic. However, interpretations of sediment records can be complicated by direct input from non-atmospheric sources and dynamic lake processes, including bioturbation, sediment focusing, and resuspension. Factors unique to Arctic lakes, such as long periods of ice cover and low sedimentation rates, may limit inputs to bottom sediments and make them a less significant reservoir for hydrophobic organics than temperate lakes (Diamond 1994).

Sediment core profiles of PCBs for 12 lakes are shown in Figure 6·47 in which concentrations (ng/g dw) are plotted against median age of each sediment slice. The green line in Figure 6·47 represents the atmospheric input function of PCBs derived by Rapaport and Eisenreich (1988) from PCB accumulation rates in peat cores taken in the mid-latitudes of eastern North America. The USA and British sales curves of PCBs also follow this function, although results are available only as far back as the mid-1950s (Brinkman and DeKok 1980, Sanders *et al*. 1992). Most of the subarctic (60-70°N) lakes show the appearance of PCBs in the 1940s, in reasonable agreement with the source function. Profiles in the sediment cores from Belot, Great Slave, Kusawa, Hawk, and Far Lakes show subsurface maxima suggesting maximum inputs in the 1970s and early 1980s, corresponding to maximum PCB use in industrialized areas. Results from three dated sediment cores collected in northern Finland also show post-1950 deposition of PCBs with maxima in the 1970s and 1980s (Vartiainen *et al*. 1997). Low concentrations of ΣPCB are also seen in older slices from these cores (and in Pahtajärvi, Sierram, Far, Kusawa, and Hawk Lakes). They are generally less than 20% of maximum concentrations and lie within the intrinsic time resolution of the core, or may be explained by diffusion or by smearing of higher concentrations downward during sampling.

The cores from Amituk, Sophia, and Hazen Lakes in the Canadian High Arctic islands, as well as Schrader Lake in Alaska, differ from the others. Onset of ΣPCB inputs in these latter cores appears to be in the 1950s, although low concentrations can be seen in older slices. Profiles of ΣDDT in the same lakes also show a later onset of appearance of this contaminant compared to mid-continental lakes at 49°N and 63°N (Muir *et al*. 1995a).

#### 6.7.3.1.2. **Historical profiles and fluxes of PCDD/Fs in lake sediments**

Historical profiles of total PCDD/Fs and OCDD in two cores from Great Slave Lake and in three Finnish lakes are presented in Figure 6·48. Two of three cores from northern Finland, from Lakes L222 and Sierram, show recent declines in PCDD/F deposition following major increases since the 1940s. In the lake Pahtajärvi, a subsurface maximum for PCDD/Fs is discernible in slices dated to the mid-1970s along with an elevated concentration in the surface slice. The preindustrial contribution of PCDD/Fs, presumably due to combustion sources such as forest fires and wood burning, is evident in these Finnish

Table 6·18. Mean concentrations of OCs (µg/g lw) and 95% confidence limits of the geometric mean (in brackets) in the last year of study (usually 1994 or 1995), in muscle tissue of pike and Arctic char in three lakes in Sweden, and the mean annual rate of change in levels.

| Organochlorine | Location and species | | | | | |
|---|---|---|---|---|---|---|
| | Lake Storvindeln (northern Sweden) Pike | | Lake Bolmen (southern Sweden) Pike | | Lake Abiskojaure (northern Sweden) Arctic char | |
| | Mean and 95% CI | Annual change, % | Mean and 95% CI | Annual change, % | Mean and 95% CI | Annual change, % |
| ΣDDT | 0.068 (0.053-0.088) | −12 | 0.4 (0.289-0.547) | −9.9 | 0.025 (0.016-0.038) | −10 |
| ΣPCB | 0.87 (0.68 -1.11) | −4.2 | 1.61 (1.32-1.97) | −5.3 | 0.12 (0.108-0.221) | −6.5 |
| α-HCH | 0.005 (0.004-0.007) | −13 | 0.009 (0.005-0.015) | −9.4 | 0.005 (0.004-0.007) | −17 |
| γ-HCH | 0.005 (0.001-0.022) | −14 | 0.015 (0.011-0.020) | −13 | 0.003 (0.001-0.007) | −17 |
| HCB | 0.013 (0.008-0.021) | 0.2 ns[a] | 0.012 (0.006-0.023) | −3.9 ns | 0.019 (0.015-0.025) | −5.3 |

a. 'ns' implies the rate of change was not significantly different from zero.

cores, particularly in Lake L222. Both cores from Great Slave Lake are from the west basin, which is influenced by inputs of sediment from the Slave River, and both show core maxima for total PCDD/Fs and OCDD in the late 1950s. The appearance of elevated levels of PCDD/Fs in these cores coincides with the start-up of two chlorine-bleached kraft pulp mills within the drainage basin, with the use of pentachlorophenol and other PCDD/F-containing pesticides, and with increased industrial activity in the region to the south. Recent fluxes of OCDD in Great Slave Lake range from 6 to 10 ng/m$^2$/y compared with 0.5 to 5.4 ng/m$^2$/y in the lakes from northern Finland. By comparison, PCDD/F fluxes of 230 ng/m$^2$/y in Siskiwit Lake (Isle Royale in Lake Superior; Czuczwa *et al*. 1984) and 375 ng/m$^2$/y in Green Lake in northern New York (Smith *et al*. 1993b) have been reported.

The core profiles of OCDD in Pahtajärvi and Sierram Lakes in Finland, and in Core 23A from Great Slave Lake, are similar to observations for cores from the Laurentian Great Lakes by Czuczwa and Hites (1984, 1986), who found maxima in the 1940s-1960s. Smith *et al*. (1993b) found OCDD was the predominant PCDD/F congener (approximately 60% of ΣPCDD/F) in Green Lake. The historical records for OCDD in these cores differ from that found in a Baltic Sea core which had highest concentrations in the most recent slices (1970s-1980s) (Kjeller and Rappe 1995).

### 6.7.3.2. Temporal trends of OCs in fish in northern Scandinavia

In 1967, after the Baltic environment was found to be contaminated by DDT and PCB, a Swedish program to monitor DDT and PCB pollution trends was initiated in areas with little or no known local discharges spanning from northern to southern Sweden (Olsson and Reutergårdh 1986, Olsson *et al*. 1997). This program has yielded a comprehensive series of temporal trends for various species at different locations. Compounds analyzed included DDTs, PCB (total as well as congener specific), HCB, and HCHs. Samples consisted of Arctic char, pike, herring, cod, and also guillemot eggs which were collected annually and were selected for consistency in sex, age, size, and sampling season. Each annual sample at a site was represented by 10-25 specimens, thus, within-year variation could be estimated. Overall, the study has so far comprised 3685 samples analyzed individually.

Lake Storvindeln, a forest lake near the Swedish alps, occupying an area of 55 km$^2$, was one of two Arctic sampling sites. Muscle samples from twenty pike, *Esox lucius*, collected in the spring, have been analyzed each year since 1967, giving an unbroken time trend study based on annual samples covering a period when DDTs and PCBs, as well as other OCs were in common use. The other Arctic site was Lake Abiskojaure, 200 km north of the Arctic Circle, where muscle samples of Arctic char (*Salvelinus alpinus*) have been collected since 1980. For comparison with the Arctic regions, pike have also been sampled in Lake Bolmen, a lake of 184 km$^2$ in southwest Sweden that receives minor amounts of industrial and agricultural pollution.

Declines in levels of ΣDDT (DDT + DDD + DDE), ΣPCB (CBs 10, 138, and 153), α- and β-HCH, lindane (γ-HCH), and HCB, were noted in most species and locations, including the Arctic locations (Olsson and Reutergårdh 1986, Olsson *et*

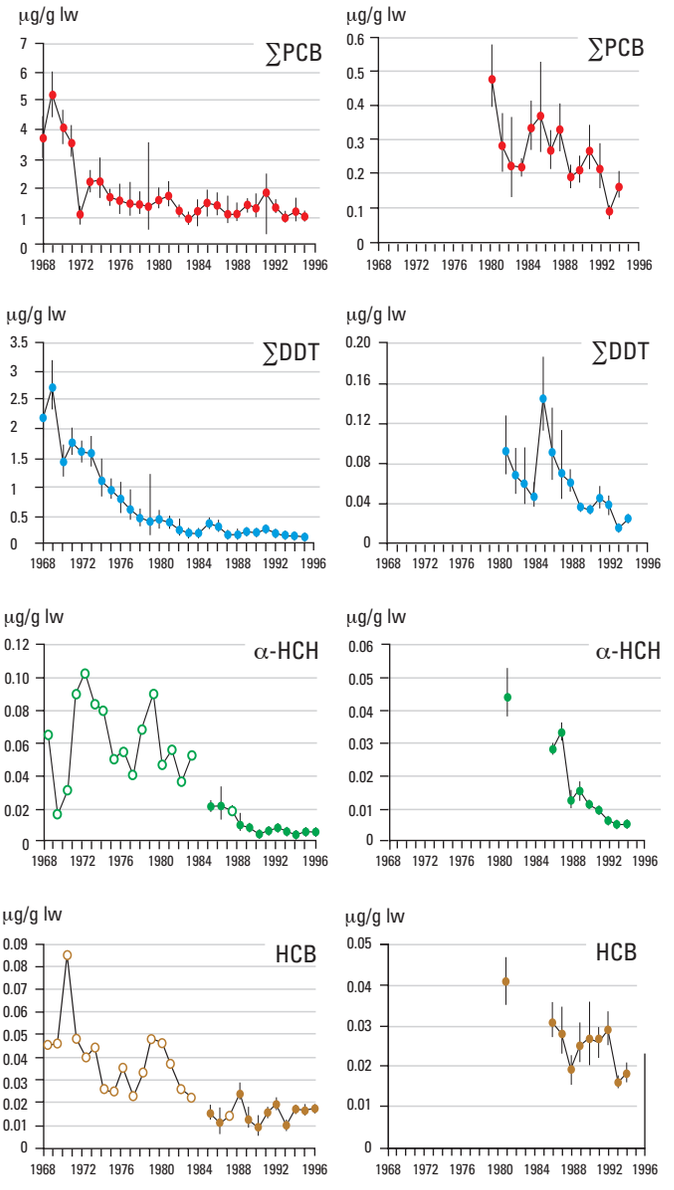



Figure 6·49. Temporal trends in levels of PCB, DDT, α-HCH, and HCB in muscle from Lake Storvindeln pike and Lake Abiskojaure char in Sweden. Mean levels (µg/g lw) and 95% confidence intervals of the mean are represented by dark points, while hollow points represent one pooled sample.

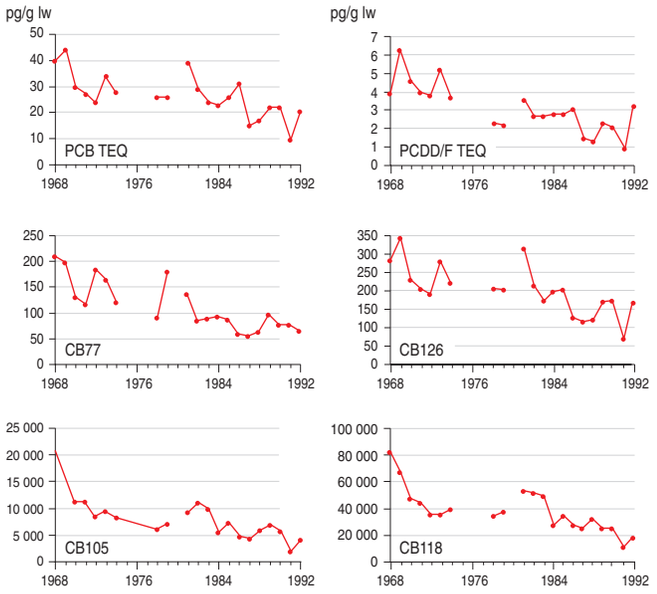



Figure 6·50. Levels of PCB congeners and PCDD/Fs (pg/g lw) in Lake Storvindeln pike muscle between 1968 and 1992.

*al*. 1997). Rates of decline of several OCs in Lakes Storvindeln, Bolmen, and Abiskojaure are given in Table 6·18, and the decline of the same compounds in Lakes Storvindeln and Abiskojaure in Figure 6·49. The char data can be expected to provide a more accurate temporal trend since the species has a higher concentration of fat, making the analysis more reliable (M. Olsson pers. comm.). HCH isomers showed significant declines in fish muscle over the 24-year period. ΣPCB and especially ΣDDT also show significant declines over the period. Much of these declines occurred in the period 1970-1980. More recent changes in ΣPCB and ΣDDT concentrations have been small (Figure 6·49). The ongoing decline in recent times is seen in the Arctic char time series starting at the beginning of the 1980s. Levels of ΣPCBs in pike in boreal Finnish lakes decreased similarly from 5-15 µg/g lw in the early 1970s to 0.5-3 µg/g lw in recent years (Korhonen *et al*. 1997).

Trends in PCDD/F TEQs, nPCBs 77 and 126, mono-*ortho* CB congeners 105 and 118, and PCB TEQs in muscle from Lake Storvindeln pike are shown in Figure 6·50 (M. Olsson and C. de Wit unpubl. data). Figure 6·50 demonstrates the correlation between the various congeners and compounds and also the large between-year variation. Overall, the declines of PCDD/F and PCB TEQs have paralleled the decline in ΣPCBs. PCB TEQs declined about four-fold from 0.05-0.06 ng/g in 1968-1970 to 0.01-0.02 ng/g in 1990-1992, while the decline in PCDD/F TEQ (0.05-0.08 to 0.015-0.03 ng/g) was smaller and more variable over the 24-year period.

With the volume of data in the Swedish study, it was possible to make generalizations about rates of decline in northern Europe. Specifically, the parameters of the log (concentration) versus time regressions for various species and OCs were examined and compared. Slopes represented the rates of decline, and the onsets of the decline were estimated visually. Olsson *et al*. (1997) found that the annual rate and the onset of the decline (1971-1972) in concentrations of DDT compounds did not differ between the Arctic and remote areas of northern Sweden or between the southern parts of the Baltic Sea and lakes in the southern part of the Swedish mainland. HCHs showed similar trends and levels. However, the concentrations of both DDT and HCHs are considerably lower in the Arctic areas.

The onset of the declines coincides with measures taken to reduce the environmental input of these OCs. Also, the sudden decline in the Russian economy at the end of the 1980s and the confirmed reduction of pesticide production as well as imports of pesticides in Russia (Libert 1995) can easily be followed not only in the Baltic Proper, but also in the remote Arctic regions of Sweden. The onset of the decline in concentrations of ΣDDT and ΣHCH is seen simultaneously in both predatory fish and birds, and in eggs of these birds. Besides a decline in total concentrations of ΣHCH, α-HCH has also declined at a similar rate both in the Arctic and more southerly areas of Sweden.

A sudden use of DDT in former East Germany in the summer of 1983 and in 1984 can be detected in the Swedish monitoring program in most studied locations, both with respect to the ratio DDT/ΣDDT as well as in total concentrations of ΣDDT (Bignert *et al*. 1990).

PCB levels also declined at all locations examined. The onset of the decline, approximately 1975-1978, occurred simultaneously in the Swedish Arctic and in the areas close to urbanized regions. The rates of decline of most OCs were generally similar in all locations. However, the overall decline for PCBs was lower than for the OC pesticides studied. This is believed to indicate continued release of PCBs to the environment.

### 6.7.3.3. Temporal trends of OCs in freshwater fish in the North American Arctic

Information on temporal trends of POPs in freshwater fish in the Canadian Arctic is very limited. Early monitoring by Reinke *et al*. (1972) on OC pesticides in Arctic char and lake trout is difficult to compare with present day levels because of the use of packed column gas chromatographic analysis and the associated 10- to 100-fold higher detection limits. Results for OCs in burbot liver from Fort Good Hope on the Mackenzie River are available for an eight-year period, from 1986 to 1994. Mean (lipid normalized log transformed) concentrations of major OCs in burbot liver are compared in Figure 6·51. Toxaphene and chlordane levels have undergone significant declines over the eight-year interval. No significant declines were observed for ΣDDT, ΣPCBs, chlorobenzenes, or dieldrin.

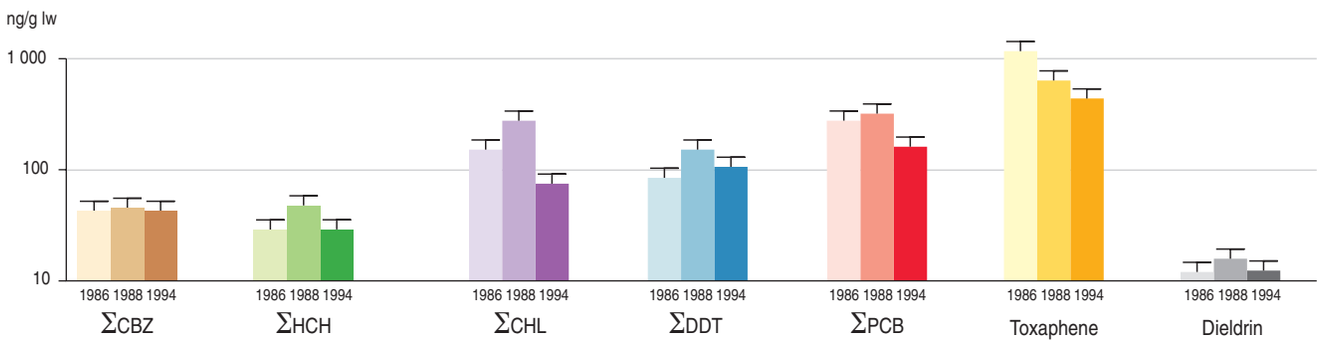



Figure 6·51. Concentrations, as geometric means, of major organochlorine groups in burbot liver at Fort Good Hope, NWT, Canada (1986-94). Vertical lines denote standard errors.

No other comparable data sets of temporal trends of OCs exist for freshwater or anadromous fish in Alaska, Arctic Canada, or Greenland. This is in contrast to the very detailed temporal trend information available for fish in the Great Lakes (Borgmann and Whittle 1991, De Vault *et al.* 1995).

## 6.7.4. Marine environment

### 6.7.4.1. Sediments

As far as we are aware, there have, as yet, been no dated marine sediment cores from the Arctic Ocean analyzed for OC contaminants. Cores have been collected, however, in the Beaufort/Chukchi Seas, Hudson Bay, and Barents Sea, and will undoubtedly be analyzed in the near future. Iwata *et al.* (1994b) analyzed OCs in a sediment core from Bristol Bay in the eastern Bering Sea. The OC profile in this core showed no significant trends with depth, possibly because the sediment surface had been disturbed. The same authors also analyzed an undisturbed core from the northern Gulf of Alaska (55°44'N, 156°14'W) which showed maximum HCB, ΣCHL, and ΣDDT fluxes in slices dated to the period 1970-1980. Maximum ΣPCB and HCH fluxes were observed in surface slices (1980-1990).

### 6.7.4.2. Temporal trends of OCs in seabirds

Several studies described below, which examined temporal trends in contaminant levels in seabird eggs in the Arctic, show downward trends in levels of OC contaminants in recent years. The declines are generally attributed to the reduction in use of contaminants at both southern and northern latitudes.

Barrett *et al.* (1996) reported significant declines since 1983 in levels of PCBs, *p,p'*-DDE, HCB, β-HCH, γ-HCH, and oxychlordane in six of the seabird species breeding in northern Norway. The reported OC levels in seabirds were at the low end of the scale of recently published data from Britain, Canada, and the Mediterranean Sea (Noble and Burns 1990, Walker 1990). An earlier study had not revealed changes in the ten-year time period between 1970 and 1980 (Barrett *et al.* 1985). The highest concentrations were found in the gulls, *Larus* spp., but the levels were considerably lower than earlier data from the south and west coast of Norway (Bergstrøm and Norheim 1986, Moksnes and Norheim 1986). The one exception was herring gulls in the south Troms/north Nordland region in which concentrations of PCB, *p,p'*-DDE, β-HCH, oxychlordane, and HCB increased. However, in general, a large decrease was observed in levels in herring gulls, with declines of 60, 85, and 78% for HCB, DDE, and PCB, respectively, compared to levels found in 1979-1981 by Moksnes and Norheim (1986). The decline corresponded to similar declines documented in marine fish in a Norwegian fjord (Skaare *et al.* 1985).

Similar declines were seen in the Barents Sea (Savinova *et al.* 1995b). Recent ΣDDT and ΣPCB levels in kittiwake were in general lower than levels reported from the Canadian Arctic (Nettleship and Peakall 1987), Bear Island (Bjørnøya) (Bourne 1976) and the coast of Murmansk (Savinova 1991) in the 1970s. Present levels of OCs in glaucous gulls from the Barents Sea are low when compared to corresponding levels from the same area published earlier (Bourne and Bogan 1972, Norheim and Kjos-Hansen 1984). Bourne and Bogan (1972), Norheim and Kjos-Hansen (1984), and Gabrielsen *et al.* (1995) collected birds in early spring, while Daelemans *et al.* (1992) and the present Barents Sea study report results collected during the breeding season.

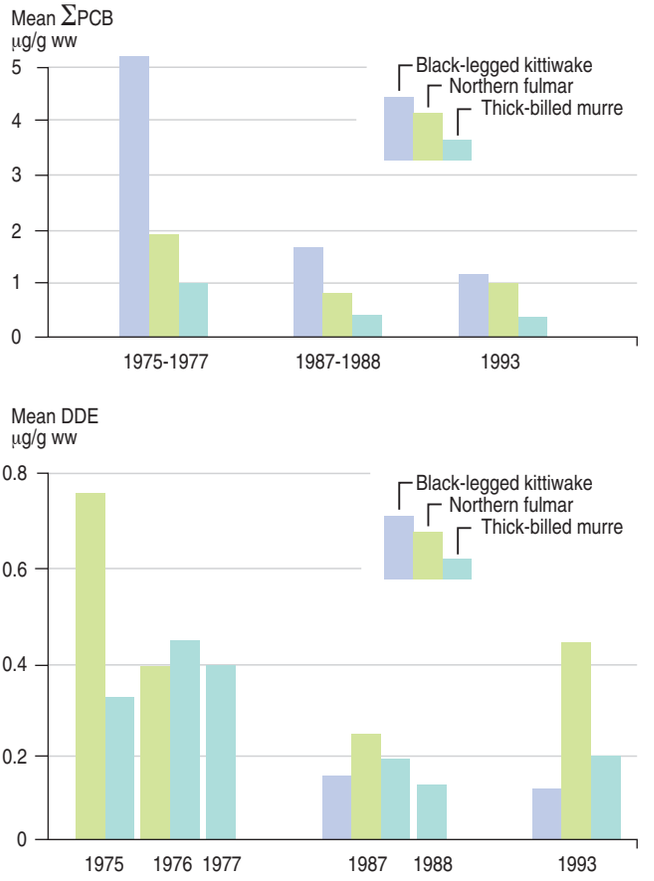



Figure 6·52. Temporal trends in PCB and DDE levels in Arctic seabird eggs from Prince Leopold Island, Canada (1975-1993). Number of eggs analyzed for respective bars are: PCBs 10, 10, 32; 3, 6, 20; and 15, 15, 15 and DDE 10, 12, 10, 11, 10; 3, 6, 10, 10; and 15, 15, 15.

OC residues have been monitored in eggs and livers of several species of seabirds at the Canadian High Arctic colony of Prince Leopold Island since the mid-1970s (Braune 1994a, 1994b). Levels of ΣPCB and ΣDDT in the eggs of black-legged kittiwake, northern fulmar, and thick-billed murre declined during the period between 1975 and 1993 (Figure 6·52). This trend is confirmed by several other limited Canadian data sets. Residue levels measured in livers of adult northern fulmars in the mid-1970s (Nettleship and Peakall 1987) and again in 1987 and 1993 also indicate a decline in residue levels. PCBs have also declined in eastern Canadian Arctic breeding fulmars, kittiwakes, and thick-billed murre (Brünnich's guillemot) since 1987 (Elliott *et al.* 1992). There was, however, a 50% increase in PCB levels and a two-fold increase in chlordane-related compounds in ivory gull eggs. The ivory gull is a resident Arctic species and, therefore, the increase probably reflects the effects of long-range transport to the Arctic, even though levels appear to be going down in the North Atlantic where most other species overwinter.

