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Arctic REGIONAL REPORT

Regionally Based Assessment of Persistent

Substances

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UNITED NATIONS ENVIRONMENT PROGRAMME



CHEMICALS

Regionally Based Assessment of Persistent Toxic Substances

Canada, Denmark, Finland, Iceland, Norway, Russian Federation, Sweden, United States of America (Alaska)

ARCTIC REGIONAL REPORT

DECEMBER 2002



GLOBAL ENVIRONMENT FACILITY

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PREFACE

The production of this report was made possible through a contract between the Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway and Global Change Strategies International, Inc. (GCSI), Ottawa, Canada. The responsible authority for this contract was Mr. Lars Otto Reiersen, Director of AMAP. The author of this report and project manager for this study was Dr. Hans Martin, Senior Associate, GCSI.

In 2000, The United Nations Environmental Program asked AMAP to participate in a global assessment of PTSs, and in particular, to produce a report on PTSs in the Arctic. This document is intended to meet that request. The report is one of twelve, which make up the global assessment.

The author is grateful to the reviewers Dr. Derek Muir and Ms. Cynthia de Witt for their critiques and advice as well as to Mr. Simon Wilson for his technical assistance.

Hans Martin, Ph. D. Global Change Strategies International Ottawa, Canada November 2002

EXECUTIVE SUMMARY

Introduction

In the late 1980's, the eight circum-polar countries launched a major multi-year research program, the Arctic Monitoring and Assessment Programme, AMAP, to evaluate and document the occurrence and consequences of persistent toxic substances (PTSs) and other pollutants in the Arctic. By far the largest discussion was that concerned with PTSs. In 1998, the AMAP secretariat published its first comprehensive report, the "AMAP Assessment Report: Arctic Pollution Issues", referenced as the AAR.

The present UNEP assessment of PTSs in the Arctic is based on the AAR. The pertinent material in the AAR has been extensively edited to meet the guidelines for the UNEP regional reports.

Conclusions

In comparison with most other areas of the world, the Arctic remains a clean environment. However, for some pollutants, combinations of different factors give rise to concern in certain ecosystems and for some human populations. These circumstances sometimes occur on a local scale, but in some cases may be regional or circumpolar in extent.

Contaminant sources and pathways

Knowledge of sources of contamination of the Arctic is improving and in some cases, the information is quantified. The pattern that is emerging is of two major types of source –sources remote from the Arctic and sources found within the Arctic.

Sources

- Outside of the Arctic, sources exist for a number of the persistent organic pollutants (POPs). The main contaminants of concern are: organochlorine pesticides (e.g., HCHs) and their metabolites from agricultural activities/practices; industrial chemicals (e.g., PCBs); and anthropogenic and natural combustion products, e.g. chlorinated dioxins/furans (see Annex I of the report). These outside sources are discussed in the UNEP reports from other regions.
- Over much of the Arctic, the levels of POPs cannot be related to known use and/or releases from potential sources within the Arctic and can only be explained by long-range transport from lower latitudes.
- Of the heavy metal contamination in the Arctic, industrial sources in Europe and North America account for up to one-third of the deposition, with maximum input in winter.
- Regulatory actions in Europe and North America are reducing the sources of some POPs and heavy metals.
- PCBs from decommissioned DEW (Distant Early Warning) Line sites in Canada, and dioxins/furans from smelters in Norway are examples of identified sources of POPs within the Arctic; other such sources probably exist but are presently unknown.
- Two-thirds of heavy metals in air in the High Arctic originate from industrial activities on the Kola Peninsula, the Norilsk industrial complex, the Urals (outside the Arctic) and the Pechora Basin.
- At point sources such as mine sites, heavy metals may exceed local background concentrations at distances up to 30 km from the site.
- Mineralization of geological formations provides significant, non-anthropogenic local inputs of heavy metals.

Pathways

The Arctic is a focus for major atmospheric, riverine, and marine pathways, which result in the long-range transport of contaminants into and within the Arctic. The Arctic is, therefore, a potential contaminant storage reservoir and/or sink. Various processes remove these contaminants from the atmosphere, oceans and rivers and make them available to plants and animals. Food chains are the major biological pathways for selective uptake, transfer, and sometimes magnification of contaminants by Arctic plants and animals, many of which are subsequently consumed by Arctic peoples.

- Strong south to north air flows, particularly over west Eurasia in winter, transport contaminants from lower latitudes. Special mechanisms selectively favour the accumulation of PCBs and certain pesticides in the Arctic.
- Arctic rivers are a significant pathway for contaminant transport to the Arctic, often associated with extreme seasonal fluctuations due to freeze-up and melt water flushing characteristics. Suspended solids carry high levels of PCBs and DDT in the Ob and Yenisey river deltas, as do sediments in the Indigirka and Pechora rivers.
- Ocean waters are a major storage reservoir and transport medium for water-soluble POPs. Sea ice may be important in transporting POPs and other contaminants from coastal sediments during the winter and from deposition from the atmosphere, with subsequent redistribution during ice melt.

In marine, freshwater and terrestrial ecosystems, contaminants are selectively taken up by microorganisms and higher plants from water, sediment and soils. Consumption by herbivores and carnivores results in the transfer of contaminants and in some cases increased concentrations (biomagnification), within the food webs. Food web structure and length of the food chain, therefore, significantly influence the transfer and redistribution of contaminants within the Arctic.

- Freshwater and marine ecosystems contain higher levels of POPs than terrestrial ecosystems due to longer and more complex food webs. Biomagnification of POPs is especially significant in food webs dominated by organisms with high fat contents. Many upper trophic level carnivores are long-lived and may transfer POPs to offspring during extended gestation and lactation.
- In several marine mammals, geographical differences in contamination, e.g. mercury contamination, may be explained by differences in geology, diet, and growth processes related to temperature. Biomagnification of metals is often very selective, e.g., there is no indication that lead levels increase in higher trophic levels although mercury levels clearly do.
- Some species and/or their prey contain large metal and POP burdens from over wintering at lower latitudes and deliver these to the Arctic on their return in the summer.

The combination of long-range transport processes, climate conditions and physical, chemical and biological properties results in the accumulation of some contaminants in traditional foods at levels often exceeding those in foods from outside of the Arctic.

Contamination levels, trends and effects

Low temperatures, extreme seasonal variations in light and lack of nutrients are some of the physical and chemical characteristics that cause environmental stress to organisms, limit productivity of Arctic ecosystems and make them potentially more vulnerable to environmental contaminants.