Studies from more temperate areas of North America and Eurasia showed similar downward trends. Included among these are declines in levels of DDE and PCB in guillemot eggs since 1985 (Olsson and Reutergårdh 1986, Olsson *et al.* 1997), DDT in storm petrels, rhinocerous auklets, great blue herons, and pelagic cormorants in British Columbia, Canada since the 1970s (Elliott and Noble 1993), and PCB in puffins in the Bay of Fundy, Nova Scotia, Canada since the 1970s and 1980s (Elliott *et al.* 1992). However, levels in eggs of double-crested cormorants, herring gulls, and razorbills from the St. Lawrence estuary in Canada did not change (Noble and Burns 1990).

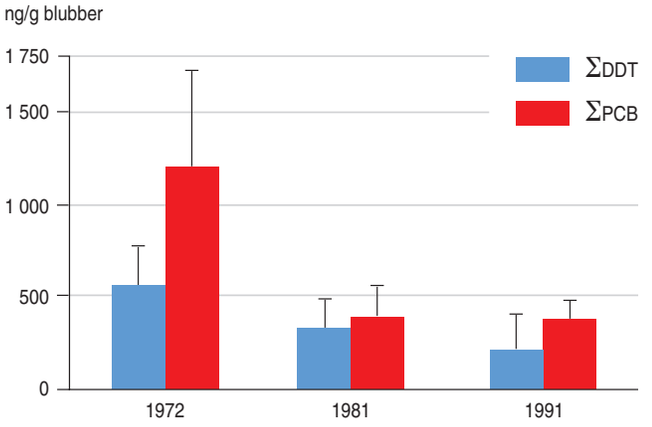



Figure 6·53. Temporal trends of ΣPCB and ΣDDT in blubber of female ringed seals from Holman Island in the western Canadian Arctic (Addison 1995b). Vertical lines represent one SD.

### 6.7.4.3. Temporal trends of OCs in pinnipeds and cetaceans

#### 6.7.4.3.1. Pinnipeds

There are relatively few long-term (multi-decade) studies of OCs in Arctic marine mammals. The longest running study of temporal trends in marine mammals is at Holman (NWT) in the western Canadian Arctic. A population of ringed seals at Holman has been studied since the late 1960s to provide information about Arctic seal population dynamics, and since the early 1970s to measure OC concentrations in the seal blubber. Addison *et al.* (1986) showed that between 1972 and 1981, PCB concentrations in the Holman ringed seals fell by about 40%. ΣDDT concentrations in this population did not fall significantly over that interval. Furthermore, *p,p'*-DDE represented the same proportion of the DDT-group as it had in the early 1970s, implying continued inputs of *p,p'*-DDT to the Canadian Arctic.

A further analysis of the Holman ringed seal population was carried out in 1989-91, and compared with the 1972 and 1981 analyses. The results, which are summarized in Figure 6·53, are only for females, to avoid confounding effects of age and sex on trends in levels. Addison (1995a, 1995b) and Addison and Smith (1996) concluded that between 1972 and 1991, PCB concentrations in blubber declined about five-fold or to about 20% of their initial value

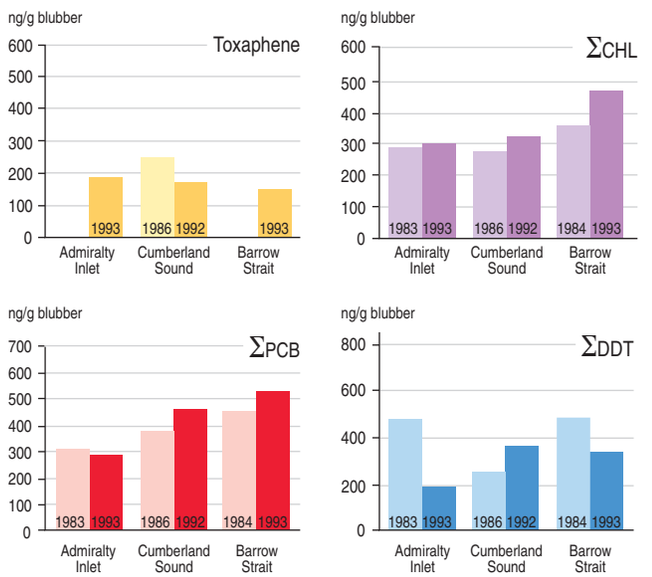



Figure 6·54. Trends in concentration of major organochlorines in blubber of female ringed seals over six to ten years at three sites in the eastern Canadian Arctic (N = 10-14 animals for each year/site).

with the major decline occurring in the 1970s. ΣDDT concentrations also declined slightly over the same interval. Most of the decline occurred during the 1980s rather than the 1970s.

Time trends in OC concentrations are also available for other sites in the eastern Canadian Arctic and at Svalbard, but only for the period 1984 to 1994. Daelmans *et al.* (1993) compared PCB levels in ringed seal blubber at Svalbard (1990) with results from earlier studies of samples collected in 1986 (Oehme *et al.* 1988) and 1984 (Carlberg and Bøler 1985). PCB levels in 1990 were half of those found in the 1986 study and *p,p'*-DDE levels were similar for both years. However, the 1990 results were higher than those found in the 1984 study. No data were available on the age or sex of these latter animals, making comparison difficult.

In the eastern Canadian Arctic, trends in OC levels over a 6-10 year period (1983-1993) have recently been examined in female ringed seals from three locations (Figure 6·54). Concentrations of ΣDDT, ΣPCB, ΣCHL, and toxaphene have not declined significantly in female ringed seal blubber from Cumberland Sound (E. Baffin Island), Barrow Strait in Lancaster Sound, or Admiralty Inlet (N. Baffin Island). The results are reasonably consistent with those for PCBs and ΣDDT at Holman Island (Addison 1995a, 1995b, Figure 6·53) because most of the decline in PCB concentrations there appears to have occurred during the 1970s.

Results for walrus from Thule (W. Greenland) show similar trends to those for seal blubber in the eastern Canadian Arctic. ΣDDT levels in walrus blubber are very similar in samples from females from 1975/76 (50 ± 50 ng/g blubber, n = 20; Born *et al.* 1981) and those from 1988 (70 ± 39 ng/g blubber n = 10; Muir and Born 1996). PCB levels are also similar (1975/76, 180 ng/g blubber versus 1988, 244 ng/g), however, the PCB results from earlier packed column GC work may not be directly comparable to more recent analyses of individual congeners.

#### 6.7.4.3.2. Cetaceans

Male narwhal and beluga from the Canadian Arctic also show little change in concentrations of major OCs over a 10-year period (Figure 6·55). Males were selected for tempo-

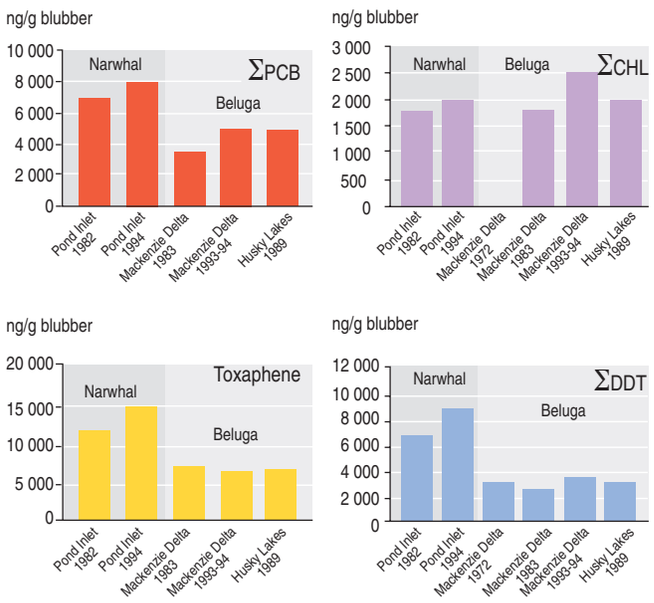



Figure 6·55. Trends in ΣPCB, ΣDDT, ΣCHL, and toxaphene over a ten year period (20 years for DDT in the Mackenzie Delta) for male narwhal and beluga blubber samples from the Canadian Arctic (N = 8-26). Data for beluga from 1972 are from Addison and Brodie (1973) and data for narwhal are from Muir (1994, 1996).

ral trend comparison because (unlike seals) they show little trend in PCB concentrations with age (Stern *et al*. 1994). Male narwhal from the Lancaster Sound region (Pond Inlet) show no significant decline in concentrations of ΣDDT, ΣCHL, or ΣPCB over a 12-year period. Toxaphene levels are higher in 1994 than in 1982 in comparable samples; this may be due to changes in the method of quantitation and the results are being reevaluated. Three sets of samples of male beluga (n = 8-26) from the Mackenzie Delta region have been analyzed since 1983 using identical methodology. Results for DDT levels from this stock were also available from 1972 (Addison and Brodie 1973). No significant decline was observed in any of the four major OC groups over the 10-year period, and over 20 years in the case of DDT.

#### 6.7.4.4. Temporal trends of OCs in polar bear

Average ΣPCB levels in bears from the Canadian Arctic in the 1989-1993 circumpolar study (section 6.6.4.8) were similar to those in 1982/84, while average ΣCHL and DDE levels were 35-44% lower, and dieldrin levels were 90% lower (Norstrom *et al*. 1988). However, the significance of the temporal trends during the 1980s, derived using these data, is not conclusive because of problems of comparability. The 1982/84 samples were not controlled for age or sex, and were composites (equivalent to an arithmetic mean). Archived samples of polar bear fat collected from R6, R7, R9, and R14 for 1984, and 1990, and R11 for 1969, 1984 and 1990, are being reanalyzed to study temporal trends. Results from this study are not yet available.

#### 6.7.4.5. Temporal trends of OCs in Arctic fox (Svalbard)

Wang-Andersen *et al*. (1993) examined concentrations of PCBs in Svalbard Arctic fox fat and liver samples collected in 1983-84, and compared these with earlier samples collected in 1973-1974. ΣPCB, based on seven selected congeners, did not differ significantly between sampling periods.

### 6.7.5. Summary and conclusions – temporal trends

The results of a number of temporal trend studies of Arctic biota indicate that ΣPCB and ΣDDT levels in the Arctic have declined over the past 20-25 years since the first controls on DDT and open use of PCBs began. Evidence from dated sediment and snow cores indicates leveling off, if not actual declines, in inputs to the Arctic. With the exception of studies in Swedish biota, the results in support of this conclusion are limited for several reasons. Temporal trend monitoring in the Canadian, Greenlandic, Norwegian, and Finnish Arctic has been limited to small and infrequently collected samples (2-4 times over 25 years, compared to yearly for several species in the Swedish Arctic). Combined with high intrasite variability in OC levels, statistically significant trends are difficult to discern from a small number of sampling times. Furthermore, there have been changes in analytical methodology, which made comparison with older results problematic in some cases (e.g., ringed seals from Holman Island in the western Canadian Arctic).

Less is known about the temporal trends of many other persistent OCs, including HCH, HCB, ΣCHL, toxaphene, dieldrin, and PCDD/Fs.

There is convincing evidence for declines of some persistent OCs in the atmospheric and terrestrial Arctic environments. A nine-fold decrease in concentrations of ΣHCH in Arctic air has been observed, based on measurements in the Bering/Chukchi Seas and at several locations in the Canadian Arctic Archipelago. Air monitoring from 1984 to 1992 on Svalbard also showed a decline of up to two-fold in α-HCH. However, γ-HCH levels were higher in 1992 at all three Norwegian air-monitoring sites than in 1984, reflecting the increased use of lindane (γ-HCH) and declining use of α-HCH as use of technical HCH products ceased in Europe during the 1980s. Long-term monitoring of OCs in air, as has been done at Ny-Ålesund on Svalbard and on ocean cruises in the Bering/Chukchi Seas by several groups, has the advantage of detecting changes in concentrations of inputs without the confounding effects of selective transformation of some OCs in the food web or selective removal during sedimentation and burial in profundal sediments.

Evidence for the decline of airborne PCBs in the European Arctic comes from a study of mosses in northern Norway. This study, although flawed because of the use of air-drying which may have introduced PCB contamination, showed a consistent three-fold decline of ΣPCB concentrations in mosses from both coastal and inland areas over the period 1977-1990. Over the same time period, ΣPCB in southern Norway declined about four-fold. As moss depends entirely on the atmosphere for delivery of nutrients and lacks both cuticle and internal transport mechanisms, this reduction is indicative of a decrease in atmospheric PCB concentrations. The decline of PCBs in Arctic mosses is consistent with observations in the nearby Swedish Arctic of declining ΣPCB levels (about a three- to four-fold decrease over 26 years) in pike and char muscle.

Other evidence of declines or leveling off of OC inputs comes from the analysis of dated lake sediment cores. Most of the subarctic (60-70°N) lakes show the appearance of PCBs and DDT in the 1940s, in reasonable agreement with the known source functions for these compounds. Sediment profiles in subarctic lakes in Canada (Belot, Great Slave, Kusawa, Hawk, Far) and Finland (Pahtajärvi, Sierram) show subsurface maxima suggesting maximum inputs in the 1970s and early 1980s, corresponding to maximum PCB use in industrialized areas.

The cores from the Canadian High Arctic island lakes Amituk, Sophia, and Hazen, as well as Schrader Lake in Alaska, differ from the others. Onset of ΣPCB inputs in these latter cores appears to be in the 1950s, although low concentrations can be seen in older slices.

This later appearance of PCBs is in agreement with the global fractionation model, which predicts that persistent, semi-volatile organics will be more prominent in polar regions and temporal trends in deposition will be delayed and prolonged relative to temperate regions (Wania and Mackay 1993). Unfortunately, there are no analyses of sediment cores from the Eurasian High Arctic (e.g., Spitsbergen and Bear Island) which could confirm whether observations in the Canadian High Arctic are a generalized circumpolar phenomenon.

Recent declines in PCDD/F deposition following major increases since the 1940s are discernible in two of three cores from northern Finland. These cores, which are from relatively undisturbed lakes receiving atmospheric inputs of contaminants, provide good evidence of historical inputs of PCDD/Fs in Arctic Scandinavia. They show a major preindustrial contribution of PCDD/Fs, presumably due to combustion sources such as forest fires and wood burning, as well as sources coinciding with chlorine-based chemical production starting in the 1930s. Both cores from Great Slave Lake in Canada provide evidence for industrial inputs to the west basin of the lake because the appearance of elevated levels of PCDD/Fs in these cores coincides with the start up of two chlorine-bleached kraft pulp mills within the drain-

age basin, and with the use of pentachlorophenol and other PCDD/F-containing pesticides. Kjeller and Rappe (1995) also found increased PCDD/F concentrations in the period 1970-1985 and a change in congener pattern from a core in the Baltic Sea which they attributed to increased use or emissions of chlorophenols. The profiles for PCDD/Fs in the Finnish and Canadian lakes are also similar to observations by Czuczwa and Hites (1984, 1986) for cores from the Laurentian Great Lakes with maxima in the 1940s-1960s.

The time trend study of Swedish reindeer from Abisko, based on annual samples, showed a significant change in α-HCH over the time period 1983-1994. A ten-fold decline was found which is quite close to the decline observed in air over the Bering/Chukchi Seas. Other OCs studied, such as PCB, DDT, and β-HCH, did not change significantly in reindeer, but the between-year variation was substantial, implying that the time period was probably too short to allow a proper evaluation. Pike and char from Swedish Arctic lakes show α-HCH concentration decreases similar to those found in reindeer.

Declining concentrations of ΣPCB and ΣDDT and increases in chlordane-related compounds in eggs of peregrine falcons and other birds of prey have been observed in both the European and North American Arctic, consistent with changes in the use of these OCs. Mean *p,p'*-DDE levels in peregrine falcon eggs declined from 7.6 μg/g in the 1980s to 4.5 μg/g in the 1990s. PCB (as Aroclor 1254 : 1260, 1 : 1) showed no significant change (means of 8.7 μg/g ww in 1981-1985 and 8.31 μg/g ww in 1991-94). Dieldrin, oxychlordane, and HCB also showed no change in concentration over the 10-13 year period. Levels of heptachlor epoxide actually increased over the period 1966 to 1987, but may have declined significantly during the late 1980s.

The best evidence for declining OC concentrations in Arctic birds of prey comes from the study of Lindberg *et al.* (1985) and Lindberg (1995b), who compared eggs collected during 1972-1981 and in 1991-1994, and found declines in concentrations of about two-fold for PCBs and five-fold for *p,p'*-DDE.

A problem with utilizing contaminant levels in eggs of migratory birds of prey to assess temporal trends in the Arctic is that the levels in eggs may also reflect exposure at wintering areas. Food for carnivorous birds may also largely consist of migratory species in some cases. Nevertheless, the egg monitoring programs enable an assessment of regional or hemispheric changes in inputs of bioaccumulating contaminants to terrestrial and aquatic environments.

Monitoring of fish in Lake Storvindeln and Abiskojaure in northern Sweden has provided some of the strongest evidence for declining inputs of persistent OCs to the aquatic environment in northern Scandinavia. Declines in levels of ΣDDT, ΣPCB, and HCB were noted in these Arctic locations. For DDT and PCB, a sudden decline occurred immediately after European measures to reduce the discharges of DDT in the beginning of the 1970s and PCB in the middle of the 1970s (Olsson and Reutergårdh 1986, Bignert *et al.* 1995, Olsson *et al.* 1997). After the initial decline, the annual change in concentration continued and there is still an annual decline of DDT and PCB of 3-8% a year (Olsson *et al.* 1997). While these recent changes in ΣPCB and ΣDDT have been small, there is no indication that concentrations are leveling off. Levels of ΣPCBs in pike in subarctic Finnish lakes also decreased 5- to 10-fold from the early 1970s to the early 1990s. Toxaphene, ΣHCH, and chlordane levels underwent significant declines in burbot liver from the lower Mackenzie River in Canada over the period 1986-94. No information is available on long-term trends of OCs in fish from Russia, Alaska, or Greenland.

The declining concentrations of persistent OCs in pike and char muscle from Scandinavia parallels observations in lake trout in the Great Lakes (DeVault *et al.* 1995). The decline in concentrations of ΣDDT and PCBs in Lake Ontario lake trout was greater during the 1970s immediately following bans on use of DDT and open use of PCBs, than during the 1980s (Borgmann and Whittle 1991).

The declines of PCDD/F and PCB TEQs observed in fish in the Swedish Arctic have been paralleled by declines in ΣPCBs over the 24-year period. These declines are not consistent with other observations in fish. For example, concentrations of 2,3,7,8-TCDD in Lake Ontario lake trout did not decline significantly over the period 1980-1992 (DeVault *et al.* 1995). The many sources of PCDD/Fs to the environment (e.g., chlorine bleached kraft mills, chlorophenols, and combustion) may mean that declines in TCDD TEQs may be very site specific in comparison to trends for PCBs and semi-volatile OC pesticides.

The Swedish time-trend studies provide a proven design on which future AMAP monitoring programs for temporal trends could be developed. The program is sufficiently long-term and detailed (annual sampling) that it has been able to demonstrate changes in inputs in source regions. For example, the sudden decline in the Russian economy at the end of the 1980s and the confirmed reduction of pesticide production, as well as imports of pesticides in Russia (Libert 1995) can be easily followed not only in the Baltic Sea, but also in the remote Arctic regions of Sweden. The onset of the decline in concentrations of ΣDDT and ΣHCH is seen simultaneously in both predatory fish and birds, and in eggs of these birds. A sudden use of DDT in former East Germany in the summer of 1983 and in 1984 was detected in the Swedish monitoring program at most study locations, both with respect to the ratio DDT/ΣDDT as well as in total concentrations of ΣDDT (Bignert *et al.* 1990).

Results from monitoring of marine biota provide good evidence for declining concentrations of major OCs in both the European and North American Arctic. Significant declines were found over the period 1983-1993 in levels of PCBs, *p,p'*-DDE, HCB, β-HCH, γ-HCH, and oxychlordane in six of the seabird species breeding in northern Norway. A large decrease was also observed in levels in herring gulls, previously examined in 1979-1981 by Moksnes and Norheim (1986), where levels of HCB, DDE, and PCB dropped by 60, 85, and 78%, respectively. The decline corresponded to similar declines documented in marine fish in a Norwegian fjord (Skaare *et al.* 1985). Present levels of OCs in glaucous gulls from the Barents Sea are also low when compared to corresponding levels from the same areas published earlier (Bourne and Bogan 1972).

Levels of PCBs and DDTs in the eggs of black-legged kittiwake, northern fulmar, and thick-billed murre from the High Arctic colony on Prince Leopold Island, Canada have also declined during the period between 1975 and 1993. Most of the decline was observed in the 1970s and early 1980s. These are migratory species and, thus, declines may reflect an overall reduction in the OC levels of the North Atlantic where many of these birds overwinter. In one of the few examples of increasing concentrations, there was a 50% increase in PCB levels and a two-fold increase in chlordane-related compounds in ivory gull eggs.

In general, OC levels in Canadian Arctic seals and whales do not show the same steep decline in concentrations observed in seabirds from eastern Canada (Addison and Smith 1974, 1996, Elliot *et al.* 1988). There are relatively few long-

term (multi-decade) studies of OCs in Arctic marine mammals. The longest running study of temporal trends in marine mammals is at Holman (NWT) in the western Canadian Arctic, where Addison (1995a, 1995b) concluded that, between 1972 and 1991, PCB concentrations in blubber declined about five-fold, while ΣDDT declined about three-fold, with most of the decline occurring during the 1980s. DDT results for beluga from the southern Beaufort Sea show no significant decline over the same period.

In the eastern Canadian Arctic and Greenland, temporal trends in marine mammals can only be examined over a 10-12 year period. No significant declines have been observed in concentrations of ΣDDT, ΣPCB, ΣCHL, and toxaphene in female ringed seals at three locations or in male narwhal blubber from Lancaster Sound from the mid-1980s to early 1990s. Results for walrus from Thule (West Greenland) show similar trends to those for seal blubber in the eastern Canadian Arctic. These results are reasonably consistent with those for PCBs and ΣDDT in ringed seals at Holman Island.

There is insufficient information at present to discern temporal trends in marine mammals from Svalbard, northern Norway, and Russia. Although limited OC data are available for marine mammals from the European Arctic during the 1970s and 1980s, there is, in some cases, insufficient information on key covariates, such as the age, sex, or season of collection of the animals to enable correct comparisons. Similarly, there are problems with comparability of samples of polar bear tissues collected in the 1970s and early 1980s in the Canadian Arctic which has made assessment of temporal trends problematic. At present, there are no long-term data on temporal trends in polar bears from Svalbard.

In conclusion, temporal trends for PCBs and DDT in Arctic biota show declining concentrations from maximums observed in the 1970s. Data from sediment cores and archived moss samples also support this observation. However, considering all of the POPs of interest, the information on temporal trends in the Arctic is very limited. Continuous monitoring over the past 30 years of the Baltic Sea and the northern areas of Sweden have shown that between-year variation is large, and individual variation within this, even larger. These observations reinforce the importance of judicious sampling and archiving programs, which would allow continuous monitoring of key populations and retrospective analysis for new contaminants.

## 6.8. Biological effects

Biological effects can be measured at different levels of biological organization, from the molecular to the ecosystem level (Figure 6·56). Biomarkers measurable at a molecular level respond early, but are not readily interpreted ecologically, while measures with established ecological relevance, such as population declines or reduced reproductive rates, respond too late to have diagnostic or preventative value.

Although Arctic biota contain a range of organic and inorganic contaminants, there is relatively little knowledge of the biological effects of these chemicals in Arctic species. Unlike the situation in the Baltic or the Great Lakes, there is currently little evidence to support or refute arguments that chemical contaminants are present in sufficient quantities to have detrimental effects on any species.

Two approaches have generally been taken in identifying possible effects. The first involves extrapolation and comparison. Researchers determine possible effects by comparing levels of OCs in Arctic species of interest to known detrimental levels, this knowledge coming from laboratory tests or from observations on affected animals in the wild. These types of comparisons have inherent weaknesses. Laboratory animals are most often exposed to single OCs or technical products at high doses for short periods of time, and it is difficult to extrapolate the toxic effects seen at high acute doses to possible adverse effects at lower, but chronic exposures. Wild animals are generally exposed to lower concentrations of OCs, but they are exposed to mixtures of POPs and other stressors, and they are exposed over their entire lifetime. Also, species vary in their sensitivities to the effects of OCs, which can make it difficult to know which of the tested species best represents those in the Arctic. For example, several Arctic species have delayed implantation (mink, otter, other mustelids, seals, walrus, polar bears), which may make them more sensitive to the reproductive effects of OCs than tested laboratory animals without delayed implantation (Sandell 1990).

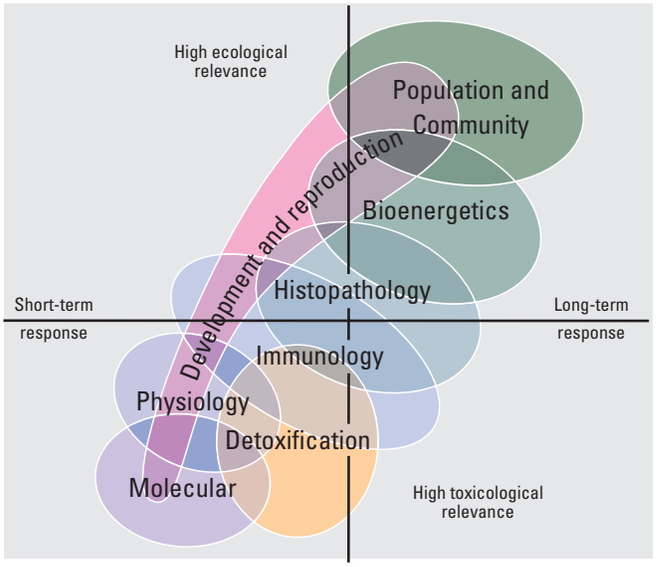



Figure 6·56. The relationship between response levels of biological organization and the toxicological relevance and time scale of responses.

The second approach studies biological effects by examining subtle indicators of biological responses (biomarkers) to contaminants. Examination of the animals for responses known to be associated with the contaminants found in them is perhaps the only way to make a convincing case either for or against the hypothesis that trace contaminants are acting biologically on the animals. Almost any biological change, from molecular to ecological, can serve as a biomarker, however, the term most often refers to changes at sub-cellular levels (McCarthy and Shugart 1990, Huggett *et al.* 1992, Peakall 1992). Biomarkers typically are measures of normal processes which take on abnormal values as a result of exposure to chemicals of interest. Most of the biomarkers studied have established sensitivities (in laboratory animals) to some of the same contaminants measured in Arctic marine mammals and fish, notably several PCB congeners, PCDD/Fs, and polycyclic aromatic hydrocarbons including petroleum oil.

The mixed function oxidase (MFO) cytochrome P450 system, a ubiquitous enzyme system common to mammals, birds, fish, and microorganisms, has probably been one of the most widely used biomarkers to date, with numerous laboratory and field cases of responses established (Payne *et al.* 1987, Rattner *et al.* 1989, Goksøyr and Förlin 1992, Haasch *et al.* 1993, Beyer *et al.* 1996, Hylland *et al.* 1996). The preferred field study design has been the comparison of

an exposed group of individuals with similar groups not exposed to the same source. Another design, less commonly encountered, is the comparison of individuals within a group to search for linkages between biomarker values and exposure as indicated by chemical residues. This is a valuable approach for those contaminants stable enough to remain identifiable as residues.

In the following assessment, results from studies of biomarkers in Arctic biota are presented first if these have been performed. Where possible, levels of OCs in Arctic biota are also compared to no-adverse-effect-levels (NOAEL) or no-effect-levels (NOEL) and lowest-adverse-effect-levels (LOAEL) or lowest-effect-levels (LOEL) known to cause subtle effects in sensitive species. An attempt has also been made to compare OC levels in the diet of selected Arctic biota to known dietary no-adverse-effect-concentrations (NOAEC) or no-effect-concentrations (NOEC) and lowest-adverse-effect-concentrations (LOAEC) or lowest-effect-concentrations (LOEC) or to environmental quality criteria/guideline values for protecting aquatic biota in various countries (Table 6·14). There are considerable limitations in this latter approach as there is a general lack of knowledge of the diet of many Arctic organisms. It is also assumed that predators eat only one type of food, and even where food preferences are known, there may not be analytical data for these particular food items.

## 6.8.1. Terrestrial environment

### 6.8.1.1. Caribou and reindeer

Information on potential or actual biological effects of environmental contaminants in Arctic terrestrial mammals is virtually non-existent (Elkin and Bethke 1995). The levels of OCs (α-HCH, HCB, sum of 43 PCB congeners, chlordanes, ΣDDT, PCDD/Fs, and dieldrin) in caribou (*Rangifer tarandus* spp.) from across the Yukon and Northwest Territories were very low, and in general, were substantially lower than the levels found in Arctic marine mammals. The same is true for reindeer and caribou from Norway, Sweden, and Russia (Annex Table 6·A4). The OC levels found are several orders of magnitude lower than those expected to lead to subtle biological effects.

PCDD/F, nPCB (CB 77, 126, 169), and mono-*ortho* (CB 105, 118) PCB levels are low in reindeer fat, withcombined TEQ values of 0.92-1.9 pg TEQ/g lw in Sweden and 0.3-3.29 pg TEQ/g lw in Canada (Annex Table 6·A22) (C. de Wit unpubl., Hebert *et al.* 1996). PCDD/F levels in reindeer fat from two sites in Norway vary from 0.7-1.2 pg TEQ/g lw at Stilla to 8.6-14.7 pg TEQ/g lw at Jarfjord (near a smelter) (Schlabach and Skotvold 1996a, 1996b). These levels are below those associated with immunosuppression in harbour seal (Figure 6·57) and reproductive effects in mink.

### 6.8.1.2. Waterfowl

No biological effects studies have been performed on Arctic waterfowl. Levels of ΣDDT and ΣPCB in most Canadian species analyzed are below those expected to lead to reproductive effects if it is assumed that levels in whole body or breast muscle are similar to those in eggs on a wet weight basis (Annex Table 6·A4; Table 6·9). However, there are a few exceptions. For ΣPCB, some molluscivores and piscivores from the eastern Canadian Arctic (Table 6·9) and some individuals of semipalmated plover (*Charadrius semipalmatus*) have levels that exceed most NOEL and LOEL values for reproductive effects in white leghorn

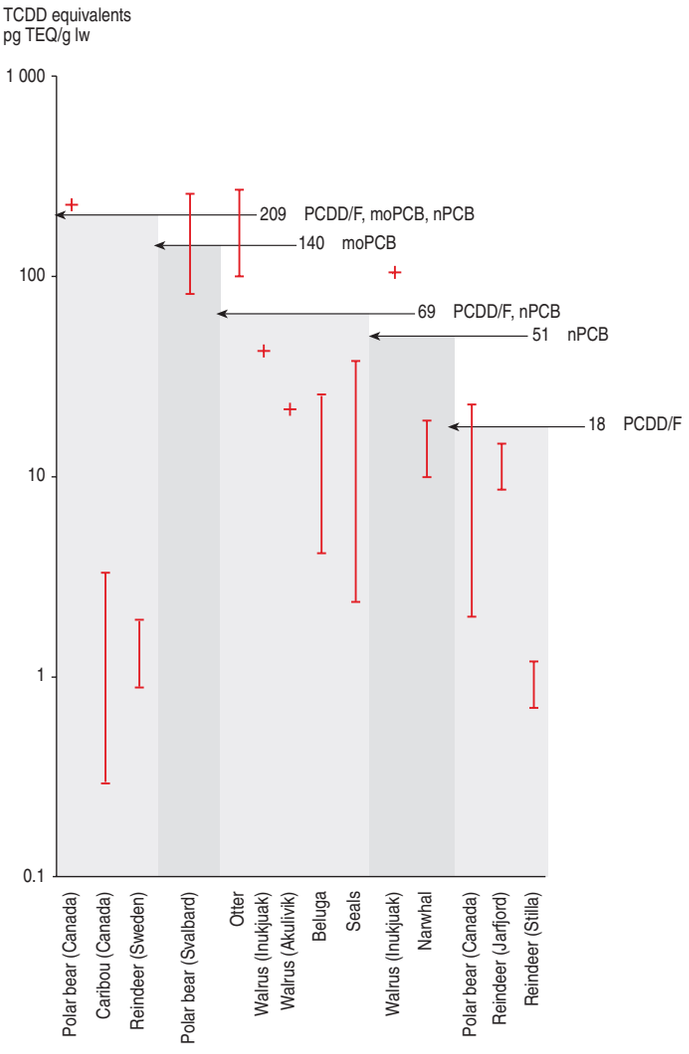



Figure 6·57. Body concentrations of PCDD/Fs and non-*ortho* and mono-*ortho* PCBs as TCDD equivalents (pg/g lw) in Arctic mammals compared to thresholds for immunosuppression in harbour seal for the same combinations of substances analyzed (Ross *et al.* 1995).

chicken (Figure 6·59). Mean ΣPCB levels in pintail (2.11 μg/g ww) also exceed these NOEL and LOEL levels. Oldsquaw have even higher ΣPCB levels of 6.88 μg/g ww (range 2.88-18.9), which overlap or exceed most NOEL and LOEL values for reproductive effects in several other bird species.

Analytical results are available for some waterfowl from Russia, but it is difficult to assess these as the tissue analyzed was liver (Annex Table 6·A16).

### 6.8.1.3. Birds of prey

The decline of populations of birds of prey in both temperate and Arctic regions of the world since the introduction of organochlorine pesticides is well documented (Ratcliffe 1967, Peakall 1976a, Newton 1979, Helander *et al.* 1982, Odsjö and Sondell 1982, Peakall *et al.* 1990, Lindberg 1995a). A large number of species suffered from high concentrations of OCs with associated eggshell thinning and lowered reproductive capacity during the 1950s-1970s. The decline in environmental concentrations of ΣDDT and ΣPCB after being banned in most countries has been followed by the recovery of most populations of predatory birds (Lindberg 1995b, Helander 1996, Odsjö and Sondell 1996). Other than reproductive effects, no other biological endpoints have been studied in birds of prey.

6.8.1.3.1. **Peregrine falcon**

• *Reproduction*

In the 1970s, populations of the North American Arctic breeding peregrine falcon (*Falco peregrinus tundrius*) declined to 35% of their numbers prior to the introduction of DDT (Kiff 1988, Schempf 1989). Perhaps because they were not exposed to high levels of contaminants all year round, populations of the tundrius race did not appear to suffer such extreme effects as the more southerly *Falco peregrinus anatum* populations (Ratcliffe 1967, Peakall 1976b, Newton 1979). Thomas *et al.* (1992) concluded that ΣDDT levels in *F. p. tundrius* had peaked in the mid-1970s, but decreased by the early 1980s.

Court *et al.*'s (1990) studies of the tundrius population from the Keewatin area of the Canadian Arctic, near Rankin Inlet, NWT, between 1981 and 1986, concluded that, while remaining a productive population, significant amounts of organochlorine pesticides and PCB residues were still accumulated. Eggs collected from 1981-1986 were 16% thinner than eggs collected prior to the introduction of DDT. About 10% of breeding attempts failed due to egg breakages. DDE levels in eggs were negatively correlated with eggshell thickness. Eggshell thinning, levels of DDE residues in eggs, and levels of contaminants found in prey items were all considered to be close to the levels considered to result in decreased production in this species (Peakall and Kiff 1988, Court *et al.* 1990, Peakall *et al.* 1990). All but one of 11 prey species measured showed measurable levels of OCs. According to Court *et al.* (1990), reproductive success was still reasonably high because the highly contaminated prey species were not as available on summer breeding grounds, and clean prey was eaten before laying. Court *et al.* (1990) concluded that about 10% of all breeding attempts between 1981 and 1985 failed due to pesticide contamination.

Johnstone (1994), in later studies of the same population, concluded that there has not been any improvement in eggshell quality since the early 1980s for this population of tundra peregrines (Figure 6·58). There was no difference in mean eggshell thickness between the sample collected by Johnstone (1994) and that collected by Court *et al.* (1990) between 1982 and 1986. Peregrine eggshell fragments collected since 1991 are on average 15% thinner (mean 0.306 ± 0.022 mm, n = 54) than those laid in the Nearctic prior to the introduction of pesticides (0.360 mm; Berger *et al.* 1970, Anderson and Hickey 1972, Walker *et al.* 1973). Of the 54 clutches in Johnstone's (1994) sample, 28% showed thinning equal to or greater than levels associated with reproductive failure and population decline in this species, that is, 17% of average pre-DDT shell thickness (Peakall and Kiff 1988).

Johnstone (1994) believes that although DDE levels appear to be decreasing in the population of *F. p. tundrius* nesting around Rankin Inlet, the changes are small. Levels of ΣPCB show little change over the same time period. Also, Johnstone (1994) believes that eggshell thickness and residue levels in peregrine eggs, blood plasma, tissue samples, and prey species are within the range that a portion of reproductive attempts will fail each year due to contamination by organochlorine pesticides and PCBs.

Just as in other countries, peregrine falcon populations in Sweden declined dramatically after the 1940s. It is estimated that about 350 breeding pairs existed in Sweden in the mid-1940s. In 1965, there were 35 pairs, which declined to nine pairs by 1974 (Alsberg *et al.* 1993). The Swedish Arctic population increased to about 25 pairs in the beginning of the 1990s. For Fennoscandia as a whole, the population of peregrine falcon is now increasing and the number of nesting pairs has increased from 65 known pairs in 1975 to 360-410 pairs in 1994 (Lindberg 1995a).

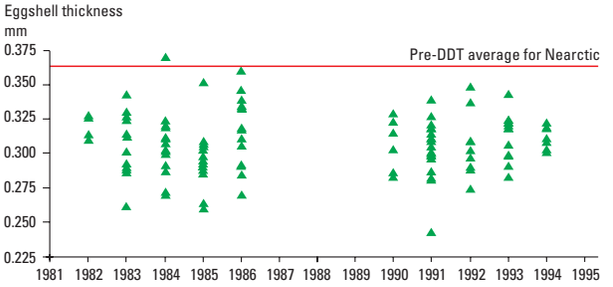



Figure 6·58. Shell thickness (mm) of peregrine falcon eggs representing 109 clutches produced in Rankin Inlet, NWT, Canada from 1981 to 1986 (Court *et al.* 1990) and from 1990 to 1994 (Johnstone 1994).

Studies of peregrine falcons from the Kola Peninsula, Russia, in 1977 and again in 1987-1991 indicate that the population there is also recovering (Henny *et al.* 1994). In 1987-1991, nestling production was 1.94 young per nest and the number of breeding pairs had increased from four to ten. Mean eggshell thickness was found to be 0.310 mm, corresponding to eggshell thinning of 11.4% as compared to Swedish peregrine falcon eggs from 1861-1946, which had pre-DDT egg shell thicknesses of 0.350 mm (Lindberg 1975).

• *Levels and intake assessment*

In several recent review articles (Bosveld and van den Berg 1994, Giesy *et al.* 1994b, Barron *et al.* 1995), the no-effect and low-effect levels for eggs and adults as well as dietary intakes associated with no or low effects have been compiled from the literature for ΣPCB and dioxin-like compounds. For eggs of fish-eating and predatory birds (bald eagle, herring gull, caspian tern, double-crested cormorant, common tern, Forster's tern, great blue heron, black-crowned night heron) the following ranges were found. For reproductive success, the NOEL range for ΣPCB was 1.3-11 μg/g ww in eggs. The LOEL range for various endpoints of reproductive success (hatching success, egg morality, deformities, parental attentiveness) ranged from 3.5-22 μg ΣPCB/g ww in eggs. For adults, ΣPCB concentrations in brain tissue higher than 300 μg/g ww are associated with mortality. Dietary LOEC levels for ΣPCB range from 2 to 50 μg/g ww food for reproductive endpoints in a number of different bird species.

For HCB, results from Vos *et al.* (1972) indicate a NOEL in kestrels (*Falco tinnunculus*) of 40-49 μg/g ww in liver and a dietary NOEC of 100 μg/g ww in food. Lindane concentrations of 100-200 μg/g ww in eggs of quail and chickens are associated with decreased egg production (Whitehead *et al.* 1972a, 1972b, 1974).

It has been concluded that peregrine falcon eggs with DDE residues of 15-20 μg/g ww would experience reproductive failure (Peakall *et al.* 1975). The mean DDE concentrations in Canadian peregrine eggs range from 4.5 to 13 μg/g ww (Annex Table 6·A5), lower than this critical threshold for reproductive failure. However, two clutches (10%) in the 1991 sample contained eggs with residue levels exceeding threshold levels, indicating that the potential for reproductive failure still exists.

Mean ΣPCB levels in Canadian peregrine eggs (both races) are 5.6 to 9.8 μg/g ww for the 1980s and 1990s (Annex Table 6·A5), which exceed NOEL and LOEL levels for hatching success, deformities, and egg production in white leghorn chicken (Figure 6·59). These levels also overlap or

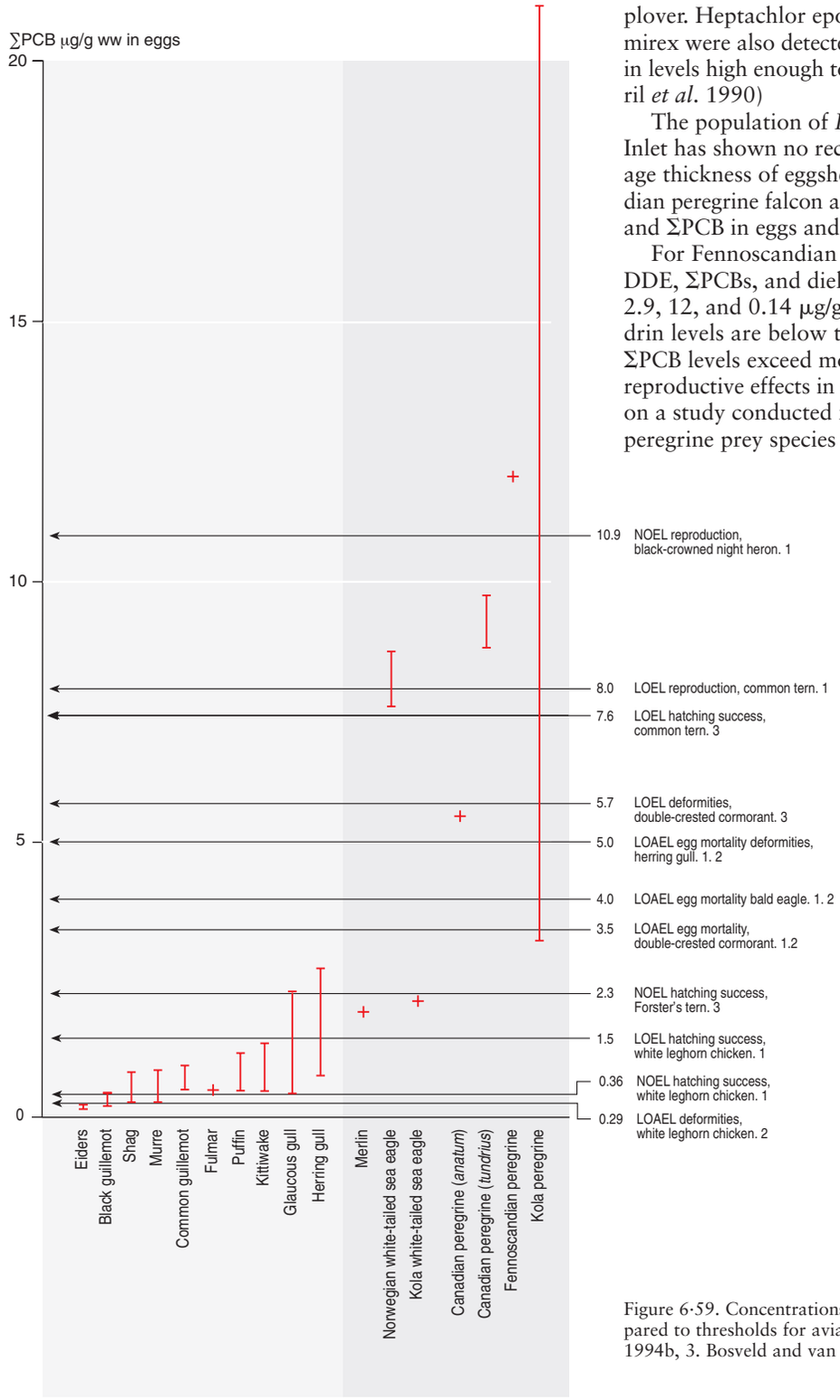



Figure 6·59. Concentrations of ΣPCB (µg/g ww) in Arctic bird eggs compared to thresholds for avian effects (1. Barron *et al.* 1995, 2. Giesy *et al.* 1994b, 3. Bosveld and van den Berg 1994).

exceed NOEL and LOEL levels for reproductive endpoints in a number of wild bird species. HCB levels in peregrine falcons are several orders of magnitude below the NOEL found in kestrels and lindane has not been detected in the few samples analyzed.

Baril *et al.* (1990) concluded that only three residues in prey species – DDE, ΣPCBs, and dieldrin – are likely to affect reproduction in peregrines. Dietary LOEC levels for ΣPCBs (5 µg/g ww) were exceeded in oldsquaw and pintail. Dietary LOEC levels for DDE (5 µg/g ww) were exceeded in oldsquaw, water pipit (*Anthus spinoletta*), and semipalmated plover. Dietary LOEC levels for dieldrin (0.1 µg/g ww) were exceeded in oldsquaw, pintail, and semipalmated plover. Heptachlor epoxide, ΣDDT, hexachlorobenzene, and mirex were also detected in some of the prey samples, but not in levels high enough to affect reproduction in peregrines (Baril *et al.* 1990)

The population of *F. p. tundrius* nesting around Rankin Inlet has shown no recent overall improvement in the average thickness of eggshells. Thus, Arctic populations of Canadian peregrine falcon are still at risk from high levels of ΣDDT and ΣPCB in eggs and in their food.

For Fennoscandian peregrine falcons, arithmetic means of DDE, ΣPCBs, and dieldrin for 1991-1994 (32 clutches) were 2.9, 12, and 0.14 µg/g ww, respectively. The DDE and dieldrin levels are below those expected to cause effects, but ΣPCB levels exceed most of the NOEL and LOEL levels for reproductive effects in other bird species (Figure 6·59). Based on a study conducted in the 1970s (Lindberg *et al.* 1985), no peregrine prey species had ΣDDT or ΣPCB levels that exceed the LOEC for reproductive effects. As OC levels seem to have decreased in northern Sweden since the 1970s, this is probably still the case.