There is considerable variability among species in their exposure and response to different contaminants and their rate of recovery from the effects of exposure. Apart from areas of intense local contamination, the major concern at present is focused on PCBs and pesticides (see Annex I of the report), mainly because of the sensitivity of species to these contaminants and the biological processes, which enhance levels and effects.

- The most exposed animals to many contaminants are those high in the food webs, such as marine mammals, including polar bears, and birds of prey, but also some fish species.
- Contaminant levels in some Arctic birds and mammals exceed some thresholds associated with reproductive, immunosuppressive, and neurobehavioral effects in laboratory animals and some studied wildlife species. Besides eggshell thinning in some Arctic predatory birds from DDE, other subtle biological effects have been seen in a few studied Arctic mammal species. These effects appear to be associated with high levels of POPs, particularly PCBs.
- Biomagnification is a major factor influencing species exposure, with the long, marine-based food webs being particularly vulnerable. In contrast, migratory birds are vulnerable through over wintering in polluted environments at mid-latitudes and/or from consumption of other contaminated migratory birds.
- Based on a few dated sediment core studies and long-term temporal trend monitoring in fish and seabird eggs, levels of PCBs and DDT decreased in the sub-arctic from the 1970s to the 1980s. However, trends

for the 1980s to 1990s are less obvious and more difficult to interpret. Long-term data on time trends in the High Arctic are lacking.

• Mercury seems to be increasing in aquatic sediments and in marine mammals. It is biomagnified but its effects appear to be suppressed by current levels of selenium.

Geographical areas of concern

Contaminants are widely, but not uniformly, distributed around the Arctic. Geographical variation in levels results from point sources of contamination, which result in high local pollution concentrations, and from environmental convergence mechanisms, e.g., convergence of physical pathways or areas of sediment accretion. Geographical variation in sensitivity for effects, results from environmental condition that make similar concentrations bio-available in one area but not in another, and, among humans, variations in production, harvesting and utilization of traditional foods.

- Industries on the Kola Peninsula, Norilsk, and eastern Finnmark emit a wide spectrum of major local pollutants, resulting in strong spatial gradients along atmospheric, terrestrial, riverine and marine pathways. Effects can be locally catastrophic and sub regionally damaging, e.g., areas adjacent to nickel smelters.
- PCBs and DDT levels in suspended solids in the Ob and Yenisey river deltas and sediments in the Indigirka and Pechora rivers are high, even compared to urban areas in temperate regions.
- Levels of PCBs and DDT seem to be higher in both biotic and abiotic media around Svalbard, the southern Barents Sea and eastern Greenland than in the Canadian High Arctic. Levels of HCHs seem to be higher in the Canadian Arctic. Causes and mechanisms in focusing these and similar important contaminants are not fully understood. Other such regions may exist, but inadequate data coverage, in particular for Alaska and parts of Russia, may mean that all such areas have not yet been identified.

Human exposure

Several groups of people in the Arctic are highly exposed to environmental contaminants. Persistent contaminants, derived from long-range transport or local sources, accumulate in animals that are used as traditional foods. Thus, variation in human exposure depends on a combination of 1) varying environmental concentrations of contaminants, 2) local physical and biological pathways that make the contaminants available, and 3) the local dietary habits of the people.

- Exposure to persistent organic pollutants is the primary concern. People are most exposed to PCBs and certain pesticides through the long marine food webs, which result in high concentrations in mammals, birds and, to a lesser extent, fish. The use of different foods determines contaminant intake. Some indigenous groups are exposed to levels that exceed established tolerable intake levels. Transfer to infants can result in newborn levels that are 2-10 times higher than in regions further south.
- Of the heavy metals, mercury tends to accumulate in the long marine food webs. Methylmercury, partly because it is fat-soluble, is efficiently taken up following consumption and therefore poses the main potential risk. Like POPs, methylmercury can be transferred to the fetus and to breast-fed children, and in certain areas, levels are high enough to indicate a need for public health measures. Although mercury levels can be high, interaction with selenium may reduce the risk to people.
- Controls on emissions have resulted in measurable reductions in input of some contaminants (e.g., lead and possibly PCBs and DDT). There is considerable variation across the Arctic, however, and recycling of accumulated pools of long-lived contaminants can result in continued exposure long after controls have been enforced.

Potential threats

- Production and use of 'new' organic chemicals, including new generation pesticides.
- Increased emissions of heavy metals and other elements or compounds from increased development of industries within the Arctic and developing regions outside the Arctic (e.g., Southeast Asia).
- Release of POPs through accidents during production, transport, waste disposal, and storage, including existing dumps (e.g., leakage from landfill sites).

- Natural events, e.g., floods, storms, volcanic eruptions, climate warming, and earthquakes, which can release, mobilize, or redistribute contaminants.
- Unexpected interactions between contaminants or between a contaminant and particular environmental conditions, may significantly change contaminant mobility through food webs.
- Accidental releases, for which the extreme environmental conditions and isolated localities in much of the Arctic greatly increase the difficulties of detection and taking remedial measures.

Gaps in current understanding

Current understanding of transport processes and the ability to quantify them is inadequate. In particular, determination of transport processes and their relative importance or magnitude within and between compartments (air, land, water, ice, sediments and biota) is essential. The prioritization of Data Gaps is highlighted in Annex I of the report. Specific gaps and needs concern:

- Contaminant inputs to the Arctic from various sources and pathways, including increased knowledge of local sources within the Arctic, which may as yet be unknown or insufficiently quantified.
- Poor understanding of pathways of transport and deposition of heavy metals and POPs, from land to rivers, estuaries, deltas and the continental shelf. In particular, determining contaminant focusing zones (i.e., zones of convergence of contaminant transport pathways) and understanding the processes of sequestration by sediments need further attention. The use of natural and anthropogenic tracers to mimic contaminants and distinguish sources has been underutilized.
- Ocean transport processes for different contaminants, including ice transport and subsequent contaminant release in melting (focusing) zones.
- Improved understanding of the influence of Arctic conditions, especially temperature and light, on the transformation and fate of contaminants.
- Understanding of the changes in contaminant concentrations, transformations, and interactions that occur within food web pathways, including dynamics of the transfer of radionuclides into traditional foods arising from both terrestrial and freshwater pathways.
- Information on contaminant levels and trends, which is still lacking for certain contaminants and media in certain areas.
- Better understanding of physiological and toxicological effects of contaminants on humans and species identified as most at risk, especially on development of offspring, and/or immunosuppression and endocrine disrupting properties
- Detailed information on the diet and food consumption patterns of specific Arctic populations, including necessary information on other factors (e.g., smoking) which can influence contaminant exposures, to allow better estimates of dietary intakes of contaminants and permit more reliable estimates of associated risks.
- Integration of physical and biological models with information on environmental measurements of sources and pathways, to aid the design and implementation of monitoring, research, and management, including mitigation.
- Knowledge about combined effects of contaminants on biota and humans, both at the individual and ecosystem level.
- Knowledge about combined effects between climate change and contaminant pathways, including improvements of models for assessments. Existing models on climate change and transport processes do not have the resolution and accuracy needed to fully assess environmental consequences of anthropogenic emissions to the Arctic.