Peregrine falcon eggs collected from the Kola Peninsula, Russia, in 1991 had a geometric mean of 3.5 µg DDE/g ww (Henny *et al.* 1994), which is well below levels expected to cause effects. The geometric mean for dieldrin was 0.059 µg/g ww, which is also below effect levels. However, concentrations of ΣPCB ranged between 3 and 21 µg/g ww (geometric mean of 7.3 µg/g ww). These ΣPCB levels exceed most of the NOEL and LOEL levels for reproductive effects in other bird species (Figure 6·59). The PCDD/F levels found (14-95 pg TEQ/g ww) are above some NOAEL and LOAEL

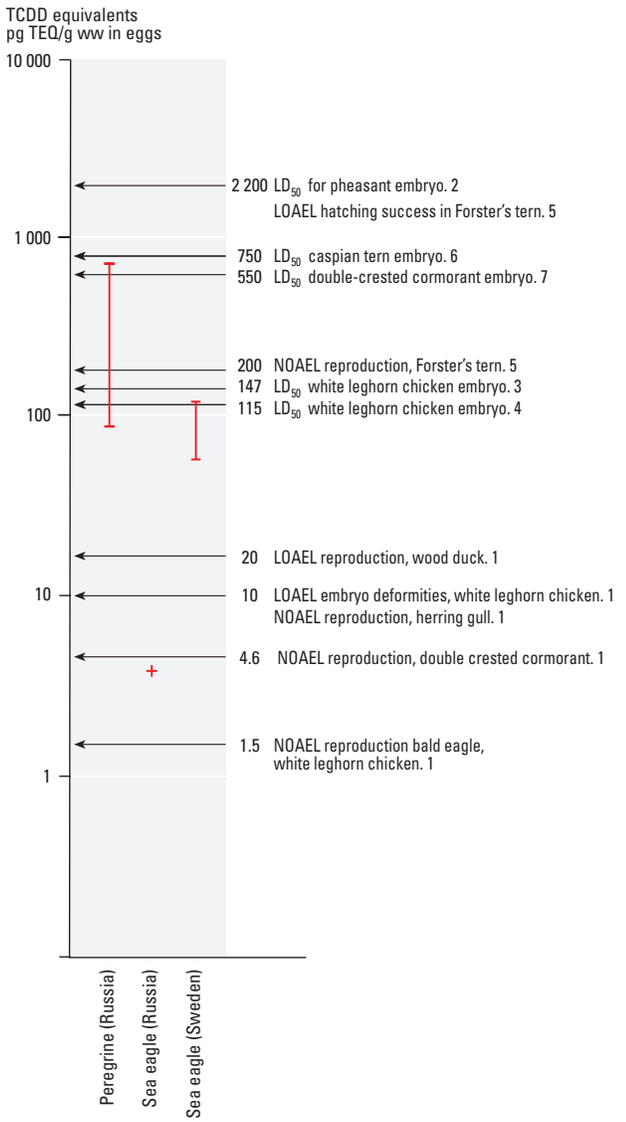



Figure 6·60. Concentrations of PCDD/Fs and non-*ortho* and mono-*ortho* PCBs as TCDD equivalents (pg/g ww) in Arctic bird eggs compared to thresholds for avian effects (1. Giesy *et al.* 1994b, 2. Nosek *et al.* 1992, 3. Verrett 1976, 4. Henschel 1993, 5. Kubiak *et al.* 1989, 6. Ludwig *et al.* 1993, 7. Tillitt *et al.* 1992).

levels for reproductive effects in other bird species (Figure 6·60). However, if the planar CB congeners 77, 126, 169, 105, and 118 are included in the TEQ calculations (72-545 pg TEQ/g ww), the combined TEQ levels (86-640 pg TEQ/g ww) exceed most NOAEL and LOAEL levels for reproductive effects in birds, and exceed the $LD_{50}$ for white leghorn chicken and double-crested cormorant embryos.

### 6.8.1.3.2. **Merlin**

• *Reproduction*

The Fennoscandian population of merlin (*Falco columbarius aesalon*) started to decline during the 1950s, and was very low around 1960. The population has recovered to a certain extent since then (Wallin 1984), and in 1991, the breeding population in Norway was estimated to be 2000-6000 pairs (Gjershaug 1991). From 1947 up to 1990, eggs were on average 15% thinner than normal, and some eggs were 20-30% thinner than normal during the 1960s and 1970s. After 1990, the thinning has been close to 10%. The number of merlins on autumn migrations in southern Sweden and Norway were depressed during the years of thinning (Wallin 1984, Nygård *et al.* 1994).

• *Levels and intake assessment*

Merlin studied after 1990 still suffer from about 10% eggshell thinning. The mean concentration of ΣDDT was 26.8 μg/g ww, and for ΣPCB 1.82 μg/g ww, in merlin eggs collected from Alta, Norway. The ΣDDT levels are above those expected to cause reproductive failure in peregrine falcons, but the degree of eggshell thinning is less than expected from the DDE levels. This may mean that merlin are less sensitive to the effects of ΣDDT than peregrine falcon. Mean ΣPCB levels are in the range of NOEL and LOEL levels for reproductive effects in white leghorn chicken (Figure 6·59). No information is available on OC levels in merlin prey.

### 6.8.1.3.3. **White-tailed sea eagle**

• *Reproduction*

White-tailed sea eagle (*Haliaeetus albicilla*) populations in Lapland and along the coast of the Baltic Sea were studied between 1965-1978 for reproductive success (Helander *et al.* 1982, Helander 1983). Both populations showed reduced breeding success during this period. Reproduction was studied in relationship to DDE, PCB (total), dieldrin, HCB, and mercury concentrations in eggs from both populations. Reproduction was found to be significantly negatively correlated with egg residue levels of DDE and total PCB in the Baltic population. After the introduction of DDT (and PCB) during the 1940s, the average number of fledglings per nest of the white-tailed sea eagle declined from 1.7 a year to 1.2 in the 1980s (Helander 1990).

In 1995, the number of nesting pairs of the Baltic population of white-tailed sea eagle and the total number of fledglings had increased to pre-1940 levels (Helander 1996). At the same time, the northern population partly inhabiting the Arctic regions increased slightly, from about 20 pairs to more than 30 pairs. The increase was most probably explained by better protection.

New data from the 1980s and 1990s show that the reproductive success seems to be primarily linked to the residue levels of DDE in the eggs (Helander 1994). The DDE concentrations have declined such that no eggs collected during the 1980s and 1990s had DDE concentrations exceeding 600 μg/g lipid. During the 1960s and 1970s, two-thirds of the eggs exceeded this concentration. The reductions in DDE concentrations have in turn led to an improvement in productivity. During the 1960s and 1970s, only 8% of the eagles had a productivity of 1.0. For the 1980s and 1990s, this figure has risen to 27%.

The mean reproductive rate for white-tailed sea eagles from several populations along the Norwegian coast showed no correlation with OC concentrations in the eggs (Nygård and Skaare 1996). Significant correlations were found between eggshell thickness and concentrations of DDE, ΣPCB, and HCB. However, the degree of eggshell thinning in the 1990s was only 2-5%, well below that considered to be critical.

• *Levels and intake assessment*

Previous studies of white-tailed sea eagle populations in the Swedish Arctic (Lapland) and the Baltic Sea coast have related DDE, ΣPCB, dieldrin, HCB, and mercury concentrations in eggs to reproductive success. Critical ranges were estimated to be 500-600 μg DDE/g lipid (25-29 μg/g ww) and/or 800-900 μg ΣPCB/g lipid (39-44 μg/g ww). The Lapland eagle population had DDE concentrations of 117 μg/g lw (5.7 μg/g ww) and ΣPCB concentrations of 263 μg/g lw (13 μg/g ww) during the period 1965-1978, which are well below these critical concentrations. The decline in breeding success in the Lapland population was found to be due to

food shortages and human disturbance. The decline in ΣDDT levels in northern Sweden from the 1970s to the present indicates that Lapland sea eagles are not currently at risk from the reproductive effects of ΣDDT.

The PCDD/F levels found in Swedish sea eagle eggs collected in 1989 (7.5-8.4 pg TEQ/g ww) (Annex Table 6·A22) are above some NOAEL levels for reproductive effects in other bird species (Figure 6·60). However, if the planar PCBs (CBs 77, 126, 169, 105, 118) (50-110 pg TEQ/g ww) are included in the TEQ calculations, the combined TEQ levels exceed most NOAEL and LOAEL levels for reproductive effects in birds, and approach the $LD_{50}$ for white leghorn chicken embryo.

Mean DDE and ΣPCB levels in Norwegian white-tailed sea eagle eggs from the coastal population between 66 and 69°N (Annex Table 6·A5) are in the same range as those for peregrine falcons (Figure 6·59). However, sea eagles seem to be less sensitive to the effects of ΣPCB and ΣDDT than peregrine falcons. These concentrations are also below the levels found to reduce breeding success in the Swedish population of white-tailed sea eagles. On the other hand, populations at more southerly latitudes along the Norwegian coast have higher concentrations, some of which exceed these levels. Levels of mirex, dieldrin, and HCB were not considered to be of biological significance (Nygård and Skaare 1996). However, the mean ΣPCB levels in the northerly population do exceed or overlap most NOEL and LOEL levels for subtle reproductive effects.

Results of OC analyses in one white-tailed sea eagle egg from the Kola Peninsula, Russia are available (Henny *et al*. 1994). The DDE level was 0.7 μg/g ww, well below the effect level given above. The ΣPCB level was 2.0 μg/g ww, which exceeds a few NOEL and LOEL levels for reproductive endpoints in other bird species (Figure 6·59). Levels of PCDD/Fs were 5 pg TEQ/g ww and nPCBs (CBs 77, 126, 169) and mono-*ortho* PCBs (CBs 105, 118), 12 pg TEQ/g ww. The combined TEQs for PCDD/Fs and planar PCBs exceed some NOAEL and LOAEL values for reproductive effects in other bird species (Figure 6·60).

Based on mean OC levels in reindeer and fish from the Swedish Arctic (Annex Tables 6·A4 and 6·A9), dietary concentrations of ΣDDT and ΣPCB are much lower than those estimated to lead to reproductive effects. The ΣDDT and ΣPCB levels in freshwater fish in the Swedish Arctic are also below the various environmental quality guidelines established to protect fish-eating wildlife (Figures 6·61 and 6.62; next page). PCDD/F and planar PCB levels in burbot liver (23 pg TEQ/g ww) exceed all the guidelines (Table 6·13).

Norwegian sea eagles prey mainly on deep-water marine fish species (Staven 1994). Based on dietary LOEC levels for ΣPCB (5000 ng/g ww), HCB (100 000 ng/g ww), DDE (5000 ng/g ww), and dieldrin (100 ng/g ww) in peregrine falcon prey, levels in Arctic marine fish are several orders of magnitude below those expected to cause effects. However, ΣPCB and ΣDDT levels in a range of fish species from many Arctic sites do exceed some, and in a few cases for ΣPCB, all of the environmental quality guidelines for protecting fish-eating wildlife given in Table 6·13 (Figures 6·61 and 6·62). This implies that dietary intakes of ΣDDT and ΣPCB in sea eagles may be high enough to lead to effects. PCDD/F levels have been analyzed in Atlantic cod (Annex Table 6·A19) from northern Norway (Schlabach and Skotvold 1996a, 1996b) and the levels were below guidelines.

For ΣDDT, the fish species around Norway that exceed both Canadian and USEPA guidelines are Arctic cod (liver) from some Barents Sea sites, Atlantic cod (liver) from all sites in the Barents Sea, and redfish (liver) from most sites in the North Atlantic. None of the fish species analyzed had ΣDDT levels that exceeded the International Joint Commission guideline of 1000 ng/g ww (Figure 6·62). For ΣPCB, the species that exceeds Canadian, International Joint Commission, and USEPA guidelines is Atlantic cod (liver) from the Barents Sea (Figure 6·61).

No information is available on OC levels in white-tailed sea eagle prey from the Kola Peninsula, Russia.

#### 6.8.1.3.4. **Gyrfalcon**

No biological effects studies have been conducted on gyrfalcon. DDE and ΣPCB levels in Canadian gyrfalcon eggs from the 1980s are several orders of magnitude lower than those considered to cause reproductive effects. Analytical results are available for gyrfalcon from Iceland, but it is difficult to assess these, as the tissue analyzed was muscle (Annex Table 6·A5). Based on dietary LOEC levels for ΣPCB, HCB, DDE, and dieldrin in peregrine falcon prey, levels in gyrfalcon prey (hare, lemming, ptarmigan) in NWT Canada are several orders of magnitude below those expected to cause effects.

### 6.8.1.4. **Wolf and red fox**

No biological effects studies have been conducted on wolf or red fox. Concentrations of ΣPCB in these species from the Canadian Arctic are several orders of magnitude lower than those expected to result in effects on reproduction. The levels in caribou/reindeer are also much lower than the dietary concentrations expected to cause such effects.

### 6.8.1.5. **Mustelids**

Mink, and probably other mustelids, are extremely sensitive to the effects of PCB. Aside from observations on population changes associated with decreased reproduction, no other biological effects have been studied in Arctic mustelids.

#### 6.8.1.5.1. **Mink**

• *Reproduction*

Population indices derived from age and sex ratios of the mink harvest (Strickland and Douglas 1987) suggest light harvest pressure and healthy reproductive performance in mink from the Northwest Territories in Canada. These indices, coupled with the comparatively low levels of contaminants, suggest little or no effects on reproduction or population health of NWT mink as a result of these contaminants.

The American mink (*Mustela vison*) was introduced to Scandinavia in the 1940s, after which there was a rapid increase in numbers followed by a leveling-off in the 1960s (Gerell 1967, 1971). The species was spread over the entire country, including the remote Arctic areas, and the development of the population has been similar in various parts of the country. At the end of the 1970s, the population suddenly started to increase rapidly and mink are now more common than during the 1960s (Alsberg *et al*. 1993). The increase followed a more than 50% reduction in environmental concentrations of PCB between 1975 and 1978 (Olsson and Reutergårdh 1986).

• *Levels and intake assessment*

Platonow and Karstad (1973) reported that 1230 ng/g ww of Aroclor 1254 in mink liver tissue corresponded to impaired reproductive success. Reduced growth and survival of mink kits were observed in female mink with 2000 ng/g ww Aroclor 1254 in liver tissue (Wren *et al*. 1987a, 1987b).

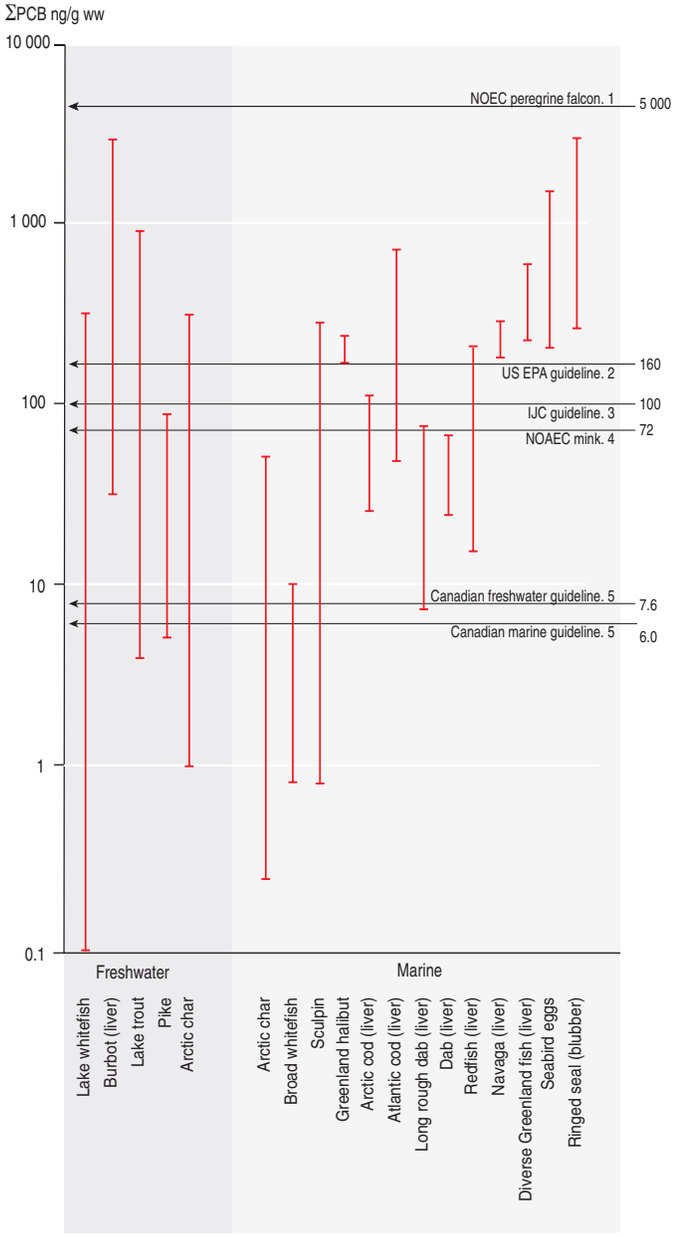



Figure 6·61. ΣPCB (ng/g ww) in Arctic animals as food items compared to no-effect-concentrations (NOAEC) and environmental quality guidelines for protecting fish-eating/aquatic wildlife (1. Baril *et al.* 1990, 2. USEPA 1995, 3. De Vault *et al.* 1995, 4. Giesy *et al.* 1994a, 5. Environment Canada 1996).

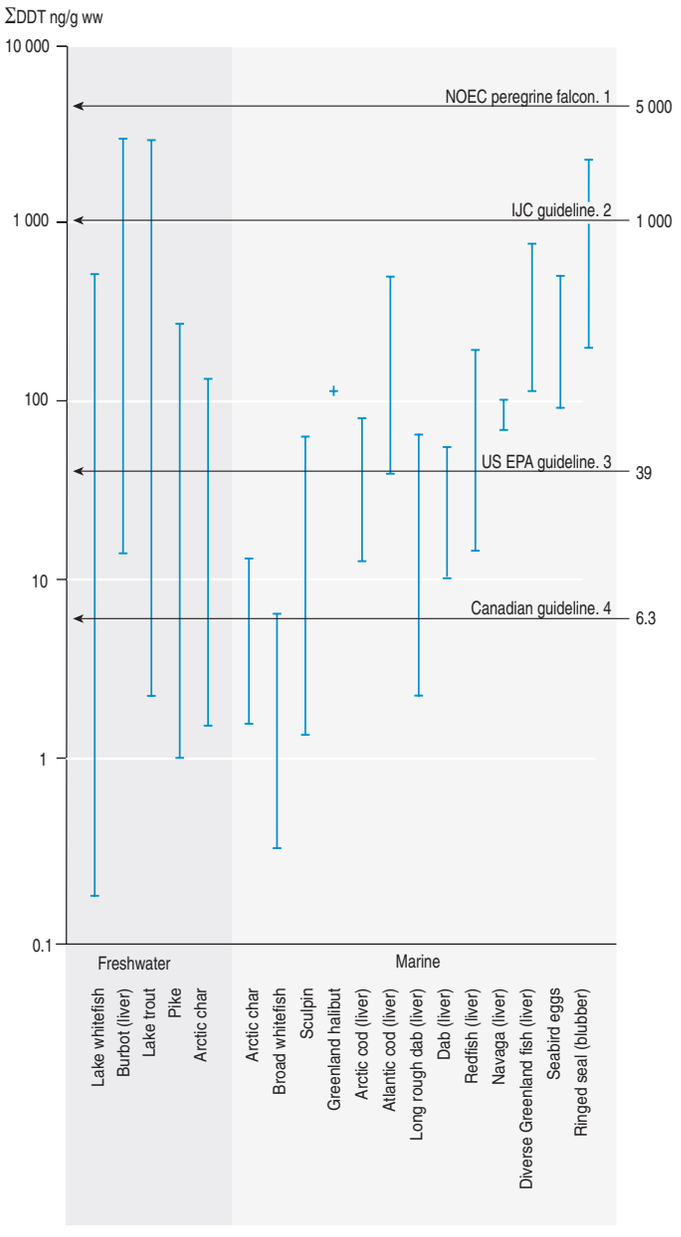



Figure 6·62. ΣDDT (ng/g ww) in Arctic animals as food items compared to no-effect-concentrations (NOAEC) and environmental quality guidelines for protecting fish-eating/aquatic wildlife (1. Baril *et al.* 1990, 2. USEPA 1995, 3. De Vault *et al.* 1995, 4. Environment Canada 1996).

Olsson and Sandegren (1991a, 1991b) proposed an $EC_{50}$ of 50 000 ng total PCB/g lw and Kihlström *et al.* (1992) proposed an $EC_{50}$ of 65 000 ng/g lw and a no-effect-level of 9000 ng/g lw for litter size in mink, based on muscle concentrations.

In a recent reassessment of all reproductive studies of PCB on mink, the $EC_{50}$ in adult females for litter size was calculated to be 40 000-60 000 ng total PCB/g lw (ca. 1200 ng total PCB/g ww) and 2400 ng/g ww for kit survival in muscle (Leonards *et al.* 1995). Similarly, the $EC_{50}$ for dioxin-like compounds was calculated to be 160 pg TEQ/g ww for litter size and 200 pg TEQ/g ww for kit survival (Leonards *et al.* 1995). In experiments, Heaton (1992) found the LOAEL for kit survival to be 490 pg TEQ/g ww.

Giesy *et al.* (1994c) have estimated the dietary no-adverse-effect-concentration (NOAEC) of ΣPCB, dioxin-like compounds, dieldrin, and ΣDDT for reproductive effects in mink to be 72 ng ΣPCB/g ww food, 2 pg TEQ/g ww food, 5000 ng dieldrin/g ww food, and 100 000 ng ΣDDT/g ww food, respectively. For heptachlor epoxide, the dietary NOAEC for adult mink was 50 000 ng/g ww food (Aulerich *et al.* 1990) and the LOAEC for kit growth was 6250 ng/g ww (Crum *et al.* 1993). No information on the sensitivity of mink to toxaphene is available, however, a NOAEC of 4000 ng toxaphene/g ww food for thyroid effects has been estimated from studies in rats and dogs (Chu *et al.* 1986).

The mean values of ΣPCB (43 congeners) found in mink liver from the western Canadian Arctic range from 5.3 ng/g to 92 ng/g ww (93-1670 ng/g lw) (Poole *et al.* in press). These values are considerably lower than the $EC_{50}$'s for kit survival and litter size and the NOEL for kit survival (Figure 6·63). ΣPCB levels in mink from Grand Baleine, Quebec, Canada are considerably higher, 160-350 ng/g ww (2700-8000 ng/g lw), and, on a lipid weight basis, are just below the NOEL for litter size in mink. Recent assessments based on subtle neurobehavioral effects in offspring of rhesus monkeys treated with PCBs and human mothers eating PCB-contaminated fish have resulted in an estimated LOAEL for effects

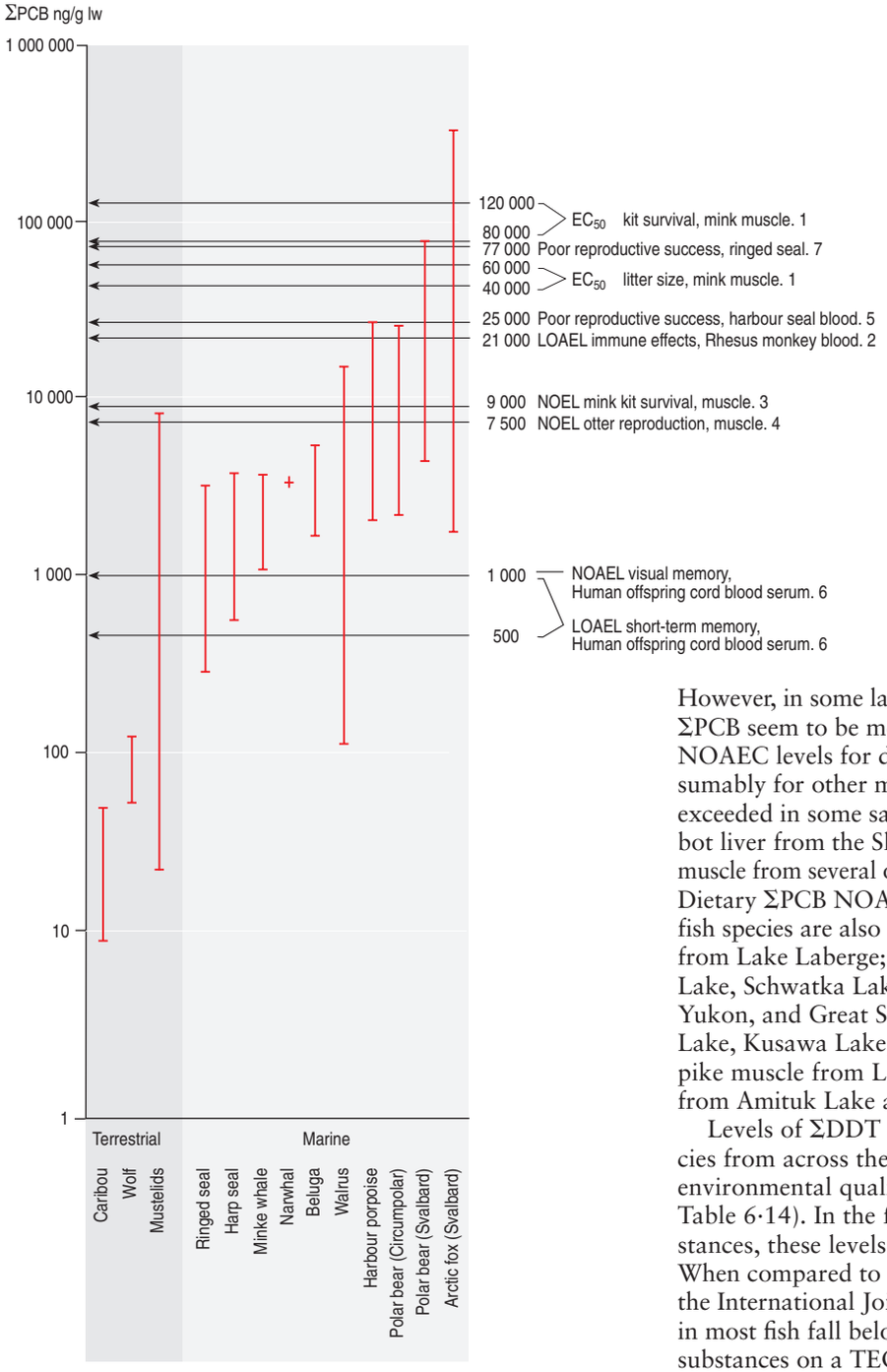



Figure 6·63. ΣPCB (ng/g lw) in Arctic mammals compared to thresholds for mammalian effects (1. Leonards *et al.* 1995, 2. Tryphonas 1994, 3. Kihlström *et al.* 1992, 4. Olsson *et al.* 1996a, 5. Boon *et al.* 1987, 6. Ahlborg *et al.* 1992, 7. Helle *et al.* 1976b).

on short-term memory of 500-1000 ng/g lw and a NOAEL for effects on visual memory of 1000 ng/g lw in blood serum (Ahlborg *et al.* 1992). Only mink from Fort Providence and Grand Baleine exceed these values.