Recommendations

Human health

Weighing the well-known benefits of breast milk and traditional food against the suspected but not yet fully understood effects of contaminants, it is recommended that:

- Consumption of traditional food continues, with recognition that there is a need for dietary advice to Arctic peoples so they can make informed choices concerning the foods they eat.
- Breast-feeding should continue to be promoted.

To ensure the interest and active involvement of Arctic indigenous peoples and other Arctic residents, the Arctic countries should:

- Improve the use of indigenous knowledge in environmental research, including local participation, and policy.
- Establish a long-term communication program to provide public information concerning environmental contaminants, linked to AMAP, which gives access to sound and regularly updated information in an understandable language.
- Integrate contamination issues for different educational levels in order to raise general environmental and scientific literacy among Arctic residents, including indigenous peoples.

Source – receptor relationship

To develop international strategies to protect the Arctic from environmental contamination, the input to and the significance of the different pathways to the Arctic must be better quantified:

- Procedures for source apportionment of contaminants need to be further developed to better identify the magnitude and relative contribution over time from natural and anthropogenic sources.
- Procedures for the improved quantification and reporting on anthropogenic emissions need to be developed to better quantify inputs to the Arctic.
- Procedures need to be developed to identify the fraction of contaminants entering the Arctic as the result of current usage or recent emissions.
- The significant transport processes distributing contaminants within the Arctic need to be quantified; in particular, the contaminant transformations and interactions within the food web pathway need to be better understood and, if possible, quantified.
- Further development of existing models (atmospheric and oceanographic) to simulate/predict the transport of and exposure from contaminants to and within the Arctic and their use together with appropriate analytical tools (such as Geographical Information Systems), is required to better define action plans and priorities (e.g., emission controls, critical loads, dietary advice, etc.).

Contaminant levels, trends and effects

There exists uncertainty as to whether or not the levels of some environmental contaminants are decreasing. It is essential that temporal trends be intensively monitored in appropriate abiotic and biotic media at a few key locations, and occasionally over wider areas. Such programs necessarily imply a long-term monitoring commitment. In this context, the next phase of AMAP should promote the design and establishment of a coordinated circumpolar network of long-term reference monitoring sites to include:

- Establishment of additional air monitoring master stations to fill geographical gaps and complement existing sites.
- Continuation of existing time trend series which have proven to be useful and informative, while replacing those that have generated less useful data with more appropriate monitoring strategies.
- The use of retrospective time trend techniques (e.g., soil, sediment and ice-core studies, analysis of specimen bank samples).

• The use of specimen banks for archiving abiotic and biotic samples.

Processes behind trends for heavy metals should be studied to resolve the relative impacts of significant natural or anthropogenic sources. There is a need to obtain a spatial distribution of the magnitude of contaminant levels on a circumpolar basis. Priority should be given to:

- Significant data gaps, particularly from the United States and Russian sites.
- Mercury and POPs in organisms for which there are concerns for biological effects.

Chemical and biological effect monitoring should be encouraged in Arctic species having body burdens of POPs and mercury levels at or above levels of concern.

There is a need for improved information on spatial and temporal trends to clarify the adverse effects of POPs and methylmercury on human populations, especially on child development. The relative importance of local and distant, and natural and anthropogenic sources of heavy metals in the Arctic that are causing elevated dietary exposures should be determined.

Surveys of tributyltin (TBT) in harbor sediments in the Arctic should be carried out to assess the extent of TBT contamination.

To ensure inter comparability, future AMAP monitoring programs should continue to address and include improved quality assurance/quality control protocols, possibly linked to other international programs, for sampling and analysis, including inter-laboratory comparison; storage and archiving of samples; and handling, reporting and analysis of data.

Remedial actions relating to contaminants

The Arctic countries should take all necessary steps to ensure that their domestic responsibilities and arrangements to reduce contaminant inputs to the Arctic region are fully implemented. If these responsibilities and arrangements are not addressed in an appropriate manner, the justification for recommending actions aimed at reducing transboundary contaminants with sources outside of the Arctic will be accordingly diminished. There is a need for actions to clean up contaminants from industrial and military sites.

The LRTAP protocol for heavy metals can only address emissions to the atmosphere from anthropogenic sources. Where there are cases of transboundary effects in the Arctic resulting from releases to the aquatic or terrestrial environment, AMAP countries should explore other appropriate mechanisms to address these concerns, including other legal mechanisms. AMAP countries which are party to other international agreements aiming at reductions in releases to the environment of heavy metals and POPs should strongly support implementation plans of those agreements where these actions will lead to improvements in the Arctic environment (e.g., the London Dumping Convention, the International Maritime Organization's MARPOL Convention, the Oslo-Paris Convention for the North East Atlantic Ocean, etc.)

Levels of many contaminants in the Arctic are likely to remain at, or close to, existing levels for decades because of their resistance to degradation, the slow rate of degradative processes, and the recycling of existing accumulations. Thus, ameliorative actions to reduce exposure to humans and to protect wildlife are an essential adjunct to emission controls.

1. INTRODUCTION

1.1. SCOPE OF THE ARCTIC REGIONAL ASSESSMENT

1.1.1. Existing assessments

There has been only one definitive assessment of the Arctic to date. That assessment is described here.

In the late 1980's, the eight circum-polar countries launched a major multi-year research program, the Arctic Monitoring and Assessment Programme, AMAP, to evaluate and document the occurrence and consequences of persistent toxic substances (PTSs) and other pollutants in the Arctic. The other pollutants were those associated with the radioactivity, acidification, petroleum hydrocarbon contamination, climate change and stratospheric ozone depletion. By far the largest discussion was that concerned with PTSs. The programme is on going. In 1998, the AMAP secretariat published its first comprehensive report, the "AMAP Assessment Report: Arctic Pollution Issues", referenced herein as AAR (AMAP Assessment Report).

The present UNEP assessment of PTSs in the Arctic is based on the AAR. The pertinent material in the AAR has been extensively edited to meet the guidelines for the UNEP regional reports.

Throughout the preparation of this report, the material has been reviewed and approved by members of the AMAP coordinating committee, which has acted in place of the Regional Committees and the Regional Experts in the other regions.

1.1.2. Inter-regional links and collaboration

The AAR does not include a detailed discussion of 'Regional Capacity And Need To Manage PTSs'. Consequently, Chapter 5 of this report is limited to some general comments and recommendations. This approach is appropriate since the majority of PTSs in the Arctic originate in the highly-populated, industrialized regions to the south and, for the most part, national administrative and regulatory centers, which manage Arctic pollution issues, are not located in the Arctic but rather in the neighbouring regions to the south. Therefore, the discussion of capacity and needs to manage PTSs will be covered in detail as follows; for Canada and Alaska by Region II (North America), for Denmark, Norway, Sweden, and Finland by Region III (Europe) and Russia by Regions III and VII (Central and North East Asia). The one exception is Iceland, which is entirely in the Arctic region. For Iceland, the 'Regional Capacity And Need To Manage PTSs' will be discussed here only.

During the preparation of chapters on 'Regional Capacity And Need To Manage PTSs', as well as for many other topics, active inter-regional discussions and collaboration were maintained in order to insure a coherent set of regional reports.

1.1.3. Omissions/weaknesses

The time available for the production of the Arctic regional reports is short. Consequently, the gathering of additional data for the UNEP assessment has not been done. This Arctic report is based primarily on material published prior to 1997, which was assembled and analyzed before the publication of the AAR in 1998. There are a few exceptions. Certain sections of the AAR have been updated in the interim. These updates have been incorporated into the report where appropriate. An official update of the AAR will be released in 2002.

The lists of chemicals for each region are not the same. As a result, some corrective strategies for individual chemicals in one region may not reflect abatement needs in the other regions. For the Arctic, the specific differences between the UNEP basic list of chemicals and the AMAP list are discussed in Section 1.3 below.

1.2. METHODOLOGY

Because the assessment of the Arctic region is advanced relative to the other eleven regions, the preparation of the Arctic report has mainly involved editing and updating material in the AAR. The procedure did not including some of the activities in the other regions, such as, striking regional committees, holding organizational meetings, gathering data and undertaking analyses. Given the difference in approach, it is appropriate that a brief description of AMAP be included here.

1.2.1. Arctic Monitoring and Assessment Programme: Organizational Background

Up until the mid-1980s, international cooperation on environmental protection in the Arctic was relatively poorly developed. It took the form of a number of largely uncoordinated national initiatives and loosely structured

bilateral and multilateral arrangements focusing on specific sub-regions (Young 1995). In the late 1980s, a number of events occurred which radically changed this situation. Most significant among these were the developments in the political climate with respect to the former Soviet Union. This event, amongst other things, lead to an expansion of environmental cooperation, as exemplified by the 1987 Gorbachev 'Murmansk Initiative'. The First Arctic Ministerial Conference was held in Rovaniemi, Finland, June 1991. This conference represented a breakthrough in the development of international cooperation for the protection of the Arctic, with its most significant outcome being the adoption of the Arctic Environmental Protection Strategy (AEPS 1991b). To implement the AEPS, five programs were instituted, including the Arctic Monitoring and Assessment Programme (AMAP), which had the responsibilities to monitor the levels of, and assess the effects of, anthropogenic pollutants in all compartments of the Arctic environment, including humans.

1.2.2. The development of AMAP and its activities from 1991 to 1996

During its first period (1991-1996), AMAP implemented a monitoring programme. In addition, at an expert meeting in March 1994 (AMAP 1994), the strategy for the preparation of the AMAP assessment reports was developed. It was agreed that two reports presenting the work during the first phase of AMAP would be prepared:

- The 'AMAP Assessment Report: Arctic Pollution Issues' (AAR) comprising a fully referenced, comprehensive, technical and scientifically presented assessment and
- The Arctic Pollution Issues: A State of the Arctic Environment Report (SOAER) a more concise report including an executive summary with recommendations specifically addressed to Ministers.

The AAR is the document used here.

1.3. GENERAL DEFINITIONS OF CHEMICALS

Persistent toxic substances include two main groups of pollutants, persistent organic pollutants (POPs) and organometallics. POPs are separated into three subgroups, pesticides, industrial compounds and unintended byproducts.

One compound, hexachlorobenzene, belongs to all three groups, pesticides (fungicide), industrial compounds (by-product) and unintended by-products.

1.3.1. Pesticides

Dieldrin and Aldrin

Dieldrin was mainly used as a soil insecticide. It is manufactured and is also a degradation product of aldrin. Dieldrin 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.

Chlordane and Heptachlor

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 -chlordane, 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). 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.

DDT (Dichlorodiphenyltrichloroethane)

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).

Toxaphene (Polychlorobornanes and camphenes)

Toxaphene is produced by the chlorination of technical camphene or X -pinene and can consist of over 300 congeners, mainly bornanes and camphenes substituted with 6-10 chlorines, with an average composition of C10H10Cl8. 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.

Mirex

Mirex is of interest because of its high Kow and its persistence. It was used as an insecticide and fire retardant. 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 (VPL = 4.76 Pa; H = 52 Pa m³/mol) capable of undergoing long-range transport. Its presence in the Arctic at low levels is consistent with its volatility and persistence.

Hexachlorobenzene

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 Kow = 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).

1.3.2. Industrial Compounds

Polychlorinated biphenyls (PCBs)

PCBs were introduced in 1929 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).

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. As with other POPs, the molecular weights and Kow of PCB congeners are correlated with their Henry's Law constant, vapour 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 dibenzo-furans. The nPCBs and mono-ortho CBs (CBs 105 and 118) are collectively referred to as 'planar' PCBs in this report.

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 g/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).

1.3.3. Unintended by-products

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. The incineration of halogenated sludge is also 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-tetra-chlorodibenzo- p-dioxin (2,3,7,8-TCDD) and 2,3,7,8-tetra-chlorodibenzofuran (2,3,7,8-TCDF). 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).