PCDD/F levels in the few mink liver samples analyzed range from 0.2-0.5 pg TEQ/g ww, which are several orders of magnitude lower than the $EC_{50}$ for litter size and kit survival in mink (160-200 pg TEQ/g ww).

Based on mean OC levels in fish from sampled lakes in the Canadian and Swedish Arctic (Annex Table 6·A9), and assuming that mink only eat fish, concentrations of dieldrin, ΣDDT, and heptachlor in fish are generally lower than those estimated to lead to reproductive effects in mink, and toxaphene levels are below those associated with thyroid effects. However, in some lakes, levels of PCDD/Fs, planar PCBs, and ΣPCB seem to be more problematic. For Canada, dietary NOAEC levels for dioxin-like compounds in mink, and presumably for other mustelids such as marten and ermine, are exceeded in some samples of walleye (whole body) and burbot liver from the Slave River as well as char and lake trout muscle from several other Canadian lakes (Annex Table 6·A22). Dietary ΣPCB NOAEC levels in tissues from a number of fish species are also exceeded. These include whitefish muscle from Lake Laberge; burbot liver from Lake Laberge, Atlin Lake, Schwatka Lake, Teslin Lake, several other lakes in the Yukon, and Great Slave Lake; lake trout muscle from Peter Lake, Kusawa Lake, Lake Laberge, and several Yukon lakes; pike muscle from Lake Laberge; and, Arctic char muscle from Amituk Lake and Char Lake (Annex Table 6·A9).

Levels of ΣDDT and ΣPCB in most freshwater fish species from across the Arctic approach or exceed Canadian environmental quality guidelines (Figures 6·61 and 6·62, Table 6·14). In the few fish analyzed for dioxin-like substances, these levels also exceed the Canadian guidelines. When compared to the USEPA assessment as well as that of the International Joint Commission, ΣPCB and ΣDDT levels in most fish fall below the guidelines, except for dioxin-like substances on a TEQ basis. ΣPCB and ΣDDT levels exceeding these guidelines are found primarily in fish from lakes known to have been contaminated.

#### 6.8.1.5.2. Otter

• *Reproduction*

The otter (*Lutra lutra*) populations in Sweden, which inhabit both the southern parts as well as the Arctic parts of Sweden, declined suddenly in the entire country during the 1950s-1970s (Erlinge 1980, Olsson *et al.* 1996a, 1996b). PCBs have been suggested to have caused the decline (Sandegren *et al.* 1980, Olsson and Sandegren 1991a, 1991b). In the 1980s, the species was only found in southern Sweden in some very restricted areas and in the Arctic north, inventory studies clearly showed that the species was found only in isolated groups (Olsson *et al.* 1988, Olsson and Sandegren 1991a, 1991b). A sudden increase of the northern population was seen at the end of the 1980s and in the 1990s (Olsson *et al.* 1996a).

• *Levels and intake assessment*

Median values for total PCB concentrations in otter muscle samples from the northern and Arctic areas of Sweden during the 1990s were 7500 ng/g lw (Olsson *et al.* 1996a). The available data from time trend studies in otter, comparing ΣPCB concentrations and reproductive success, suggest that 7500 ng/g lw is approximately the NOEL for reproductive effects. This is also close to the estimated NOEL of 9000 ng/g lw given above for mink. Thus, otters from the Swedish Arctic and Norwegian Arctic coast seem to have ΣPCB levels at or below the NOEL for reproductive effects. However, they exceed the NOAEL and LOAEL levels for subtle neurobehavioral effects (Figure 6·63). Levels of PCDD/Fs and nPCBs (1.5-12 pg TEQ/g ww) are also below the LOAEL and $EC_{50}$ values for mink reproduction given above. However, the lipid weight levels of PCDD/Fs and nPCBs (100-270 pg TEQ/g) are above those associated with immunosuppression in harbour seals (Figure 6·57).

For Sweden, current ΣPCB and ΣDDT levels in fish do not pose a threat to mink or otter. The dietary PCDD/F and planar PCB NOAEC levels for mink, and presumably for otter as well, (on a TEQ basis) are exceeded only in burbot liver from Pajala, along the River Torneälv. The OC levels in fish are also just below the various guidelines for protecting fish-eating wildlife, except for PCDD/F and planar PCB levels in burbot liver (23 pg TEQ/g ww) which exceed all the guidelines.

## 6.8.2. Freshwater environment

### 6.8.2.1. Fish

• *Cytochrome P450 activities*

A variety of studies examining mixed function oxidase enzyme activity in freshwater fish, including studies with different species, at different latitudes, and in systems with different known degrees of contamination have been performed. In general, results show that, in various comparisons, higher levels of enzyme induction relate to 1) more southerly latitudes, 2) higher trophic levels, 3) polluted systems, and in one case, 4) an inferior consumable product for human beings.

Figure 6·64 shows liver microsomal AHH (aryl hydrocarbon hydroxylase) activities in collections of lake trout

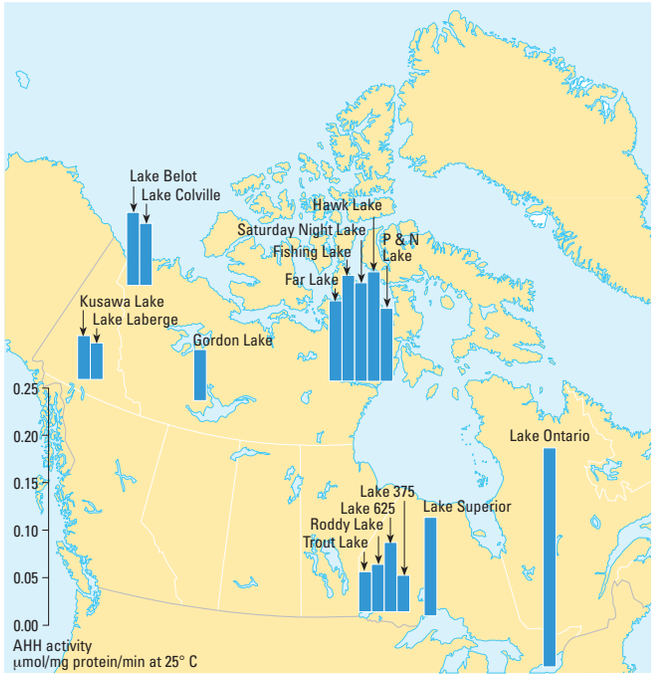



Figure 6·64. AHH induction in lake trout from Canada (Muir *et al.* 1997).

Table 6·19. Mixed function oxidase enzyme activity in liver from fish in Lake Laberge and Kusawa Lake.

| Location/ Species | Sex | N | EROD | AHH | TEQ (pg/g) [a] |
|---|---|---|---|---|---|
| *Lake Laberge* | | | | | |
| Lake trout | M | 5 | 0.0130±0.0040 | 0.0442±0.0247 | 47±42 |
| | F | 2 | 0.0085 | 0.0350 | 11 |
| Burbot | M | 10 | 0.0095±0.0040 | 0.0077±0.0040 | 255±130 |
| | F | 10 | 0.0041±0.0020 | 0.0061±0.0036 | 83±14 |
| Whitefish | F | 6 | 0.0132±0.0061 | 0.0373±0.0142 | |
| *Kusawa Lake* | | | | | |
| Lake trout | M | 12 | 0.0226±0.0139 | 0.0547±0.0234 | |
| | F | 8 | 0.0134±0.0107 | 0.0359±0.0185 | |

a. Results from Muir (1993).

from several locations in northern Canada. Two collections from the Great Lakes are also included. The collection from Lake Ontario shows higher levels than any of the Arctic collections. It is assumed that this is due to the fact that these fish are exposed to higher amounts of a variety of chemical contaminants.

Muir and Lockhart (1993b) examined mixed function oxidase enzyme activity in liver from fish in Lake Laberge and Kusawa Lake in the Yukon. Liver samples were assayed for two cytochrome-P-450 1A-related activities, ethoxyresorufin-O-deethylase (EROD), and AHH levels (Table 6·19). EROD activities in lake trout liver from both lakes were similar despite higher PCB levels in the Lake Laberge fish. Higher levels of TEQs were found in burbot (*Lota lota*) liver than in lake trout muscle from Lake Laberge, but EROD and AHH activities were lower in burbot. EROD activities in male lake trout from Lake Laberge are about two-fold higher than those in Buchanan Lake (79°N, Canadian High Arctic) Arctic char (*Salvelinus alpinus*), but similar to levels observed in char from Amituk Lake (75°N, eastern Canadian Arctic). (Lockhart and Stewart 1992). There appeared to be no correlation between TCDD equivalents (TEQ) for planar PCBs and enzyme activity.

Lockhart *et al.* (in press) examined liver of burbot from western and northern Canada for four mono-*ortho* PCB congeners (CBs 105, 114, 118, and 156) and for EROD and AHH. Collections ranged from a small lake in northwestern Ontario (51°N, 96°W) to the Peel River in the Northwest Territories (67°N, 135°W). Congeners varied among sites, but there was no consistent correlation between congeners, nor was there a geographic pattern.

Inferior fish quality has been linked with hydrocarbon-induced stress, which is also linked with induction of liver mixed function oxidase enzymes (Muir *et al.* 1990a). Coincident with oil field expansion at Norman Wells, NWT, fishermen in downstream communities complained that the quality of fish had deteriorated. Specifically, the liver of burbot was reported to have become small and dark in color, and the flesh of whitefish 'watery'. Examination of the burbot revealed that the liver condition was associated with a low content of fat, and analysis of the whitefish revealed some of the highest tissue water contents on record.

• *Bone collagen and hydroxyproline*

Bone hydroxyproline has been shown to be depressed in fish during exposure to toxaphene (Mayer *et al.* 1977). Lake trout collected in 1992 from Lake Laberge had significantly lower levels of hydroxyproline than lake trout from Kusawa Lake or from northwestern Ontario (Lake 260) (Lockhart and Muir 1996). To further identify causes of this depression,

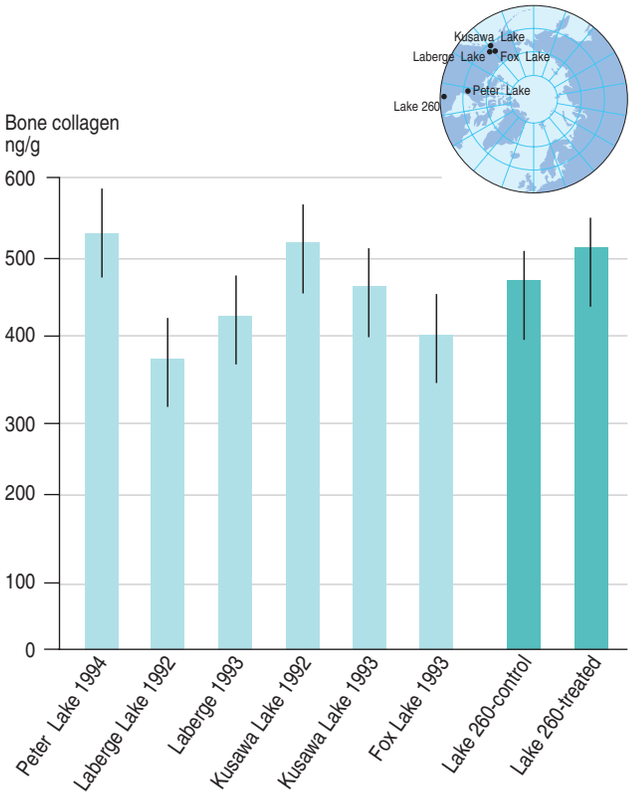



Figure 6·65. Comparison of bone collagen in lake trout from Peter Lake, NWT and from Yukon lakes, and in toxaphene-treated and control fish from Lake 260 (Delorme 1995).

fish in Lake 260 were treated with a single intraperitoneal dose of 7000 ng/g toxaphene, tagged and returned to the lake (Delorme 1995). No differences were seen in mean levels of hydroxyproline or calcium between treated and control fish from L260. A repeat analysis of trout bone collected in 1993 from Lake 260 also showed lower, but not significantly different, bone collagen than from Kusawa Lake and very similar levels to those for trout from Fox Lake. Lake trout from Peter Lake, NWT had higher bone collagen and lower toxaphene concentrations than the trout from Yukon lakes (Figure 6·65). No correlation was observed between toxaphene levels and collagen or hydroxyproline in 19 trout from Peter Lake. Thus, bone collagen did not prove to be a strong indicator of toxaphene exposure in lake trout in contrast to results for fathead minnows (*Pimephales promelas*) in the laboratory.

• *Levels and intake assessment*

Mayer *et al.* (1977) found that adult fathead minnows and channel catfish (*Ictalurus punctatus*) exposed to toxaphene had no decreases in hydroxyproline levels, but levels in exposed offspring were significantly decreased. Toxaphene concentrations of 3400 ng/g ww in channel catfish fry tissues were associated with decreased growth and 600 ng/g led to altered bone development (Stickel and Hickey 1977), identifying offspring as being more sensitive than adults to toxic effects. Mayer *et al.* (1975) exposed brook trout to toxaphene in water and found higher mortality during spawning. A 50% mortality was associated with toxaphene concentrations of 870 ng/g ww in muscle and 2400 ng/g ww in whole body. Delorme *et al.* (1993) found decreased survival in lake trout treated with 7000 ng/g body weight, which was extrapolated to be equivalent to a whole body concentration of 4800 ng/g ww.

Mean toxaphene levels in freshwater fish do not exceed effect levels. However, fish in a number of lakes have mean toxaphene levels that are not far from those expected to affect bone development and mortality during spawning, indicating that some individuals may possibly exceed these. Besides Lake Laberge, these include several lakes in the Yukon (Bennett, Tagish, and Marsh Lakes), Atlin Lake in northern British Columbia, and Great Slave Lake in the NWT (Annex Table 6·A9). Information on toxaphene levels in fish from other parts of the Arctic is limited relative to other OCs.

## 6.8.3. Marine environment

### 6.8.3.1. Invertebrates

Exposure to TBT in harbors with significant boat moorage is responsible for the development of imposex in females of the common whelk (*Buccinum undatum*) in Kongsfjorden, Svalbard, dog whelks (*Nucella lapillus*) in Norway and Iceland, and a marine snail (*Nucella lima*) in Alaska (Short *et al.* 1989, Harding *et al.* 1992, Brick and Bolte 1994, Berge 1995, Berge *et al.* 1995, Svavarsson and Skarphédinsdóttir *et al.* 1996). The banning of TBT use on small boats has resulted in some signs of recovery for invertebrate species near marinas and other heavy-use areas. Some recovery from imposex in populations of whelks has been observed in the Pacific northwest (Tester *et al.* 1996), but it is probably still too early to have an overall view of the environmental response to this partial regulation (Stewart and Thompson 1994). The difficulty in evaluating the environmental response is that 1) imposex is initiated by extremely low concentrations of TBT making it difficult to relate cause and effect below analytical detection limits, 2) many of the indicator species live for more than six years, therefore, since effects are irreversible, observed responses may be indicative of past rather than current exposure and, 3) other compounds or environmental circumstances may also produce imposex (Evans *et al.* 1995, Ellis 1991), although if so, none have yet been identified.

No information on the biological effects of OCs is available for Arctic marine invertebrates.

### 6.8.3.2. Fish

No studies of biological effects have been carried out on Arctic anadromous or marine fish. As was stated under section 6.8.2.1 on freshwater fish, effect levels for toxaphene in channel catfish fry were 3400 ng/g ww for growth and 600 ng/g ww for bone development, and for brook trout, 870 ng/g ww for mortality during spawning. Based on mean toxaphene levels in muscle (Annex Table 6·A17), only Greenland halibut (*turbot*) from the Canadian Arctic have levels close to the effect levels for bone development and mortality during spawning (570-670 ng/g ww).

### 6.8.3.3. Seabirds

• *Levels and intake assessment*

In general, ΣPCB levels in seabirds analyzed from the Canadian and Norwegian Arctic approach or exceed reproductive NOEL and LOEL levels at the low range of the scale compared to peregrine falcons (Figure 6·59). However, no studies of biological effects have been carried out on Arctic seabirds. Mean ΣPCB levels in eider eggs are below those expected to result in reproductive effects (Figure 6·59). ΣPCB levels in shag (a species of cormorant) and fulmar are at or exceed the LOAEL for embryo deformities and the lower NOEL for hatching success in white leghorn chicken. In addition to the above, mean ΣPCB levels in puffin, murre, common guillemot, black guillemot, and kittiwake exceed

the upper NOEL for hatching success in white leghorn chicken (Figure 6·59).

Mean ΣPCB levels in glaucous gull and herring gull eggs are somewhat higher and approach or exceed the LOEL for hatching success in white leghorn chicken and the NOEL for hatching success found for Forster's tern. None of the Arctic seabirds studied appear to have ΣPCB levels high enough to be associated with egg mortality. However, it is difficult to assess the situation for Russian seabirds and for several seabird species at some Norwegian sites as other body tissues were analyzed rather than eggs. Mean ΣDDT, HCH, dieldrin, and HCB levels in all seabirds studied are several orders of magnitude below those expected to cause effects.

Based on dietary LOEC levels for ΣPCB (5000 ng/g ww), HCB (100 000 ng/g ww), DDE (5000 ng/g ww), and dieldrin (100 ng/g ww) in peregrine falcon prey, levels in anadromous and marine fish are several orders of magnitude below those expected to cause effects. Mean dieldrin levels in Arctic anadromous and marine fish from Canadian, Norwegian, Greenlandic, Icelandic, and Russian waters (Annex Table 6·A17) do not exceed any of the guidelines for protecting fish-eating wildlife (Table 6·14). However, ΣPCB and ΣDDT levels in a range of fish species from many Arctic sites do exceed some, and in a few cases for ΣPCB, all of the environmental quality guidelines for protecting fish-eating wildlife given in Table 6·14 (Figures 6·61 and 6·62). This implies that dietary intakes of ΣDDT and ΣPCB in fish-eating seabirds may be high enough to lead to effects if seabirds prey on these fish species. PCDD/F and/or nPCB levels have been analyzed in a few anadromous and marine fish (Annex Table 6·A19) from Canada and northern Norway. Sculpin from Cambridge Bay and anadromous Arctic char from several Canadian sites have PCDD/F or nPCB levels (1.1-4.5 pg TEQ/g ww) that exceed the guidelines.

For ΣDDT, the fish species that exceed both Canadian and USEPA guidelines are short-horned sculpin from Scoresbysund, Greenland; Greenland halibut from Cumberland Sound, Beaufort Sea and Davis Strait; Arctic cod (liver) from some Barents Sea sites and southern Novaya Zemlya; Atlantic cod (liver) from all sites in the Barents Sea, around Iceland, Halten Banken, and the Faeroe Islands; long-rough dab (liver) around Iceland; dab (liver) from some Icelandic sites; redfish (liver) from most sites in the North Atlantic as well as Davis Strait (West Greenland); navaga (liver) from the east Pechora Sea; and liver from tusk, blue hake, black dogfish, roughhead grenadier, and smalleyed rabbit fish from Davis Strait. None of the fish species analyzed had ΣDDT levels that exceeded the International Joint Commission guideline of 1000 ng/g ww (Figure 6·62).

For ΣPCB, the species that exceed Canadian, International Joint Commission, and USEPA guidelines are four-horned sculpin from Cambridge Bay, Greenland halibut from Cumberland Sound and the Beaufort Sea, Atlantic cod (liver) from the Barents Sea and Halten Banken, redfish (liver) from Halten Banken, navaga from the eastern Pechora Sea, and tusk, blue hake, black dogfish, roughhead grenadier, and smalleyed rabbit fish (liver for all species) from Davis Strait (Figure 6·61). Many of these fish species are deep sea or bottom predators that are probably not generally available for seabirds, except possibly if brought up by commercial fishing.

Herring and glaucous gulls prey on eggs, chicks, and even adult seabirds. Based on OC levels found in eggs from guillemots, puffin, cormorant, and fulmar from the Canadian and Norwegian Arctic (Annex Table 6·A16), dieldrin, HCB, ΣDDT, and ΣPCB levels are below dietary LOEC levels for reproductive effects in peregrine falcon. Mean dieldrin levels in these seabird species' eggs also do not exceed any environmental guidelines for protecting aquatic wildlife. However, ΣDDT levels in these species from these sites exceed Canadian and USEPA guideline levels (Figure 6·62), and ΣPCB levels exceed all guideline levels (Figure 6·61).

### 6.8.3.4. **Cetaceans**

#### 6.8.3.4.1. **Beluga**

• *Cytochrome P450 activity*

The principal biomarker that has been used is the microsomal cytochrome P450 system since it has been associated both with exposures to planar aromatic compounds and with a cascade of other biological responses. The cytochrome P450 system in beluga whale from the Canadian Arctic has been characterized (White *et al.* 1994). EROD, PROD, and AHH activities were found to correlate with one another and to correlate with cytochrome P450 1A levels. The cytochrome P450 1A levels were also correlated with the concentrations of non-*ortho* and mono-*ortho* PCBs found in blubber. Little cytochrome P450 2B activity was found.

There is a strong correlation between liver microsomal EROD or AHH activities and blubber residues of several PCB congeners in a group of beluga whales from the western Canadian Arctic (Figure 6·66) (Lockhart and Stewart 1992, White *et al.* 1994). AHH activity in liver of beluga whales from the Mackenzie Delta was found to be correlated with the sum of mono-*ortho* and nPCB congeners, which probably carry most of the TCDD TEQs, since PCDD levels are low in Arctic marine mammals (Norstrom *et al.* 1990). These particular whales entered a freshwater lake system during the open-water season and became trapped there by ice at the onset of winter. Breathing holes in the ice became smaller and smaller and native hunters decided to take the whales in mid-winter. They were about 200 kg lower in body weight than typical for whales of their length should have been. The hypothesis suggested by this information is that the whales mobilized blubber fats during the period of poor feeding in freshwater and concomitantly mobi-

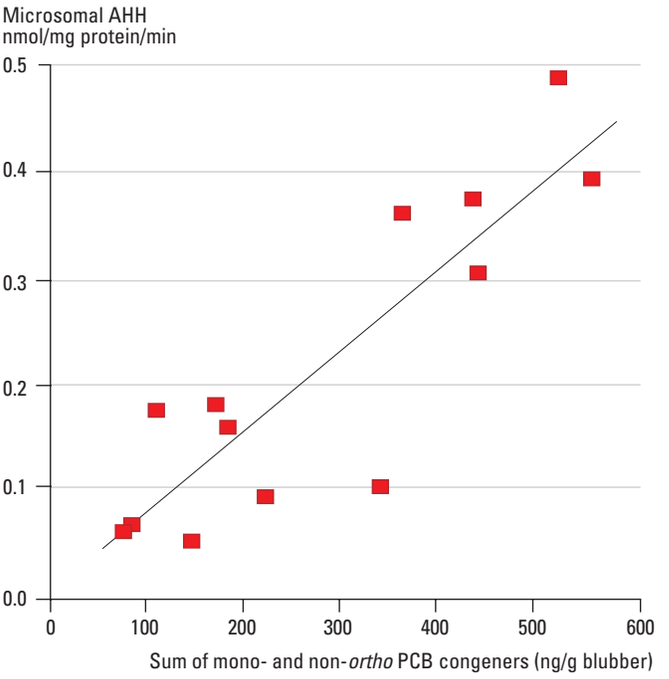



Figure 6·66. Aryl hydrocarbon hydroxylase (AHH, nmol/mg microsomal protein/min) activities in liver microsomes of beluga taken from the Eskimo Lakes area of the Mackenzie Delta, as a function of PCB congener concentrations in blubber (White *et al.* 1994).

lized fat-soluble contaminants which then acted to produce the correlations. While this is not a proof of a causal relationship between the PCBs and the bioindicators, it is probably the closest we can come to connecting residues and responses in animals that are not available for experimental study.

These observations on the beluga whales indicate that current body burdens of contaminants in an Arctic species can be associated with subtle effects on the animals. Clearly, they point to a possible interaction between diet and the pharmacology of organochlorines and suggest that existing body burdens may be sequestered effectively when blubber reserves are high, but may become a significant problem during a poor feeding season.

For obvious logistical reasons, this observation on whales cannot be reproduced. Nevertheless, the effect has also been demonstrated in fish in the laboratory. Arctic char received an initial low dose of PCB congener 126 and were then maintained at different feeding rates for several months. Fish receiving the low rations responded by increased cytochrome-P-450 catalytic activities (L. Lockhart unpubl. results).

• *Levels and intake assessment*

The mean ΣPCB levels range from 1880-6263 ng/g lw, which exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, but are below the NOEL for otter reproduction and mink kit survival (Figure 6·63). Belugas from three Canadian sites have been analyzed for nPCBs and, at one of these sites, for PCDD/Fs (Annex Table 6·A19). Experiments in harbour seals fed Baltic herring, indicated immunosuppression at total TEQ levels of around 210 pg/g lw in blubber. Of these, PCDD/Fs accounted for 18 pg TEQ, nPCBs for 51 pg TEQ, and mono- and di-*ortho* PCBs for 140 pg TEQ (Ross *et al*. 1995). The combined TEQs in beluga range from 4.2-25.3 pg/g ww in blubber, which is usually about 90% lipid. These TEQ levels are considerably lower than those associated with immunosuppressive effects (Figure 6·57).

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin, ΣDDT, and chlordanes in marine fish are several orders of magnitude below those expected to result in effects on reproduction in beluga whale. Toxaphene levels in marine fish are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in tissues from several fish species exceed the dietary NOAEC for reproduction of 72 ng/g ww (Figure 6·61). These include fourhorn sculpin from Cambridge Bay, short-horn sculpin (liver) from Scoresbysund, Greenland, Greenland halibut (muscle and/or liver) from Cumberland Sound, the eastern Beaufort Sea and Davis Strait, Arctic cod (liver) from several sites, Atlantic cod (liver) from all sites, redfish (liver) from several sites, navaga (liver) from the eastern Pechora Sea, tusk, blue hake, black dogfish, roughhead grenadier, and smalleyed rabbit fish (liver for all species) from Davis Strait (Annex Table 6·A17).

The assessment for dietary intake of dieldrin, PCB, DDT, and PCDD/F is the same as given for seabirds (section 6.8.3.3).

6.8.3.4.2. **Narwhal**

• *Levels and intake assessment*

No biological effects studies have been carried out on narwhal. OC levels are available from individuals from only one site in Baffin Bay. Mean ΣPCB levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (Figure 6·63). Narwhal from two Canadian sites have been analyzed for nPCBs and, at one of these sites, for PCDD/Fs. The nPCB TEQs range from 10-19 pg/g ww in blubber, which is about 90% lipid. PCDD/F TEQ levels were less than 1 pg/g ww. These levels are considerably lower than those associated with immunosuppressive effects (Figure 6·57).

Narwhal eat squid, Arctic cod, shrimp, and Greenland halibut (Muir *et al*. 1988a, 1992a). Mean dieldrin levels in Arctic cod and Greenland halibut (Annex Table 6·A17) do not exceed any of the guidelines for protecting fish-eating wildlife (Table 6·14). ΣDDT levels in marine fish do not exceed the NOEC for reproductive effects in mink. Mean ΣPCB levels exceed the NOEC for reproductive effects in mink in Greenland halibut from Cumberland Sound, the Beaufort Sea, and Davis Strait and Arctic cod (liver) from some Barents Sea sites and southern Novaya Zemlya. For ΣDDT, Canadian and USEPA guidelines are exceeded in Greenland halibut from Cumberland Sound, the Beaufort Sea, and Davis Strait and Arctic cod (liver) from some Barents Sea sites and southern Novaya Zemlya. For ΣPCB, Canadian, International Joint Commission, and USEPA guidelines are exceeded in Greenland halibut from Cumberland Sound and the Beaufort Sea (Figure 6·61).

6.8.3.4.3. **Minke whale**

• *Levels and intake assessment*

No biological effects studies have been carried out on minke whale. ΣDDT and ΣPCB levels are only available from individuals from northern Norway (southern Barents Sea). Mean ΣPCB levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (Figure 6·63).

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin, ΣDDT and chlordanes in marine crustaceans and fish are several orders of magnitude below those expected to result in effects on reproduction in minke whale. Toxaphene levels in marine fish are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in tissues from several fish species exceed the dietary NOAEC for reproduction of 72 ng/g ww (Figure 6·61). These are the same as given for beluga (section 6.8.3.4.1).

The assessment for dietary intake of dieldrin, PCB, DDT, and PCDD/F is the same as given for seabirds (section 6.8.3.3).

6.8.3.4.4. **Harbour porpoise**

• *Levels and intake assessment*

No biological effects studies have been carried out on harbour porpoise. The mean ΣPCB levels in animals from West Greenland are 2600 ng/g ww in blubber and 24 500 ng/g ww in blubber of animals from the southern Barents Sea. The NOAEL and LOAEL for subtle neurobehavioral effects are exceeded in harbour porpoise from both sites (Annex Table 6·A18). ΣPCB levels in the southern Barents Sea porpoises also exceed the NOEL for otter reproduction and mink kit survival and the levels associated with immunosuppression, and approach the levels associated with poor reproductive success in harbour seal (Figure 6·63).

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin, ΣDDT, and chlordanes in marine fish are several orders of magnitude below those expected to result in effects on reproduction in harbour porpoise. Toxaphene levels in marine fish are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in tissues from several fish species exceed the dietary NOAEC for reproduc-

tion of 72 ng/g ww (Figure 6·61). These are the same as given for beluga (section 6.8.3.4.1).

The assessment for dietary intake of dieldrin, PCB, DDT, and PCDD/F is the same as given for seabirds (section 6.8.3.3).

### 6.8.3.5. **Pinnipeds**

#### 6.8.3.5.1. **Seals**

• *Cytochrome P450 system*

EROD, AHH, and APND activities and cytochrome P450 levels were measured in 52 ringed seals (*Phoca hispida*) from Arviat, Canada and compared to organochlorine concentrations in blubber (Lockhart and Ferguson 1994). EROD, AHH, cytochrome P450 levels, and APND were found to correlate with each other and were all significantly correlated with dieldrin concentrations, also after correcting for age, sex, and lipid content. No relationship was found between APND activity and ΣDDT or chlordane concentrations. Some association was seen in female seals between APND activity and toxaphene concentrations. Associations were also seen between EROD, AHH and ΣPCB. A significant correlation was observed between ΣPCB levels in blubber and EROD levels in liver of hooded seal from the West Ice (Goksøyr and Skaare unpubl.).

• *Levels and intake assessment*

The mean ΣPCB levels in Arctic harp, ringed, harbour, and grey seals range from 241-5700 ng/g ww in blubber (85-95% lipid), which exceed the NOAEL and LOAEL for subtle neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (Figure 6·63). These levels are also considerably lower than those associated with poor reproductive success in harbour and ringed seal (Figure 6·63). Ringed seal and harp seal have been analyzed for PCDD/Fs or nPCBs from Svalbard, the Barents Sea, the Greenland Sea, and several sites in Canada (Annex Table 6·A19). Both groups of substances were analyzed at only two sites (Greenland Sea West Ice and Broughton Island, western Baffin Bay) (total TEQ of 29 and 21 pg/g ww, respectively), with the nPCBs comprising at least half of the combined TEQ value. The PCDD/F or planar ΣPCB TEQs for all other sites range from 2.4-38 pg/g ww in blubber (about 90% lipid). These levels are somewhat lower than the sum of PCDD/F and nPCB TEQs (69 pg TEQ/g lw) associated with immunosuppressive effects in harbour seal (Figure 6·57).

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin, ΣDDT, and chlordanes in marine fish and crustaceans are several orders of magnitude below those expected to result in effects on seal reproduction. Based on the NOAEC for rats and dogs, toxaphene levels in fish are below those associated with thyroid effects. ΣPCB levels are also below effect levels in crustaceans. However, assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in tissues from several fish species exceed the dietary NOAEC concentration for reproduction of 72 ng/g ww (Figure 6·61). These are the same as given for beluga (section 6.8.3.4.1).

The assessment for dietary intake of dieldrin, PCB, DDT, and PCDD/F is the same as given for seabirds (section 6.8.3.3).

#### 6.8.3.5.2. **Walrus**

• *Levels and intake assessment*

No studies of biological effects have been carried out in walrus. The mean ΣPCB levels range from 111-13 600 ng/g lw in blubber. The NOAEL and LOAEL for subtle neurobehavioral effects are exceeded in walrus from eastern Baffin Island (1990 ng/g lw), eastern Hudson Bay (5800-13 600 ng/g lw), northeastern Hudson Bay (220-1400 ng/g lw), and Svalbard (11 500 ng/g lw) (Annex Table 6·A18). ΣPCB levels in walrus from eastern Hudson Bay (Inukjuak) and Svalbard also exceed the NOEL for otter reproduction and mink kit survival (Figure 6·63). However, no walrus exceed ΣPCB levels associated with immunosuppression or poor reproductive success in harbour and ringed seal (Figure 6·63).

Walrus from two sites in eastern Hudson Bay (Inukjuak, Akulivik) have also been analyzed for PCDD/Fs and/or nPCBs (Annex Table 6·A19). The PCDD/F and nPCB TEQs for the samples from Akulivik combined are 22 pg/g ww in blubber (about 80% lipid). These levels are lower than the sum of PCDD/F and nPCB TEQs (69 pg TEQ/g lw) associated with immunosuppressive effects in harbour seal (Figure 6·57). The combined levels in female walrus from Inukjuak are higher, 44 pg/g ww. Only nPCBs were analyzed in male walrus from Inukjuak, but these levels (104 pg TEQ/g ww) were higher than the combined TEQs from females. These nPCB TEQ levels exceed the combined PCDD/F and nPCB TEQ levels associated with immunosuppression in harbour seals.

Based on the dietary NOAECs and LOAECs given for mink, mean levels of dieldrin, ΣDDT, and chlordanes in marine invertebrates are several orders of magnitude below those expected to result in effects on walrus reproduction. ΣPCB and PCDD/F levels are also below effect levels in invertebrates. Levels of ΣDDT, ΣPCB, and PCDD/Fs in invertebrates are also below the various environmental guidelines.

However, some walrus are known to prey on ringed seal. Mean levels of dieldrin, ΣDDT, and chlordanes in ringed seal blubber are below those expected to result in effects in walrus. Based on the NOAEC (4000 ng/g ww) for rats and dogs, toxaphene levels in ringed seal are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in ringed seal blubber from all sites exceed the dietary NOAEC concentration for reproduction of 72 ng/g ww. Mean levels of ΣDDT in ringed seal from most sites exceed both Canadian and USEPA guidelines for protecting aquatic organisms (Table 6·14, Figure 6·62). The International Joint Commission guidelines are exceeded in ringed seal from Svalbard, the southern Barents Sea, Scoresbysund, Hudson Bay, northern Baffin Island, and southwestern Ellesmere Island. For ΣPCB and dioxin-like substances expressed as TEQs, ringed seal blubber from all sites exceed all environmental guidelines for protecting aquatic wildlife (Figure 6·61).

### 6.8.3.6. **Polar bear**

• *Reproduction*

Reproductive success for ten female polar bears on Svalbard was followed by satellite telemetry (Skaare *et al.* 1994a). Their success was examined in relation to ΣPCB concentrations (sum of 22 congeners) in blood. Eight of the ten females had cubs, but no correlation was found between reproductive success and ΣPCB concentrations.

Bernhoft *et al.* (1996) found no difference in the OC levels between females available for mating that became pregnant and those that did not become pregnant. However, the sample sizes for this comparison were very small. The reproductive success of adult female polar bears at Svalbard is about 0.75, which is similar to values found in other Arctic populations (Wiig 1995). However, Wiig (1995) found a very high mortality of young polar bears at Svalbard. Polischuk *et al.* (1995) found higher levels of the lipophilic persistent contaminants in polar bear cubs-of-the-year than in

yearlings. High intake of OCs occurs at a crucial period of growth and development of the young polar bears. This could adversely influence the early development of cubs and lead to a higher mortality.

During field research conducted in the spring of 1996, researchers at the Norwegian Polar Institute and the University of Oslo captured two female siblings accompanied by their 11-year-old mother on Svalbard. During field examinations, the two siblings were both noted to have abnormal external genitalia (J. Skaare pers. comm.). Photographs were taken and examined by Dr. Marc Cattet (DVM) at the University of Saskatchewan and he concurred that the two females were either pseudo-hermaphrodites or hermaphrodites. Causes of the abnormal development are unknown, but possible causes are: a natural but rare event known as the freemartin effect; occurrence of excessive maternal androgens, likely from a tumor; or a consequence of toxic substances. Samples collected from the bears are undergoing analysis.

• *Cytochrome P450 activity*

Studies of cytochrome P450 activity in polar bears or in ursids are very recent. The cytochrome P450 enzyme system of the polar bear was characterized by Bandiera *et al*. (1995). EROD, pentoxyresorufin (PROD) and benzyloxyresorufin (BROD) activities in polar bears were found to increase with cytochrome P450 1A content, and did not correlate with cytochrome P450 2B content, suggesting that all three activities were primarily cytochrome P450 1A-mediated (Letcher *et al*. 1996).

EROD and PROD activities, hepatic microsomal P450 1A1, 1A2 and 2B1 protein levels, as well as organochlorine and $MeSO_2$-metabolite concentrations were determined in 13 adult male polar bears collected near Resolute, Northwest Territories, Canada (Letcher 1996, Letcher *et al*. 1996). Cytochrome P450 1A1, 1A2, and 2B1 were found to be induced. The mean cytochrome P450 content in polar bear liver, measured as EROD activity, was about two-fold higher than in beluga liver (White *et al*. 1994) and 10-fold higher than activity in male hooded seals (*Cystophora cristata*) (Goksøyr *et al*. 1985). Assuming that the interlaboratory activities are comparable, this means that the cytochrome P450 1A-mediated activity was elevated in polar bear. P450 1A1 protein levels were ten-fold higher than 1A2 levels.

Cytochrome P450 1A1 and 1A2 content in male polar bear hepatic microsomes was correlated with levels of planar PCBs (non-*ortho* and mono-*ortho*-substituted) as equivalents of 2,3,7,8-TCDD (TEQ). EROD activities in polar bear liver correlated with total TCDD-equivalents (TEQs) to a concentration of 350 pg/g lipid (Letcher *et al*. 1996). EROD activity remained relatively constant above this TEQ concentration. Thus, EROD activity was not significantly correlated with TEQ or ΣPCB for the data set as a whole. The non-linear response is probably due to inhibition of enzyme activity by increasing levels of organochlorines. Immunoquantitated cytochrome P450 1A protein concentrations remained highly correlated with TEQs, even at high TEQ levels, indicating that maximal induction had not occurred. Cytochrome P450 2B content in male polar bear hepatic microsomes was significantly correlated ($r^2 = 0.784$) with concentrations of total chlordane (mainly oxychlordane and nonachlor) and total *ortho*-substituted PCBs in liver.

The strong positive correlation of cytochrome P450 1A with nPCBs and mono-*ortho* PCBs in polar bear liver suggests that these compounds may be responsible for the activity. PCDD/Fs contributed only a small fraction of the total TCDD-toxic equivalents in polar bear liver. No PROD activity was found, although there was an indication of induced cytochrome P450 2B1 levels (Norstrom *et al*. 1994c).

• *Thyroid and retinol effects*

Concentrations of retinol, total and free thyroxine (T4) and total and free triiodothyronine (T3) were measured in plasma from polar bears from Svalbard (Skaare *et al*. 1994a). These concentrations were related to the plasma concentrations of organochlorines. A significant negative correlation was found between retinol and ΣPCB concentrations (sum of 22 congeners) in the plasma ($r = 0.37$, $p = 0.003$) and in subcutaneous fat ($r = 0.46$, $p = 0.0003$). A negative correlation between thyroid hormone concentrations and ΣPCB concentrations was also found, but was not statistically significant.

• *Levels and intake assessment*

Standardized ΣPCB levels in polar bears from 16 areas across the Arctic range from 2763-24 316 ng/g lw. All ΣPCB levels exceed the NOAEL and LOAEL levels found for subtle neurobehavioral effects in offspring if polar bear are as sensitive as offspring of rhesus monkeys and humans (Figure 6·63). The ΣPCB levels also exceed the NOEL for kit survival in mink in four of the areas – Svalbard, East Greenland, McClure Strait, and eastern Hudson Bay (Figure 6·63). ΣPCB levels are close to the kit survival NOEL in several other areas. The LOAEL for immunosuppression is 21 000 ng ΣPCB/g lw in rhesus monkeys. ΣPCB levels in polar bear from three areas are at or above this LOAEL – Svalbard, East Greenland, and McClure Strait.

In the ΣPCB results presented for different groups of Svalbard polar bears based on age and sex (Annex Table 6·A20) for 1990-1994, all groups exceed the NOAEL and LOAEL for offspring neurobehavioral effects, as well as the NOEL for kit survival. Some individuals in all groups over three years of age exceed the LOAEL for immunosuppression. Some individuals also exceed the levels known to be correlated with poor reproductive success in harbour seals, as well as those correlated with poor reproductive success in ringed seal and the $EC_{50}$ for reduced litter size in mink (Figure 6·63).

Levels of PCDD/Fs in Canadian polar bears sampled in 1983-1984 range from 2-23 pg TEQ/g ww in adipose tissue (58-73% lipid; Ramsay *et al*. 1992). The mean levels of PCDD/Fs, nPCBs, and mono-*ortho* PCBs in polar bear liver from bears collected near Resolute Bay, Canada in 1992-1994 were 27, 29, and 172 pg TEQ/g lw, respectively (Letcher 1996, Letcher *et al*. 1996). The combined TEQs exceed those associated with immunosuppression in harbour seal (210 pg TEQ/g lw). Mono-*ortho* PCB levels (CBs 105, 118, 156, 157) were measured in polar bears from Svalbard (Bernhoft *et al*. 1996). The levels ranged from 42-102 pg TEQ/g ww (82-256 pg TEQ/g lw). These levels are comparable to or higher than the mono-*ortho* PCB TEQs (140 pg TEQ/g lw) associated with immunosuppressive effects in harbour seal (Figure 6·57). Oehme *et al*. (1995a) analyzed PCDD/Fs and nPCBs in polar bear milk from Svalbard. The combined TEQs for both groups of substances ranged from 5.5-12.7 pg TEQ/g lw. These levels are lower than those associated with immunosuppression.

In a study by Polischuk *et al*. (1995), samples from female polar bears (Churchill, Manitoba) taken before and after a fasting period, and from cubs-of-the-year and yearling cubs were analyzed for OC levels. ΣPCB concentrations (lipid weight basis) in females with or without cubs were about twice as high after the fasting period, and in some individuals, the fasting concentrations approached the NOEL for kit survival in mink. ΣPCB concentrations in yearling cubs were above the NOAEL and LOAEL for neurobehav-

ioral effects and the NOEL for mink kit survival, and concentrations in cubs-of-the-year were highest (approximately 11000-15 000 ng/g lw), also exceeding these NOAEL, LOAEL, and NOEL levels. Thus, some polar bear populations may be at risk for the immunosuppressive effects of ΣPCB and dioxin-like substances as well as the reproductive and developmental effects of ΣPCB.

Based on mean OC levels in ringed seal blubber from various sites in the Arctic (Annex Table 6·A18), dieldrin, chlordanes (heptachlor epoxide), and ΣDDT levels are below the dietary NOAEC levels for reproductive effects in mink. Based on the NOAEC (4000 ng/g ww) for rats and dogs, toxaphene levels in ringed seal are below those associated with thyroid effects. Assuming that marine mammals are as sensitive as mink, mean levels of ΣPCB in ringed seal blubber from all sites exceed the dietary NOAEC concentration for reproduction of 72 ng/g ww. Mean ΣDDT levels in ringed seal exceed Canadian and USEPA guidelines for protecting aquatic wildlife, and in some cases (Svalbard, Barents Sea, Scoresbysund, northern Baffin Bay, Ellesmere Island, Hudson Bay), International Joint Commission guidelines are also exceeded (Figure 6·62). For ΣPCB and dioxin-like substances expressed as TEQs, ringed seal blubber from all sites exceed all environmental guidelines for protecting aquatic wildlife (Figure 6·61).

#### 6.8.3.7. Arctic fox

• *Levels and intake assessment*

No studies of biological effects have been performed on Arctic fox. Levels of OCs have only been determined for individuals on Svalbard. On a wet weight basis, OC concentrations in Arctic fox liver are similar in specimens collected in 1983-1984 and 1993-1994. Assuming a lipid content of 6%, ΣPCB levels in Arctic fox liver from 1993-1994 range from 2250-236 700 ng/g lw with means in the range 30 100-63 600 ng/g lw (Annex Table 6·A20). These mean ΣPCB levels exceed the NOAEL and LOAEL for subtle neurobehavioral effects in offspring of rhesus monkeys and humans, the NOEL for kit survival in mink, levels known to cause poor reproductive success in harbour seals, and the $EC_{50}$ for reduced litter size (Figure 6·63). Some individuals exceed the levels associated with poor reproductive success in ringed seals and the $EC_{50}$ for kit survival in mink as well (80 000-120 000 ng/g lw). The lowest-adverse-effect-level for immunosuppression is 21 000 ng ΣPCB/g lw in rhesus monkeys. Mean ΣPCB levels in Arctic fox from 1993 and 1994 are above this LOAEL. No information is available for PCDD/Fs or planar PCBs in Arctic fox. However, these can also be suspected to be at or above the levels found in polar bears.

As Arctic fox from Svalbard also eat ringed seal, the dietary intake assessment given above for polar bear is probably also valid, i.e., ΣPCB levels in ringed seal blubber from all sites exceed the dietary NOAEC for reproductive effects as well as all guidelines for protecting aquatic wildlife (Figure 6·61). Levels of ΣDDT and dioxin-like substances expressed as TEQs (Annex Table 6·A19) in Svalbard ringed seal exceed all of the guidelines for protecting aquatic wildlife (Table 6·13). Arctic fox from Svalbard are, therefore, at risk for the immunosuppressive effects of ΣPCB and probably also dioxin-like substances, as well as the reproductive and developmental effects of ΣPCB.