1.3.4. Organometals

In this report, reference is made to organometals, metals, and heavy metals. The use of these terms does not imply differences in the suite of chemicals but rather differences in their form, in particular elemental and organic forms. In the AAR the metals discussion is titled 'Heavy Metals' while in the UNEP assessment the section is titled 'Organometals'. This difference should cause no confusion. The substance of the discussions is the same.

In the AAR, the major focus is placed on Hg, Pb, Cd, and Se. In the case of the current UNEP Assessment, the focus is on the organic forms of mercury, lead and tin. In the AAR, the assessment does not segregate information into separate categories of elemental, inorganic or organic species and occasionally includes a larger group of metals than the three specified by UNEP. Consequently, the following discussion of metals is somewhat broader than required.

1.3.4.1. <u>Mercury</u>

Background concentrations of Hg are generally low, except in the immediate vicinity of mining sites, etc. Mercury occurs naturally as elemental Hg, and as organic and inorganic compounds (e.g., Hg vapor, Hg salts, short- and long-chain alkylmercury compounds). This speciation of Hg is of great importance in relation to uptake from soil and water, as the different chemical forms differ greatly in their physico-chemical properties. Much of the Hg in the environment is unavailable to organisms, as it is strongly bound to sediment or organic material. Inorganic forms can be methylated by microorganisms and transformed to methylmercury, which is much more readily taken up and accumulated in both aquatic and terrestrial organisms.

1.3.4.2. <u>Lead</u>

Lead in the environment is strongly absorbed onto sediment and soil particles, and is therefore largely biologically unavailable. Many of the inorganic salts formed (Pb oxides and sulfides) are not readily soluble in water and are sequestered in sediments. In aquatic systems, uptake is influenced by various environmental factors such as temperature, salinity, pH, and the presence of organic matter. Among all the chemical species, Pb (II) is considered to be most readily absorbed by biota.

1.3.5. Regional Specifics

1.3.5.1. <u>POPs</u>

The UNEP assessment allows for adjustments to the list of chemicals to be reported in the regional reports. The adjustments are based on regional priorities and data available. In the case of the Arctic region, one priority chemical is not included, endrin, and one group of chemicals has been added, hexachlorocyclohexanes (HCH).

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) is the most biologically active insecticidal isomer. It is used mainly in seed treatment (Bidleman et al. 1989). Technical HCH is used 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 Kow = 3.8) and short half-life in biota (Niimi 1987). Relatively high H's and VPL'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.

1.3.5.2. <u>Organometals</u>

The AMAP study does not assess tin in detail. In the UNEP assessment, tin is recommended. This report will reflect the AMAP list of metals thus providing only limited commentary on tin.

1.4. DEFINITION OF THE ARCTIC REGION

The vast region of the Arctic extends across northern North America, northern Europe and northern Asia, taking in eight countries and the expanses of sea and ocean in between. Its area is about 33 10^6 km². The terrestrial, freshwater and marine environments throughout this area exhibit considerable variation in climate, meteorology and physical geography.

The Arctic is often delimited by the Arctic Circle (66°32'N), which approximates the southern boundary of the midnight sun. Such a definition, however, is simplistic, given variations in temperature, presence of mountain ranges, distribution of large bodies of water, and differences in permafrost occurrence. Outlined below are some definitions of the Arctic region, which take into account physical, geographical and/or ecological characteristics. Following these, the Arctic region, as defined for the purposes of the AMAP assessment, is discussed.

Climatic Boundaries

Based on temperature, the Arctic is defined as the area north of the 10°C July isotherm, i.e., north of the region that has a mean July temperature of 10°C (Linell and Tedrow 1981, Stonehouse 1989, Woo and Gregor 1992). This isotherm encloses the Arctic Ocean, Greenland, Svalbard, most of Iceland and the northern coasts and islands of Russia, Canada and Alaska (Stonehouse 1989, European Climate Support Network and National Meteorological Services 1995). Another geographical indicator of the Arctic region that is partially determined by climate is the presence of permafrost (Barry and Ives 1974).

Vegetation boundaries

A floristic boundary used to delimit the terrestrial Arctic is the treeline (Linell and Tedrow 1981). Simply defined, the treeline is the northern limit beyond which trees do not grow. The treeline roughly coincides with the 10°C July isotherm (Stonehouse 1989, Woo and Gregor 1992). However, in some areas, the treeline lies 100200 km south of the isotherm, adding western Alaska and the western Aleutians to the Arctic region (Stonehouse 1989).

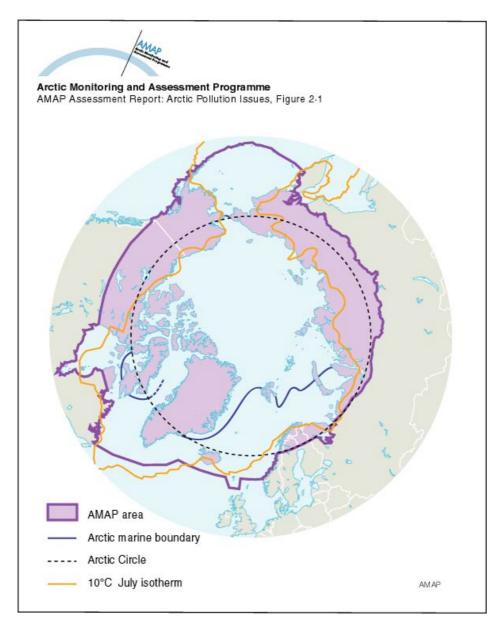
Marine boundary

Based on oceanographic characteristics, the marine boundary of the Arctic is situated along the convergence of cool, less saline surface waters from the Arctic Ocean and warmer, saltier waters from oceans to the south.

Geographical coverage of the AMAP assessment

Given the different definitions of the Arctic, based on physical geographical characteristics as described above, and those based on political and administrative considerations within different countries, no simple delineation of the Arctic region was applicable for the purposes of the AMAP assessment. To establish a geographical context for the AMAP assessment, therefore, a regional extent was defined based on a compromise among various definitions, as shown in Figure 1. This compromise incorporates elements of the Arctic Circle, political boundaries, vegetation boundaries, permafrost limits, and major oceanographic features.

Figure 1. The Arctic as defined by temperature (after Stonehouse 1989), and the Arctic marine boundary, also showing the boundary of the AMAP assessment area



1.5. PHYSICAL SETTING

1.5.1. Physical/geographical description of the terrestrial Arctic

The vast Arctic terrestrial landscape, covering an area of approximately $13.4 \ 10^6 \ \text{km}^2$ within the AMAP boundary, is very diverse, with large tracks of land covered by glacial ice. Glaciers are large masses of ice that flow under their own weight. They form where the mean winter snowfall exceeds the mean summer melting. Melting, refreezing and pressure gradually transform the snow into ice.

1.5.1.1. General geographical descriptions

Greenland, often described as the largest island in the world, is actually comprised of numerous mountainous islands almost entirely covered with a permanent ice cap up to 3000 m thick (Stonehouse 1989, Bjerregaard 1995). It covers an area of some 2 186 000 km 2.

Iceland is located south of the Arctic Circle (66°32'N). This mountainous and volcanically active island lies on the mid-Atlantic ridge. Its average height is approximately 500 meters above sea level. One quarter of the country is less than 200 m above sea level. It has an area of 103 000 km 2, with 11% of its surface covered by glaciers and more than 50% of its land surface unvegetated (Stonehouse 1989, Ministry for the Environment 1992).

The Faeroe Islands, with a total area of 1399 km 2, are located 430 km southeast of Iceland. The terrain is mountainous with an average elevation of 300 m.

Svalbard and Franz Josef Land are Arctic archipelagos of 63 000 and 10 000 km 2, respectively. These mountainous islands, and others lying to the north of Eurasia, are about 90% covered by ice.

The Fennoscandian Arctic area covers roughly 300 000 km 2, but most of this area is sub-arctic due to the warming influence of the Gulf Stream extension (Stonehouse 1989, Encyclopedia Britannica 1990).

The Kola Peninsula (ca. 145 000 km 2) on the Russian mainland is also sub-arctic and contains many lakes. Permafrost is absent, except for sporadic occurrences at the tip of the peninsula, and the coasts are ice-free (Ives 1974, Luzin et al. 1994).

The entire area of Arctic Russia within the AMAP boundary is approximately 5.5 10 6 km 2. The numerous islands of the Russian Arctic cover an area of 135 500 km 2. The largest of these is Novaya Zemlya, an archipelago with two main islands. The northern island has as area of 48 000 km 2 area. The southern island is smaller, 33 000 km 2.

Alaska's Arctic, according to the AMAP definition, extends over an area of 1.4 10 6 km 2, and is dominated by rugged mountain ranges that stretch across the state in the south and north, reaching a maximum height of 6194 m at Mount McKinley.

The Canadian Arctic landscape covers an area of approximately 4 10 6 km 2, comprised of the northern Canadian mainland in the south and the Arctic Archipelago to the north.

1.5.1.2. <u>Geology and physiography</u>

Greenland, and a vast region of the Canadian Arctic, from the Atlantic Ocean in the east to Great Bear Lake and Great Slave Lake in the west, is underlain by the Canadian Shield. This Precambrian, crystalline rock mass is exposed in some areas and covered by glacial deposits and thin soil in others. The Canadian Shield extends northward to include Baffin Island. The remaining islands in the Canadian Arctic Archipelago are primarily made up of Paleozoic and Mesozoic sedimentary rocks. Along the southwest coast of Hudson Bay are the Hudson Lowlands, comprised mainly of Lower Paleozoic rock and covered by Quaternary sediments. To the west of the Shield are the Interior Plains made up of Devonian and Cretaceous sedimentary origin, extend along western Canada and Alaska. In Alaska, these mountains merge to the north with the Arctic Foothills, which descend still farther north into a coastal plain (Linell and Tedrow 1981, Prowse 1990, Natural Resources Canada 1994).

Iceland has been created by volcanic activity along the mid-Atlantic ridge during the last 20 million years. New volcanic rock is constantly being added and about one tenth of Iceland is covered by lava deposited since the last ice age (Einarsson 1980). The Aleutian Islands to the west of Alaska are also volcanic.

Northern Fennoscandia and the Kola Peninsula are comprised of ancient, crystalline rocks forming the Baltic Shield. East of this region, from the White Sea to the Ural Mountains, lies the East European Plain, made up of sedimentary rock covered by a deep layer of glacial drift. The Ural Mountain complex, which includes Novaya Zemlya, is a region of folded Paleozoic bedrock, covered thinly with glacial deposits. The other Russian Arctic islands are also primarily formed by sedimentary formations of the Paleozoic Era. The West Siberian Lowland, comprised of till-covered sedimentary rocks, extends from the Urals to the Yenisey River. From here, eastward to the Lena River, is the Central Siberian Plateau. The Anabar Shield and peripheral sedimentary rocks underlie this till-covered region. To the north, the Taimyr Peninsula contains a folded mountain complex of sedimentary rocks, overlain by shallow soils. The region east of the Lena River is similarly comprised of folded sedimentary mountains, with some volcanic rocks. Glacial drift is discontinuous in this region (Linell and Tedrow 1981). Low plateaus and plains are characteristic of the region through which the Lena River passes and of the northern margin of Siberia from the Taimyr Peninsula to the Kolyma River (Sachs and Strelkov 1961 in Linell and Tedrow 1981).

1.5.1.3. Permafrost and soils

Permafrost, or perennially frozen ground, is defined as material that stays at or below 0°C for at least two consecutive summers (Woo and Gregor 1992). It may consist of soil, bedrock or organic matter. Spaces within the ground material may be filled with ice in the form of ice lenses, veins, layers and wedges. When very little or no ice is contained in the frozen substrate, this is referred to as dry permafrost (Linell and Tedrow 1981). Permafrost may reach depths of 600 - 1000 m in the coldest areas of the Arctic (Stonehouse 1989).

Permafrost influences soil development in the north. In general, Arctic soils are either poorly drained and underlain by solid, ice-rich permafrost, or well drained and situated over dry permafrost. Poorly drained soils are found in 85-90% of the Low Arctic and in the few wet meadows of the High Arctic. Well drained soils are

common in the extensive, sparsely vegetated areas of the High Arctic, and are scattered throughout the Low Arctic in areas where water can escape, such as on steep slopes and beach ridges. Oxidation processes in these drier soils result in lower organic matter content compared to wetter soils (Rieger 1974, Everett et al. 1981).

During the summer, the upper layer of soil in the Arctic thaws and is termed the active layer. Its depth varies according to temperature, ground material, soil moisture content and plant cover, ranging from as little as several centimeters in far northern wet meadows to as deep as a few meters in warmer, more southern, dry areas with coarse-grained soils (Ives 1974). Soil-forming processes are largely restricted to the active layer, which is unstable due to frost action during repeated freezing and thawing. Frost action results in characteristic surface features, such as frost scars, stone circles, mud circles, solifluction lobes and stone stripes (Rieger 1974, Stonehouse 1989).