### 6.8.4. Summary and conclusions – biological effects

While studies of ecological relevance, such as species diversity or population size, are ongoing in the Arctic, they are not focused specifically on effects of contaminants. Therefore, at the present time, it is very difficult to link contaminant levels or biochemical indicators of effects to effects on Arctic animals at the individual or population level. Such assessments are also complicated by the fact that the thresholds for effects of many contaminants are not well known and very little is known about effects of contaminant mixtures.

As far as organochlorines are concerned, Arctic marine mammals are often regarded as controls for much more contaminated members of their populations or related species in temperate regions. However, biological effects studies on Arctic animals do show some subtle responses that may be related to current levels of OC contaminants. Based on the results of the few biological effects studies that have been carried out, the following can be concluded.

#### 6.8.4.1. Observed effects

• *Reproduction*

Studies in Canadian, Swedish, and Russian peregrine falcon still indicate that egg-shell thinning occurs due to high ΣDDT levels in the eggs. For Canadian peregrines, these levels are high enough to still be causing reproductive failure in some cases. For Norwegian white-tailed sea eagle, correlations were found between eggshell thinning and concentrations of DDE, ΣPCB, and HCB, but the degree of thinning was below that affecting reproduction. No correlations could be found between ΣPCB concentrations and reproductive success in a study of female polar bears, however, the sample size was small. There is some evidence of reduced cub survival on Svalbard, however. Imposex has been observed in invertebrates in Kongsfjorden, Svalbard and is probably due to TBT exposure.

• *Cytochrome P450 activity*

Liver enzyme induction (EROD) seems to be correlated with concentrations of ΣPCBs in burbot from the Canadian Arctic. A clear relationship has been seen between non- and mono-*ortho* PCB levels and liver enzyme induction (EROD, AHH) in starved beluga whales from the western Canadian Arctic. A relationship has also been seen between EROD and AHH activities and ΣPCB and dieldrin concentrations in ringed seals from Arviat and between EROD activity and ΣPCB concentrations in hooded seal from the West Ice. Cytochrome P450 1A activities in polar bear seem to be elevated and are correlated with concentrations of non-*ortho* and mono-*ortho* PCBs. Cytochrome P450 2B activities in polar bear liver seem to be correlated with chlordane levels.

• *Bone development*

It is still not clear if toxaphene is the cause of depressed bone hydroxyproline levels in fish from Lake Laberge.

• *Thyroid and retinol effects*

A significant negative correlation was found between retinol concentrations and ΣPCB concentrations in polar bear plasma. A similar trend was found for thyroid hormones, but was not statistically significant.

#### 6.8.4.2. Assessment of current levels in biota

Current concentrations of some OCs in several Arctic species are at or above the known thresholds associated with effects that have been seen in other species studied either in the laboratory or in the field. The current levels of DDE in the Canadian population of tundra peregrines and the fact that there has not been any improvement in eggshell quality

since the early 1980s indicate that present DDE levels are still causing effects. ΣPCB levels in Canadian, Fennoscandian and Kola Peninsula peregrines are at or exceed most NOELs and LOELs for reproductive endpoints in a wide range of wild bird species. PCDD/F and planar PCB TEQs in Kola Peninsula peregrines exceed most threshold levels for reproductive endpoints in wild birds, indicating similar risks. The Committee on the Status of Endangered Wildlife in Canada (COSEWIC) downlisted the status of *Falco peregrinus tundrius* from 'threatened' to 'vulnerable' partly because the chemical threat did not appear to be as great as it once was. The current studies suggest that this threat still exists.

High DDE levels as well as significant eggshell thinning are also still seen in Fennoscandian merlin and white-tailed sea eagle in Arctic sites. The extent of eggshell thinning is less than seen in peregrine falcons and the populations of these species are recovering. However, ΣPCB levels as well as PCDD/F and planar PCB TEQs in sea eagle exceed most threshold levels for reproductive endpoints in wild birds, indicating similar risks for this species.

Predatory seabirds, such as glaucous and herring gulls, exceed some threshold effect levels for ΣPCB, indicating that they may be at risk for subtle reproductive effects. Piscivorous seabirds such as guillemots, kittiwake, and puffin have somewhat lower ΣPCB levels, but also exceed some reproductive threshold levels.

Mustelids, particularly mink, are very sensitive to the reproductive effects of ΣPCB and dioxin-like compounds, and populations of mink and otter in the Swedish Arctic have increased as environmental levels of these compounds have declined. Current ΣPCB levels in otter from the Swedish Arctic and in mink, marten, and ermine from Canada do not put these species at risk for reproductive effects, but possible risks do exist for subtle neurobehavioral effects in offspring. For otter, the PCDD/F and planar PCB levels expressed as TEQs are in the same range as those associated with immunosuppression in harbour seals. Moderate immunosuppression may have little effect under normal conditions, but impaired immune function is thought to have played a role in the morbillivirus-induced mass mortalities of harbour seal, grey seal, and striped dolphin populations in Europe in the period 1987-1991 (Hall *et al*. 1992, Aguilar and Borrell 1994, Ross *et al*. 1995, 1996).

Harbour porpoises from the southern Barents Sea have ΣPCB levels that exceed those associated with decreased otter and mink reproduction, and with immunosuppression and poor reproductive success in harbour seal. Some walrus populations, and most notably those individuals that feed on ringed seal, have ΣPCB levels that exceed NOELs for reproductive effects in otter and mink. In at least one case (Inukjuak, Hudson Bay), nPCB TEQ levels in walrus exceed those associated with immunosuppression in harbour seals. ΣPCB levels in most walrus studied, as well as in seals, beluga, minke whale, harbour porpoise (from West Greenland), and narwhal exceed those associated with subtle neurobehavioral effects in offspring of rhesus monkeys and humans.

Biological marker studies in polar bear show significant correlations between increased liver enzyme induction and elevated planar PCB and chlordane levels, as well as between decreased retinol levels and elevated ΣPCB levels. These correlations indicate that these substances may be the cause of the biological responses. Current ΣPCB levels in polar bear from Svalbard, East Greenland, and McClure Strait indicate that these populations are at risk for reproductive and immunosuppressive effects as well as subtle neurobehavioral effects in offspring. ΣPCB levels are highest in the Svalbard population, and some groups have levels that exceed those associated with poor reproductive success in both harbour and ringed seals and decreased survival of young in mink. In support of this, some evidence exists indicating that young polar bears on Svalbard have higher mortality rates. Other populations have lower ΣPCB levels, but still exceed some threshold levels for reproductive effects as well as those associated with neurobehavioral effects in offspring. Populations on Svalbard and at Resolute Bay are also at risk for immunosuppression from current levels of PCDD/Fs and planar PCBs expressed as TEQs.

ΣPCB levels in Arctic fox from Svalbard are actually higher in some cases than in polar bear. Thus, some individuals may be at risk for reproductive and immunosuppressive effects, as well as subtle neurobehavioral effects in offspring.

Consequently, based on known thresholds for effects, several Arctic species appear to be at risk for, primarily, reproductive and/or immunosuppressive effects from current levels of ΣDDT, ΣPCB, and/or dioxin-like substances. Those at greatest risk include peregrine falcon, white-tailed sea eagle, glaucous and herring gulls, alcids, kittiwake, otter, harbour porpoise, some walrus populations, polar bears, and Arctic fox. If the risk for subtle neurobehavioral effects from ΣPCB in exposed offspring of mammals is included, then some mink populations, beluga, narwhal, minke whale, and seals are also potentially at risk. Greenland halibut and lake trout from some sites have toxaphene levels high enough to affect fry bone development and increase mortality during spawning. Some invertebrates may be at risk for the reproductive effects of TBT, particularly those that are exposed in harbors.

An assessment of risks from dietary intake has been attempted, based on results from laboratory feeding experiments, OC levels in prey items, and information on different species' food preferences. Based on low-effect-concentrations in food, reproduction in Canadian peregrine falcons is likely to be affected by levels of DDE, ΣPCB, and dieldrin in their prey. Reproduction in mustelids in Canada and Sweden may be affected by dietary levels of, primarily, PCDD/Fs and ΣPCB in freshwater fish. Reproduction in beluga, narwhal, and seals may be affected by dietary levels of ΣPCB in marine fish. Walrus, polar bear, and Arctic fox that prey on ringed seal have dietary intakes of ΣPCB that may cause reproductive effects.

When environmental quality guidelines for protecting aquatic wildlife are used, dietary intakes of ΣDDT, ΣPCB, and dioxin-like substances are problematic for many marine and freshwater piscivorous species. The same is true for predatory seabirds feeding on bird eggs and for walrus, polar bear, and Arctic fox that consume ringed seal.

The new data thus suggest that even the relatively low levels of contaminants currently present in Arctic animals may not be without biological effects, especially during years of poor feeding or during periods of fasting. When fat reserves decrease, OC concentrations in the remaining fat increase, leading to levels that may exceed known effects thresholds. Exposure of offspring to high OC levels during lactation is also problematic. The high fat content of milk in many Arctic mammals leads to a transgenerational transfer of high levels of OCs during early development, a period of the life cycle that has been shown to be particularly sensitive to disruption by some OC contaminants. A similar situation occurs in birds and fish when fat reserves are mobilized into egg production.

As is usually the case with Arctic species, the lack of experimental dosage/response data continues to limit the ability to interpret residues in Arctic animals. Food and water stress in combination with immunosuppressive chemical exposure have been shown to affect the immune system, growth,

and reproductive potential in mice (Porter *et al*. 1984). The possibility of immunosuppression in Arctic marine mammals, especially polar bears, during times of food stress is therefore real, and warrants further study.

## 6.9. Conclusions and recommendations

All POPs considered under the AMAP monitoring program have been found in air, snow, water, sediments, and/or biota in the Arctic. In some cases, a number of Arctic species have POP levels high enough to cause effects. Therefore, the authors of this chapter believe that we must continue to 1) monitor levels of POPs in the abiotic environment and in biota with emphasis on temporal and spatial trends and improved understanding of transport and fate processes, 2) refine and develop methods of determining subtle biological effects and relate POP levels to these effects, 3) improve our ability to assess biological effects in organisms and health effects in humans, and 4) continue to promote measures to reduce levels of POPs in the environment.

### 6.9.1. Levels and effects

#### 6.9.1.1. Air and precipitation

Monitoring of air in the Arctic has shown that levels of lindane and chlordane are correlated with long-range transport episodes from use areas in the mid-latitudes of North America, Europe, and Asia. Higher concentrations of PCBs are related to transport of air masses from industrialized areas of western Europe and eastern North America in the mid-latitudes. Current and past uses of OCs in the mid-latitudes of the northern hemisphere are, therefore, the most likely source of OC contaminants to the Arctic environment. Global transport from current use areas at low latitudes is also important for some OCs. Levels of PCB and DDT were up to ten times higher in snow from the Taimyr Peninsula and Laptev Sea than in the Canadian Arctic.

*Continued studies of OC levels in air, mechanisms of dry and wet deposition, and scavenging by snow are needed in order to understand and better model transport pathways, as well as to determine sources of OCs and their fate in the Arctic. The high PCB and DDT levels in Russian samples need to be confirmed.*

#### 6.9.1.2. Seawater and freshwater

Highest levels of α-HCH in the world's oceans are found in the Canada Basin and Canadian Arctic Archipelago due to a combination of icecover conditions and circulation of older water from the European Arctic. High PCB levels are found in Russian seawaters. The PCB levels in Arctic seawater exceed some guidelines for protection of freshwater aquatic life.

There is little knowledge of OC concentrations in lake waters, with the exception of studies in the Yukon River basin and the Northwest Territories in Canada and a few lakes from Taimyr Peninsula, Russia. Higher levels of HCH, DDT, and PCB were found in the Russian lakes. Concentrations of PCBs in lake waters in Canada and Russia exceeded levels associated with negative biological effects.

Exceptionally high HCH levels are found in Russian river water, especially the Ob. Ratios of γ-HCH to α-HCH indicate use of lindane.

*More research is needed on OC levels in Arctic lake waters. The high OC levels found in lake and river waters in Russia need to be verified.*

#### 6.9.1.3. Sediments and suspended solids

OC levels are higher in freshwater sediments than marine sediments. PCB levels in both freshwater and marine sediments generally do not exceed thresholds associated with biological effects. Data are lacking on OCs in marine sediments from the Canadian and Alaskan Arctic except for a limited number of samples from the southern Beaufort Sea, Cambridge Bay area, and the Bering/Chukchi Seas. TEQ levels in most freshwater, marine, and estuarine sediments exceed some guidelines for protection of aquatic life.

PCB and ΣDDT levels on suspended solids in the Ob and Yenisey Rivers are higher than found in river water near industrialized areas in North America. Although there may be unidentified quality assurance problems with the PCB and DDT data for Russian rivers, results from independent Russian and Norwegian studies of bottom surficial sediments from the Indigirka River delta and Pechora River support these results. Thus, the data for PCBs and DDT in Russian rivers, while reflecting some sample contamination problems, may in fact be extraordinarily high even compared with surface waters of urban areas of North America and western Europe. Unfortunately, there are no measurements of PCBs in the dissolved (including dissolved organic carbon-associated) phase where the major fraction would be found.

*Verification of the extent of PCB and DDT contamination of these rivers is needed.*

#### 6.9.1.4. Biota

Levels of OCs in Arctic species and environments are generally lower than in temperate areas. However, high levels do occur due to a combination of the following processes: 1) OCs are biomagnified in long food webs, particularly in ones that are dominated by organisms with high lipid content, 2) some abiotic-biotic interfaces, for example ice edges, may be particularly conducive to transferring OCs into the food web, and 3) some species, and/or their prey, have large contaminant burdens from their southern overwintering habitats.

Knowledge of effects of low tissue residue levels or body burdens of POPs or low-level intakes is very limited and is most developed for PCDD/Fs, non-*ortho* PCBs, other PCBs, and DDT. Threshold levels have mainly been established in laboratory animals for effects on reproduction, neurobehavioral development, and immunosuppression.

POP-related effects are seen in some Arctic biota. Current concentrations in several Arctic species are at or above the known thresholds associated with, primarily, reproductive, immunosuppressive, and neurobehavioral effects.

Studies of POP levels at different trophic levels in terrestrial, freshwater, and marine ecosystems confirm that considerable bioaccumulation and biomagnification occur. Bioaccumulation models exist and have been used for OCs in Canadian Arctic lakes, but these do not consider the movement or runoff of contaminants in snowmelt and runoff from the terrestrial environment. The accumulation of PCBs with age in beluga and seals has been predicted using a model that incorporates age-dependent uptake and elimination as well as transfer of contaminants in maternal milk. This model is not yet linked with bioaccumulation/biomagnification models for the marine food chains leading to marine mammals.

*The existing database should be used, together with further studies, to calibrate models and link chemical fate/distribution model output to contaminant bioaccumulation. The role of abiotic-biotic interfaces at the base of food webs needs to be better understood in both marine and freshwater environ-*

*ments. The epontic component of the food web, ice edge effects, and the role of sediments and the benthic community in the accumulation of OCs should be examined further, particularly in the marine environment.*

Fauna in the terrestrial ecosystem, excluding migratory birds and birds which prey on them, are less contaminated with OCs than in freshwater and marine systems, mainly due to shorter food webs. The major concern in the terrestrial environment is for migratory birds of prey and piscivorous mammals, which have higher OC levels. Current PCB levels put otter from the Swedish Arctic and mink, marten, and ermine from some sites in Canada at risk for subtle neurobehavioral effects in offspring. For otter, the PCDD/F and planar PCB levels expressed as TCDD TEQs are in the same range as those associated with immunosuppression.

For predatory birds, lowest OC levels are found in the non-migratory Icelandic gyrfalcon. Migratory species such as merlin, white-tailed sea eagle, and peregrine falcon have much higher ΣDDT and ΣPCB levels, reflecting accumulation of OCs at wintering grounds farther south as well as from preying on migratory birds in the Arctic.

DDE levels in the Canadian population of tundra peregrines and the lack of improvement in eggshell quality since the early 1980s indicate that present DDE levels are still causing effects. ΣPCB levels in Canadian, Fennoscandian, and Kola Peninsula peregrines are at or exceed most thresholds for reproductive effects. PCDD/F and coplanar PCB TEQs in Kola Peninsula peregrines exceed most threshold levels for reproductive endpoints in wild birds.

High DDE levels as well as significant eggshell thinning are still seen in Fennoscandian merlin and white-tailed sea eagle in Arctic sites. The extent of eggshell thinning is less than seen in peregrine falcons and the populations of these species are recovering. However, ΣPCB levels as well as PCDD/F and planar PCB TEQs in Swedish white-tailed sea eagle and in Kola Peninsula peregrine falcon exceed most threshold levels for reproductive endpoints in wild birds.

*The 'active transport' of contaminants to Arctic food chains by migrating waterfowl and other species merits more research. The recovery of migratory birds of prey in the Arctic should be followed.*

Freshwater ecosystems contain higher levels of OCs than terrestrial ecosystems, mainly due to longer and more complex food webs. Lipid weight PCB levels in freshwater fish are very similar to levels in anadromous and marine fish, with the major differences being related to trophic level. Concentrations of OCs within the benthic and pelagic invertebrate community, or in forage fish, have been examined in only a few lakes. The major concern is high OC levels, particularly toxaphene and PCB, in fish at higher trophic levels.

*Additional surveys of contaminants in the food web are needed to understand the variability of persistent OCs such as PCBs and toxaphene in freshwater fish and the mechanisms of delivery of contaminants in meltwater to the water column and bottom sediments.*

Results for TBT (tributyltin) in sediments are limited to a few harbors in northern Norway and Iceland. There is very little data on TBT levels in invertebrates or fish, and none reported for marine mammal tissues. Imposex has been observed in Arctic invertebrates in some harbors probably due to TBT exposure. Therefore, some invertebrates may be at risk for the reproductive effects of TBT.

*Surveys of TBT in harbor sediments in the Arctic should be carried out to assess the extent of TBT contamination. Where possible, the survey should include investigations of the presence of imposex characteristics in indicator species such as dogwhelk (Nucella lapillus) or whelk (Buccinium undatum). The imposex bioindicator technique for geographical scale surveys is not likely to be universally applicable in Arctic regions as the intertidal zone is often narrow (about 1-2 meters), limited in its biodiversity, and probably does not support a sufficient dogwhelk fauna for assessment sampling. However, whelks (Buccinum undatum) have a wider distribution and may be a useful indicator species (Ten Hallers-Tjabbes et al. 1994). The risk exists that TBT biomagnifies and analyses should also be made on bottom-feeding fish, waterfowl, and marine mammals, particularly as these are components of the diet of Arctic peoples.*

Other major problems in the marine environment are seen in organisms at high trophic levels. These have the highest OC levels due to long food chains. High levels are also found in detritivorous invertebrates due to relatively high levels at the base of their food webs.

Correlations between concentrations of specific POPs (PCB, dieldrin, non- and mono-*ortho* PCB, chlordane) and liver enzyme induction were found in ringed seals and starved beluga whales in the Canadian Arctic.

Greenland halibut have toxaphene levels that may be high enough to affect fry bone development and increase mortality during spawning. Piscivorous seabirds (guillemots, kittiwake, puffin) and predatory seabirds (glaucous and herring gulls) have ΣPCB levels that exceed some reproductive effects thresholds.

Harbour porpoise from the southern Barents Sea have ΣPCB levels that exceed those associated with reproductive effects and immunosuppression. Some walrus populations, most notably those where some individuals feed on ringed seal, have ΣPCB levels that exceed thresholds for reproductive effects. In at least one case (Inukjuak, Hudson Bay), TCDD TEQ levels in walrus exceed those associated with immunosuppression. ΣPCB levels in most walrus studied, as well as in harbour porpoise from West Greenland and the southern Barents Sea, seals, beluga, minke whale, and narwhal exceed those associated with subtle neurobehavioral effects in offspring.

High PCB levels were found in polar bear with highest levels in some individuals from the Svalbard population. No information is available on OC levels in polar bear from the Russian Arctic except for Wrangel Island.

Studies in polar bear show significant correlations between biological markers and levels of specific OCs. The PCB levels in some individuals of the Svalbard population may exceed those associated with poor reproductive success and decreased survival of young, which is supported by indications of higher mortality rates for young polar bear. Other populations have OC residue levels which exceed some threshold levels associated with potential risk for reproductive and immunosuppressive effects, as well as subtle neurobehavioral effects in offspring.

ΣPCB levels in Arctic fox from Svalbard are actually higher in some cases than in polar bear, exceeding thresholds for reproductive and immunosuppressive effects, as well as subtle neurobehavioral effects in offspring. It is not known if other Arctic fox populations have high OC levels, with consequent risks for effects.

Thus, peregrine falcon, white-tailed sea eagle, glaucous and herring gulls, alcids, kittiwake, otter, harbour porpoise, some walrus populations, polar bears, and Arctic fox appear to be

at greatest risk for reproductive, immunosuppressive, and/or neurobehavioral effects from current levels of ΣDDT, ΣPCB, and/or dioxin-like substances. Some mink populations, beluga, narwhal, minke whale, and seals may be at risk for subtle neurobehavioral effects from ΣPCB in exposed offspring.

Current OC levels in the prey of many top level predators may affect reproduction in these predators.

*All the species at risk should be monitored directly for contaminant levels and possible biological effects. Biological monitoring of the most heavily contaminated species should be encouraged. High trophic level predators, such as polar bear, should be studied in the Russian Arctic to determine if OC levels here are higher than in East Greenland and Svalbard.*

A large suite of biological effects indicators are available, but only a few have been determined in Arctic animals. Most of these biomarkers have been developed for fish or mammals. There are generally no biological effects data for Arctic seabirds and little laboratory data on immunosuppression in terrestrial birds or seabirds, or on other subtle effects of OC levels in young birds. Thus, biomarkers for these types of effects in birds should be developed. The lack of data on biological effects indicators, especially immunosuppression and hormonally-based disturbances in mammals at high trophic levels, is a major gap. At present, measurements of PCB congeners and other persistent OCs are seldom linked to biomarkers.

*There is thus a need for refinement and development of sensitive biochemical or physiological level assays for use in Arctic biota. Practical problems that must be overcome are the difficulty of collection of biopsy samples from live animals or fresh tissue from hunted animals in remote regions, and problems in sample preservation and storage. Linkage of the results for planar OCs to bioassays of cytochrome P450 1A1 and 2B activity on sample extracts, a technique widely used in contaminant studies in the Great Lakes, should be considered for Arctic studies in order to explain the biological activity.*

Little is known about the immunology of peregrine falcon, white-tailed sea eagle, polar bear, or Arctic fox (Svalbard), the species found to have the highest OC levels.

*A research program directed at immunology and immunosuppression in the species identified as most at risk is a high priority, given the fact that these species have the highest levels of PCBs and other OCs.*

Toxaphene, when measured, is observed to be a major OC contaminant in air, seawater, fish, pinnipeds, and cetaceans in the Canadian Arctic. There is no information available on toxaphene levels in terrestrial animals, waterfowl, seabirds, polar bear, or Arctic fox or in most other species outside of Canada. Very little information is available on the toxicity of toxaphene in animals and this is a major gap in interpreting the levels found in Arctic birds and mammals.

*Toxaphene should be included in the suite of OC analyses in continued AMAP monitoring in order to fill the existing data gaps for parts of the Arctic and for additional species.*

Polar bear contain DDT- and PCB-methylsulfone metabolites, but no studies on their possible effects on the adrenal gland have been performed. Little is known about most POPs' hormone-disrupting effects. Current risk assessments are focused primarily on DDT/PCB/PCDD/PCDF effects as threshold data are available for these. Although toxicity data are available for many of the OC pesticides, very little of this is usable for establishing threshold levels for effects in wildlife. This has made it difficult to assess the significance of current levels of other POPs found in biota.