1.5.2. Climate and meteorology

Low air temperatures characterize the polar areas because these areas receive, on an annual basis, less solar radiation than other parts of the world. However, the radiation levels vary greatly depending on the season. In the winter months, there is a total lack of incoming solar radiation, while in the summer, the poles receive higher levels of solar radiation than any other place on Earth.

The annual amount of solar radiation received is less than that which is lost to space by long-wave radiation, since a large part of the solar radiation that reaches the Earth is reflected by extensive cloud, snow and ice cover. This radiation imbalance produces low temperatures and results in a redistribution of heat from southern latitudes via air and ocean currents (Varjo and Tietze 1987). This energy regime is the fundamental factor driving the Arctic climate. The effect of macro-scale topography of the Earth's surface, in particular the distribution of land, sea and mountains, is important for regional and local climatic conditions in the Arctic. The frequency and the preferred tracks of the persistent Pacific and Atlantic low-pressure systems and the position of the persistent high-pressure systems not only play an important role in the existence of the regional and local climates in the Arctic, but also link the Arctic climatic system to the world climatic system.

1.5.2.1. <u>Air temperature</u>

Climate conditions in the Arctic are divided into maritime and continental subtypes.

A maritime climate is characteristic of Iceland, the Norwegian coast, and the adjoining parts of Russia. These areas have moderate, stormy winters. The summers are cloudy, but mild with mean temperatures of about 10°C. The average winter temperatures are -2° C to 1°C in the Icelandic lowland, -2° C at Bodø on the Norwegian coast, and -11° C at Murmansk on the Russia coast (Barry and Chorley 1992, EEA 1996). A maritime climate is also found directly along the Alaskan coast. This zone is very narrow because of the mountains of the Alaska and Coastal Ranges. Winters here are moderate and the summers are mild, but cloudy. The average temperature at Anchorage ranges from -5° C in winter to 10° C in summer.

Continental climate is found in the interior of the Arctic from northern Scandinavia toward Siberia, and from eastern Alaska toward the Canadian Arctic Archipelago, with much lower precipitation and significant differences between summer and winter conditions (July means of 5 - 10° C; January means of -20° to -40° C) (Prik 1959). Over the ice-covered Arctic Ocean, both the ice and the underlying sea have a regulating effect on temperature. The minimum air temperature is moderated by heat conducted from seawater below the ice. Generally, in the central Arctic, the average temperature is between -30° and -35° C in winter and between 0° and 2° C in summer.

1.5.2.2. Ocean temperature

As in the atmosphere, there is both annual and inter-annual variability in ocean temperature. This is most pronounced in the warmer, water masses. Variability in cold Arctic waters is small, but important. In the North Atlantic, the ocean appears to alternate between warm and cold states. The length of these states may vary, but fluctuations with periods of 3 - 5 years are most frequent. The varying temperature condition in the western North Atlantic is opposite to that in the eastern North Atlantic. This means that when the Barents Sea is in a warm state, the coast of Newfoundland is in a cold state (Sundby pers. comm.). This feature is also found in the distribution of ice. When there are heavy sea ice conditions in the northeast Atlantic, there is little ice in the Labrador Sea, and vice versa (Gloersen et al. 1992). There is also large variability in ice distribution in the Bering Sea, but at present, there is no evidence that this is linked to the variability in the Atlantic.

The temperature state of the ocean appears to be closely linked to atmospheric circulation, with a positive feedback mechanism existing between the atmospheric and oceanic circulations. It appears that high atmospheric pressure is associated with low temperatures in the ocean while low pressure is related to a warmer ocean.

Changes in ocean climate influence transport mechanisms and ice cover. In warm years, there is an increased transport of warm water masses to the Arctic, resulting in decreased ice cover. In cold years, transport of warm water to the Arctic is reduced and sea-ice coverage is greater (Ikeda 1990a, 1990b, Aadlandsvik and Loeng 1991).

1.5.2.3. Precipitation

The total annual precipitation in the Arctic is generally less than 500 mm and typically between 200 and 400 mm (Loshchicov 1965). Along the Arctic coast, the precipitation is higher, and over the central Polar Basin, it is lower. Cold air contains less moisture; therefore, although the frequency of precipitation may be high, the overall intensity is low. This explains why the total accumulation of snow is relatively low in winter over much of the Arctic.

The spatial precipitation pattern in the Arctic can be explained in terms of the effects of elevation changes and proximity to maritime sources of moisture. Precipitation levels decline with increasing distance inland from the oceans, and in general, levels decrease in a west-east direction across the continents in the direction of movement of most low-pressure systems.

The lowest precipitation levels on land are approximately 140 mm/y, occurring in eastern Siberia, northern Canada and Greenland. In general, total precipitation increases to above 600 mm/y from these areas toward the Atlantic and Pacific Oceans (Sugden 1982).

Maritime areas in the sub-arctic have much higher precipitation. In southern Iceland, the annual precipitation ranges from below 800 mm to over 3000 mm, and on mountains and glaciers, it can exceed 4000 mm (Einarsson 1984). The precipitation decreases toward the east and north, with 700 - 2000 mm/y at Bodø to less than 400 mm/y at Murmansk, Russia and Longyearbyen, Svalbard (EEA 1996).

1.5.2.4. Cloud cover

An important climatic feature of the Arctic is the presence of persistent and extensive stratus cloud cover over the polar oceans. The cloud cover occurs in well-defined layers separated by clear interstices. The structure of the clouds is related to the large-scale transport of relatively mild, humid air into the Arctic Basin, the boundary layer turbulence, and the optical properties of the liquid water droplets. Clouds are formed largely during the summer season; the cloud cover varies from a summer amount of 70 - 90% to 40 - 60% in winter (Landsberg 1970).

During periods of cold air outbreaks from the Arctic Basin, clouds are formed by cooling of the boundary layer air previously at higher temperatures because of the relatively warm sea surface underneath. Characteristic cloud strips following the wind direction cover large areas of the open sea.

1.5.2.5. <u>Fog</u>

A characteristic feature of Arctic weather is fog. Parts of the Arctic are extremely foggy due to the juxtaposition of cold air overlying warmer ocean waters in some areas and warm air overlying cold ice in others. In some areas, it is typical to have more than 100 days per year with fog (SCOR 1979).