*More research into the toxicology of the major bioaccumulated components of toxaphene, $MeSO_2$-PCB and -DDT metabolites, chlordanes, and less persistent, current use OC pesticides is required in species at risk, particularly fish and marine mammals, in order to be able to assess biological effects. Research to establish threshold data for OCs, particularly for use in studies of wildlife, should be a high priority.*

It is not clear whether animals having delayed implantation, such as mustelids, seals, and polar bear, are more vulnerable to the reproductive effects of some POPs. The high fat content of milk in many Arctic mammals leads to a transgenerational transfer of high levels of OCs during early development, a period that has been shown to be particularly sensitive to disruption by some OC contaminants. A similar situation occurs in birds and fish when fat reserves are mobilized into egg production. No studies of such detrimental effects have been performed in Arctic biota. All of these factors combine to limit our ability to conduct proper ecotoxicological risk assessments.

*Research on reproduction in key Arctic species should be done, particularly with reference to the possible sensitivity of species with delayed implantation, including mustelids, seals, and polar bear. In addition, more research into the toxicology of known OC compounds is needed in order to fill the above knowledge gaps.*

For all species living in the Arctic in which lipids play an important role as energy reserves during starvation periods, there is a risk of redistribution of lipid-soluble OCs stored in the lipids. Detrimental effects of chemical contaminants may be enhanced by environmental stresses that occur regularly or occasionally in Arctic ecosystems. In kittiwake, beluga, harp seal, and polar bear, there is evidence that nutritional status affects the distribution of OCs in the body. After periods of fasting or starvation, OC levels increase in remaining fat tissues and may redistribute to the brain or liver. It is not known how Arctic animals respond to 'below threshold' levels of contaminants which then relatively rapidly increase above thresholds. In polar bears, there is limited evidence that detrimental effects of OCs on reproduction will occur if females are stressed by starvation. Other stresses, such as occasional extreme weather, overhunting, habitat destruction, or human harassment may affect behavior, reproduction, and social structures. Physiological stress caused by toxicants may worsen the effects of such environmental stressors.

*More research into the effects of multiple stressors is required. In particular, the effects of starvation and other environmental stressors, combined with the effects of the mobilization of lipid-associated OCs, may be important.*

PBDEs, PCNs, and chlorinated paraffins have been detected in biota from Svalbard. PCDEs have been found in biota in northern Finland. PBDEs are of concern in view of expected future trends in production or release and potential toxic effects.

*Surveys should be carried out to determine the extent to which these POPs occur in the Arctic.*

Some of the less persistent chlorinated pesticides (endosulfan, methoxychlor, pentachlorophenol, trifluralin, atrazine, chlorpyrifos, and chlorothalonil) have been found in Arctic air, snow, water, and plants. These are currently not included in the AMAP monitoring program. Although they are not expected to biomagnify in food webs, they may be accumulated by plants and phytoplankton at the base of the food web (France *et al*. 1997, Chernyak *et al*. 1996). Some (e.g., atrazine) are potent inhibitors of photosynthesis while others (e.g., methoxychlor) are known to have estrogenic activity. Little is known about the behavior of these chemicals at low temperatures.

*There is a need for research on environmental behavior, levels, and effects of the less persistent OCs, which are still in use in most circumpolar countries.*

There is a basic lack of knowledge for most Arctic species in terms of population sizes, geographical extent, and natural history, which limits interpretation of contaminant levels. This is particularly the case for dietary information and trophic status of the species.

*More research into Arctic ecology is required for interpretation of the contaminant data.*

*A combination of contaminant monitoring, biomarker studies, and population monitoring may prove to be the most powerful technique for assessing the impact of contaminants in the Arctic.*

### 6.9.2. Sources

High HCH levels are seen in Russian lake water (Taimyr Peninsula) and rivers, with high ratios of γ-/α-HCH indicating fresh use of lindane. High ΣPCB and ΣDDT levels are seen in snow, rivers, seawater, coastal sediments, and the few invertebrates, fish, reindeer, lemming, seabirds, seal, and beluga sampled in Russia. Some samples have higher relative amounts of *p,p'*-DDT, indicating possible fresh releases. Although no single set of samples is large enough for statistical comparisons with biota from other Arctic countries, the results indicate higher contamination with these compounds, possibly through current use or improper disposal.

*These high levels must be verified and, if correct, actions must be taken to reduce inputs to the Arctic.*

Significant PCB contamination is observed in the immediate vicinity and within a 20 km radius of abandoned and recently active military radar (DEW line) sites in the Canadian Arctic. PCB levels in nearshore areas of Cambridge Bay show that PCBs can be transported from dump sites to the immediate marine environment. Transfer to the terrestrial and aquatic food webs near these sites has been demonstrated. PCB contamination has also been demonstrated in marine sediments outside of landfills on Svalbard. Military and civilian sites and dump sites that contain significant amounts of electrical equipment in other circumpolar countries may have PCB-contaminated soils, and terrestrial and marine mammals and birds could be contaminated because of feeding, even infrequently, on plants or animals at these locations. Local PCDD/F contamination in the vicinity of a smelter near Kirkenes, Norway has also been demonstrated.

*Surveys of other such sites within the Arctic should be made to determine the circumpolar scope of the problem and remedial actions taken where high levels of contamination are found.*

Numerous local sources of other POPs also exist, but have not been studied. The roles these sources play in loadings to the Arctic are not known. These include burning of wastes in Arctic communities, smelter emissions of PCDD/Fs, industrial and community effluents, dumps, waste sites, etc.

*Surveys of local sources of contamination by POPs within the Arctic are needed to quantify the emissions and leakage. Where emissions are high, actions should be taken to reduce or remediate them.*

### 6.9.3. Budget modeling results

Mass balance modeling of HCHs suggests that the Arctic Ocean is in steady state with respect to γ-HCH inputs and is exporting α-HCH.

Although the modeling results for toxaphene are highly uncertain, they suggest that inputs and outputs to the Arctic Ocean are roughly in balance. Assessment of water concentrations of toxaphene and HCHs suggests that they are trapped under the polar cap and are thus 'ghosts of the past' and will be slowly drained, largely through the Canadian Archipelago, on a time scale of decades.

Mass balance modeling of PCBs, although confounded by high uncertainties for many input parameters, suggests that inputs exceed outputs by a factor of 1.5 and that up to 24% of inputs may be coming from Russian rivers. Knowledge of inputs of OCs via rivers is very limited, both geographically and temporally. Lack of information on seasonal and year-to-year variation of OCs, particularly PCB, DDT and toxaphene, in these rivers constitutes a major knowledge gap.

*More detailed measurements of riverine inputs should be performed, especially in the Eurasian Arctic. The measurements of OCs in rivers must include both the dissolved and particulate phases, and be conducted on at least a monthly basis in the largest rivers, for example, the Ob, Yenisey, Lena, Mackenzie, and Pechora, in order to more accurately estimate inputs to the Arctic Ocean.*

In seawater, little is known about concentrations of most OCs in offshore areas within the Arctic Ocean and adjacent seas, especially for PCBs and toxaphene. Profiles of these contaminants with depth, which would allow estimation of the total mass of these chemicals in the ocean, are lacking. The general lack of information on OCs in the Norwegian coastal current and Greenland current is especially critical because of the importance of these currents for inputs to and outputs from the Arctic Ocean. Knowledge of airborne OCs is much more detailed, however, only a limited number of paired air and water measurements, which are required to assess the direction of gaseous flux, are available.

*In order to make accurate estimates of loadings to the Arctic Ocean, levels of HCH, toxaphene, and PCB in seawater should be determined at key locations and in ice and snow-cover above it, as well as in paired air and water samples.*

There is no information on fluxes of OCs to marine sediment for any area of the Arctic Ocean. Knowledge of losses via sedimentation and burial are critical to make accurate budgets of PCBs and may also be important for some toxaphene components (e.g., nonachlorobornanes).

*Sediment cores should be collected at key locations in the Arctic Ocean and analyzed (including dating) in order to calculate these fluxes.*

The behavior of OCs in snow is a major knowledge gap. Uptake and release of vapor-phase OCs, especially HCHs and toxaphene, by snow and ice is expected to be significant, but was not included in the budget calculation because of uncertainties over the ultimate fate of the sorbed chemicals. Much of the contaminants in snow may be revolatilized prior to melt and then deposited into the water column by gas absorption rather than in meltwater.

*Research on the key processes of revolatilization and gas absorption of OCs by snow is required to develop more accurate models of chemical fate and OC delivery to marine and freshwater systems.*

### 6.9.4. Spatial trends

The most extensive spatial coverage of OC measurements in biota is found for polar bear, caribou/reindeer, ringed seal, seabird eggs, and Arctic char, but none of this sampling is truly circumpolar. Most spatial trends that have been identified are in Canada, and are no doubt due to the large land areas that differ in ecological characteristics, distances from pollution sources, and atmospheric deposition patterns. Generally, west to east increases in PCB and DDT levels are seen in Canadian caribou, ringed seal, harp seal and polar bear. This trend appears to extend to Greenland and Svalbard polar bear and possibly also to harp and ringed seal from Svalbard, northern Norway, and/or western Russia. HCH levels are higher in Canadian caribou, thick-billed murre eggs, and ringed seals than from sites farther east (Svalbard, northern Norway, and western Russia). No trends could be seen in the Arctic char data.

Results of PCB and DDT analyses in Arctic seabirds indicate that the Barents Sea may be more contaminated than the Canadian High Arctic. There is no information on OC contaminants levels in seabirds from the Alaskan Arctic or the Chukchi/East Siberian Seas with which to assess the geographic extent of this trend.

Complete circumpolar coverage of contaminants in biota is lacking, particularly for Alaska and the Russian Arctic. Spatial coverage of lake sediments (both surface grab sediments and dated sediment cores) is very limited, especially in Russia, Alaska, Greenland, Iceland, and the High Arctic Islands of Canada, Norway, and Russia. Species coverage for cetaceans is limited. There is much less information on contaminant levels, and very limited geographic coverage, for marine fish and invertebrates. The lack of circumpolar data limits our ability to understand sources, transport pathways, and mechanisms for focusing contaminants.

*There is a need for standardized measurement programs, particularly in biota, adopted on the advice of experts. To at least complete a general picture of the magnitude of contaminant levels on a circumpolar basis, properly sampled, complementary data for the best studied matrices are needed from Alaskan and Russian sites. An alternative approach would be to carry out new spatial studies, properly designed and coordinated for simultaneous sampling at key circumpolar sites. We believe that internationally organized studies, with one scientist or one scientific organization responsible for coordination on a circumpolar scale, have the greatest chance of producing meaningful results. Such a study is demonstrated for polar bears.*

Some data indicate that levels of some OCs in biota on and around Svalbard, the southern Barents Sea, and eastern Greenland may be higher than in biota measured in other areas. Factors which influence the input of OCs to the Barents Sea are: 1) meteorological conditions, including atmospheric depressions moving northeast along the northern low-pressure belt meeting with cold Arctic air in the Barents Sea region, 2) hydrographical conditions, including warm Atlantic water meeting with cold Arctic water and forming the Polar Front, and 3) the movement and melting of sea ice, a possible carrier of contaminants both deposited from the atmosphere and associated with particles incorporated into the ice in other areas. Riverine sources in western Russia may also play a role. The transport of contaminants either in ice and overlying snow or associated with sediment particles embedded in sea ice, and subsequent melting in marginal ice areas, has been suggested as a significant pathway for focusing contaminants from a wide area of the Arctic into these areas.

*Additional studies of oceanographic and atmospheric circulation are needed to increase our knowledge about the physical processes that deliver contaminants to the Arctic. High priority should be given to studying the role of sea ice as a pathway for focusing POPs, particularly at marginal ice areas around Svalbard and in the Barents Sea.*

### 6.9.5. Temporal trends

The accurate monitoring of temporal trends in contaminant levels in both the abiotic and biotic environments is necessary in order to determine the correlation between contaminant inputs and risks to biota in an area. It must also be known whether remedial actions to reduce inputs of contaminants are effective in reducing environmental contamination levels. Temporal trends can also provide warning signals if contamination levels increase, or if levels do not change over time in response to remedial actions. In addition, the verification of many models depends on informative time trend measurements.

A nine-fold decline in concentrations of ΣHCH in Arctic air, based on measurements in the Bering/Chukchi Seas and at several locations in the Canadian Arctic Archipelago, has been observed. However, in the European Arctic at Svalbard, α-HCH concentrations have only declined two-fold and γ-HCH concentrations appear to have increased during the period 1984 to 1992. This may possibly be due to regional differences in inputs of HCH isomers.

*Priority for future air monitoring should be to examine temporal trends at key locations, especially where data are available from past monitoring, for example, at Ny-Ålesund (Svalbard) and at Alert (Canada). Also, additional air monitoring in the Bering/Chukchi Seas would be useful as this is where the longest time series for HCH and other OCs exist.*

Studies of freshwater sediment cores show recent declines in PCDD/F deposition following major increases after the 1940s. Sediment cores from lakes in the North American High Arctic show later onsets of ΣPCB inputs (in the 1950s) compared with cores from subarctic and mid-latitude lakes, which is in agreement with the global fractionation model. There are no analyses of sediment cores from the Eurasian High Arctic (e.g., Svalbard or Bear Island) which could confirm whether observations in the North American High Arctic reflect a generalized circumpolar phenomenon.

Retrospective time trends derived from a snow core from the Agassiz Ice Cap, Ellesmere Island, Canada showed no significant changes in OC levels with time. This may be due to different processes involved in scavenging and deposition of contaminants in snow and sediments, but this is currently not understood. Interpretation of profiles of POPs in snow

and sediment cores can be problematic where melting or mixing has occurred. Although the time trends did not concur, fluxes for PCB were found to be similar for freshwater sediments and the ice core.

*OC levels should be determined in peat cores from ombrotrophic bogs (bogs formed by rain), to confirm deposition fluxes of OCs predicted from snow and sediment cores. The analysis of additional dated sediment and/or glacial snow cores would be particularly useful in the High Arctic (e.g., Svalbard, Taimyr Peninsula, Franz Joseph Land) for inferring present and past inputs where peat cores may not be available.*

Long-term (greater than 15 years), standardized, temporal trend data for the Arctic, based on annually collected, well-defined samples, are only available for fish and reindeer from the Swedish Arctic and subarctic (up to 29 years), and, based on longer intervals between sampling, for seabird eggs in the Canadian Arctic (18-20 years). Based on these time series, PCB, PCDD/F, and DDT levels in biota declined between the 1970s and the 1980s, after many POPs were restricted or banned. Based on the more precise studies in Sweden, PCBs, DDT, PCDD/F, HCH, and HCB levels have declined in biota in recent years. The decline seems to be slower for PCB. This may indicate continued low-level leakage of PCB to the environment from unknown or poorly studied sources. From the Canadian seabird data, it is not clear whether this decline continued from the 1980s to the 1990s. OC levels did not decline in peregrine falcon between the 1980s and 1990s.

There is insufficient information at present to discern temporal trends in POP levels in marine mammals from Svalbard, northern Norway, or Russia. Although limited OC data are available for marine mammals from the European Arctic during the 1970s and 1980s, there is insufficient information on key covariates such as the age, sex, or season of collection to make reliable comparisons. Similarly, there are problems with comparability of samples of polar bear tissues collected in the 1970s and early 1980s in the Canadian Arctic, which has made assessment of temporal trends problematic. At present there are no long-term data on temporal trends in polar bears from Svalbard.

Temporal trend data from other Arctic biota are very limited for most OCs, because most are based on two, or at most three, sampling occasions, and the results are not equivocal. In many cases, sampling strategies and sampling techniques were not standardized and the data may not be comparable for the different sampling occasions.

It is difficult to evaluate time trends for the High Arctic region since properly designed monitoring programs have generally not been performed with this as an objective. At present, there is a lack of well designed temporal trend programs monitoring biota, which could be used to establish trends of POPs in key species throughout the Arctic. It is not clear whether temporal trends in the High Arctic may be synchronized with those at more southern latitudes, or whether declines in the Arctic are slower, as predicted by the 'cold condensation' model.

*In view of the uncertainty as to whether or not OC levels are decreasing in the High Arctic, it is essential that temporal trends be monitored there in appropriate abiotic and biotic media at a few key locations. Existing trend series must be reevaluated for their potential to provide the information needed in the future. If programs are sufficiently powerful, they should be continued. Programs generating poor series should be discontinued, and new ones started. If new series are started, appropriate guidelines (e.g., ICES) for setting up monitoring programs and criteria for selecting species should be used. Based on experience with previous monitoring, new time series may require 10-15 years of monitoring before temporal trends are apparent.*

Despite being banned for open use in the circumpolar countries, evidence from temporal trend studies in biota and sediment cores indicates that PCB levels are not decreasing in the Arctic as quickly as other POPs.

*Due to the threat posed by PCBs in the Arctic, it is necessary to reduce the inputs of PCB into the environment both from the circumpolar countries as well as globally. Current sources within the circumpolar countries should be identified and remediated. International action is also needed to reduce global inputs of DDT and other persistent OCs that are still in use.*

The experiences from the past 30 years of continuous monitoring of the Baltic and the northern areas of Sweden have shown that between-year variation of OC residue levels in biota is large, and individual variation within this, even larger. These observations reinforce the importance of judicious sampling and archiving programs which would allow regular monitoring of key populations and retrospective analysis for contaminants, including newly detected contaminants.

The monitoring of temporal trends should include the use of specimen banks for archiving abiotic and biotic samples. The Lake Ontario and Swedish studies of contaminant trends were possible in part because tissue banks enabled a retrospective analysis of samples using current methodology. This has been done to a lesser extent for Arctic biota, for example, the reexamination of eggshells of predatory birds, and the analysis of archived moss samples from Norway.

*Consideration should be given to the establishment of AMAP sample archive centers for various media, similar in concept to the current situation with AMAP data centers for air, marine, and freshwater data. Within AMAP countries, there is already notable specimen banking experience and knowledge (Olsson and Bignert 1997). Specimen banking started in Sweden at the beginning of the 1960s. Recently, several organizations have commenced work on specimen banking for both contaminant and biological (e.g., genetic, disease) studies. The marine mammal tissue archival program operated by the National Institute of Science and Technology (NIST) in Alaska may be another appropriate model for other countries.*

### 6.9.6. General monitoring

Many laboratories submitting analytical results used in the assessment have taken part satisfactorily in interlaboratory comparisons. However, some question remains as to possible contamination during sampling as a source of high levels of some POPs, particularly PCBs in Russian samples. In a number of cases, analyses were performed on only single samples from a few sites, limiting the usefulness of the data for comparisons. Quality assurance programs exist for air sampling of OCs, where sampling is carried out at fixed locations with well-characterized media (polyurethane foam, glass fiber filters), but are less well developed for the collection of sediments and water. Concern has been expressed that field sampling, especially from ships, results in contamination from OCs such as PCBs due to shipboard contaminants.

*Future AMAP monitoring programs for POPs should include a quality assurance program with mandatory participation of laboratories in interlaboratory comparisons for key matrices. The program could be part of other international efforts. These are currently well developed in Europe, but are not circumpolar in their coverage of analytical laboratories. Quality assurance programs for sampling strategy and sample collection should also be developed.*

Many current reference materials have relatively high levels of contaminants, which are not always appropriate for use in analysis of low level samples from the Arctic, and can result in cross-contamination during sample analysis.

*There is a need for development of 'made for the Arctic' reference materials, especially for abiotic matrices, such as sediments and plants, with low levels of POPs.*

PCDD/Fs, and non- and mono-*ortho* PCBs have been studied to a limited extent in freshwater and marine sediments and a range of Arctic biota (freshwater fish, white-tailed sea eagle, peregrine falcon, mink, otter, caribou, ringed seal, harp seal, walrus, beluga, narwhal, and polar bear). Levels of PCDD/Fs in fish, caribou/reindeer, and beluga/narwhal are low. Non-*ortho* PCBs were found in all samples that were analyzed and (along with mono-*ortho* PCBs) accounted for most of the TEQs. PCDD/F levels were higher in blubber of pinnipeds (ringed seal, walrus) than in cetaceans and accounted for a greater proportion of TEQs. Initial surveys of ringed seals and polar bears, which showed higher PCDD/F levels in the High Arctic, have not been followed up.

*It may be justifiable to limit future monitoring to non-ortho PCBs, along with mono-ortho PCBs, unless specific sources of PCDD/Fs are suspected or information is needed for human intake studies. Additional measurements of PCDD/Fs may be justified in the case of seals, where PCDD/Fs account for up to 50% of TEQs and in order to verify the higher levels seen in the High Arctic.*

PCB results were often given as sums of as few as seven to as many as 90 congeners, making comparisons difficult. Most laboratories determined at least seven CB congeners (CBs 31/28, 52, 101, 118, 138, 153, and 180). However, the sum of the above seven represented 11-65% of ΣPCB in sediments, 50% in mosses, 50-66% in biota, and only 10-30% of ΣPCB in air. The lack of more than 7-10 congeners in air samples made it difficult to use the results for source and pathway studies.

*The number of CB congeners should be standardized and future analyses should require, as a minimum, a somewhat larger number of congeners to be determined, including toxicologically important mono-ortho PCBs such as CB 105 and 118. Determination of 30-40 congeners (the exact number would have to be assessed by careful consideration of congener patterns in each matrix) should give results close to those for the sum of all possible congeners because the remaining congeners contribute relatively little to ΣPCB.*

Current methods of quantifying toxaphene may overestimate levels in some samples, such as marine mammals, and underestimate levels in others, such as air. The use of different quantification methods has limited the assessment of current atmospheric loadings of toxaphene to the Arctic Ocean and is a matter that needs to be resolved.

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

In the past, suites of species that could be used for monitoring across the Arctic were identified. Such species were common, widespread, and not endangered. They were chosen because they had similar food habitats across the entire Arctic. However, experiences with contributing scientists in integrating data for this report suggest that different species in various taxonomic groups are more easily sampled in different Arctic areas of the world. Species obviously differ in abundance and economic importance in different areas of the world. In areas of low productivity, the desired species cannot be sampled or rigorous sampling programs could actually put species or ecological communities at risk. Thus, the cost of sampling sufficient numbers of particular species in some areas may be prohibitive. Regional opportunities for sampling should also be reflected in monitoring programs.

*The 'essential' species recommended in the AMAP monitoring program should be reevaluated. We recommend that AMAP take the initiative to organize a meeting with experts that have experience in temporal and spatial monitoring to discuss and define objectives with the monitoring activities as well as to select some few optimal matrices in accordance with these objectives. The aim is to discuss the monitoring activities that should be based on annually repeated measurements. However, during the discussions, it might also be possible to disclose monitoring activities that are most effectively carried out over other time periods. It is important that the aim of the meeting is to minimize the matrices to be used so that the outcome of the meeting will be realistic to fund.*

Some life stages may reflect environmental contamination better than others. For example, experience shows that immature stages of fish have less variability in OC levels compared to mature fish, and similarly for pelagic fish when compared to benthic or littoral fish (Olsson *et al.* 1997). Egg tissue from nesting birds, such as some alcidae that lay single eggs, have proven to have low variation compared to other bird tissue (Bignert *et al.* 1995). Egg tissue from nesting birds is indicative of levels in adults, with levels in eggs and female parent increasing as the parent ages. Similarly, OC levels in many species vary with age, season, and, in the case of females, with reproductive status. Thus, ancillary data on each sample is essential for proper interpretation.

*Biota should be sampled at a life stage that integrates contaminant exposure. Also, the same tissue type should be analyzed in a given species or sample type (e.g., muscle tissue in fish) in order to make comparisons possible. Lipid amounts should always be reported. Other covariates, such as sex, age, size, condition, reproductive status, and time of year should also be determined and reported.*

In the marine environment, joint cruises, such as those conducted by Norway/Russia, Russia/USA, Canada/USA, and Germany, have been critical to the development of information on POPs in seawater, ice, sediments, and some biota (e.g., invertebrates).

In the terrestrial environment, recent collaborations between Russian and USA scientists have developed a significant amount of information on metal contamination in plants on the Kola Peninsula and near Norilsk. Similar collaborations should be encouraged for future measurements of POPs in plants and other terrestrial fauna.

*Mechanisms should be found to continue to fund such cooperative projects.*

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# Annex

## Contents