In summer, the ice retreats northward, exposing open water, and warm air moves in over the ice and cold water. In August, sublimating ice and condensing water form thick fog fields that envelop the marginal ice zones, with peaks in relative humidity over water. In winter 'sea smoke' or steam fog forms over open water leads in the pack ice (SCOR 1979).

1.5.2.6. <u>Wind</u>

Winds are particularly important for the Arctic surface environment, as they can greatly augment the chilling effect of low temperatures. The generally open landscape of the Arctic region means that winds are not greatly slowed by friction at the ground level (Sugden 1982). Wind is an important factor in snow distribution, causing scouring in exposed areas and deposition in sheltered locations (Killingtveit and Sand 1991).

In the marine environment, wind affects sea surface stability and increases mixing in the water column. It also influences ice drift (Vinje 1976) and the formation of polynyas. Winds, or more precisely differences in air pressure which cause winds, are often closely related to ocean circulation as discussed by Ikeda (1990b) and Aadlandsvik and Loeng (1991).

Surface winds are greatly affected by the presence of temperature inversions. In Greenland, katabatic winds are formed as air in the dense, cold inversion layer flows down the slopes of the ice sheet toward the coast under the

influence of gravity. These winds are especially well developed in winter when the air comes into contact with the ice surface and is chilled. With the exception of these parts of Greenland, wind speeds are generally slowed by temperature inversions as the air is effectively isolated from faster moving air currents above (Sugden 1982).

Two dominant air currents in the Arctic are associated with cold air flowing in winter from the high-pressure zone over northern Siberia to the Pacific, and air flowing northwest from the high-pressure area over the Canadian Arctic toward the low pressure over the Atlantic. These winds result in very severe climatic conditions in the Arctic, and in Canada, these conditions persist into the early summer (Sugden 1982).

1.5.3. Arctic freshwater environments

1.5.3.1. Rainfall and snow

Short summers and, therefore, short periods with rainfall, characterize the Arctic. The remaining precipitation falls as snow, which accumulates as snowpack over the winter. Snowpack duration, away from the moderating influences of coastal climates, ranges from about 180 days to more than 260 days (Grigoriev and Sokolov 1994). High levels of solar radiation reaching northern latitudes in spring result in rapid snowmelt. Spring runoff comprises 80 - 90% of the yearly total, and lasts only two to three weeks (Linell and Tedrow 1981, Rydén 1981, Marsh 1990). Infiltration of this flush of water into the ground is constrained by the permafrost. Thus, spring meltwater may flow over land and enter rivers, or accumulate into the many muskegs, ponds and lakes characteristic of low-lying tundra areas (van Everdingen 1990). Summer sources of water include late or perennial snow patches, glaciers, rain, melting of permafrost, and groundwater discharge (Rydén 1981, van Everdingen 1990).

1.5.3.2. Groundwater

Groundwater levels and distribution within the Arctic are greatly influenced by permafrost. Permafrost affects the amount of physical space in which groundwater can be held and the movement of water within drainage systems. There are three general types of groundwater: supra-permafrost, intra-permafrost and sub-permafrost. Supra-permafrost water lies above the relatively impermeable permafrost table in the active layer during summer, and year-round under lakes and rivers that do not freeze. Intra-permafrost water resides in unfrozen sections within the permafrost, such as tunnels called 'taliks', located under alluvial flood plains and under drained or shallow lakes and swamps. Sub-permafrost water is located beneath the permafrost table and its depth below the surface depends on the thickness of the permafrost. In this latter case, the permafrost acts as a relatively impermeable upper barrier. These three types of aquifers, which may be located in bedrock or in unconsolidated deposits, may interconnect with each other or with surface water (Mackay and Løken 1974, van Everdingen 1990).

Generally, low levels of annual precipitation in the Arctic restrict the recharge of groundwater. In addition, infiltration of water to aquifers is restricted by permafrost year-round and by the frozen active layer for up to ten months of the year. Frozen substrate does not entirely prevent water from seeping through to aquifers, but slows the rate of infiltration by one or more orders of magnitude compared to unfrozen ground (van Everdingen 1990).

Groundwater is quite extensive in the Arctic. For example, approximately two thirds of the Yukon in the Canadian Arctic is underlain by aquifers (Hardisty et al. 1991). The largest groundwater aquifers in Iceland have been mapped in Elíasson (1994). These are generally found in highly permeable lava. Groundwater represents an important source of water in some Arctic countries.

1.5.3.3. <u>Wetlands</u>

Wetlands and saturated soils are characteristic features of the Arctic since moisture received from rain and snowmelt is retained in the active layer above the permafrost barrier. Due to the higher levels of precipitation received at lower latitudes, wetlands are more common in the Low Arctic than the High Arctic. In general, wetlands are sparsely distributed throughout the Arctic, but tend to have significant local concentrations.

Arctic wetlands are distinct due to the unique climatic conditions under which they were formed. Permafrost underlies almost all Arctic wetlands.

1.5.3.4. Wetland occurrence

In the Canadian Arctic, approximately 35% of the land area is covered by wetlands. Areas of high occurrence (between 20 and 75% coverage) include the Yukon coastal plain, the Mackenzie Delta, and parts of the Arctic islands (National Wetlands Working Group 1988).

Mires are uncommon in Greenland, and in Alaska, they are restricted to the northern mountains. In Norway, only 0.9% of the Arctic region is classified as wetland and bog area. Sweden reports 21% of its Arctic land area as mire (CAFF 1994). In Finland, approximately half of the original wetland area has been disturbed for forestry, leaving about 15% of the country covered by wetlands (Keltikangas et al. 1986). In the Russian Arctic, large areas are dominated by bogs and marshes including the Kola Peninsula, West Siberia, the Yamal Peninsula and the lowlands of the Yana, Indigirka, and Kolyma River basins (Plancenter Ltd. 1991, Bliss and Matveyeva 1992).

1.5.3.5. <u>Rivers</u>

Two main types of rivers occur in the Arctic, those that have headwaters within the Arctic and those with headwaters farther south (Woo 1992). Most of the large Arctic rivers begin their flow south of the Arctic, including the major rivers of Siberia (Ob, Yenisey, and Lena) and the Mackenzie River in Canada (Mackay and Løken 1974).

Flow in Arctic rivers is largely influenced by rain, snowmelt and ice melt because of the drainage barrier of the permafrost and the limited water storage capacity of the thin active layer (Newbury 1974 in Woo 1992). River flow is also affected by lake/reservoir storage and by groundwater input (e.g., the Lena River in winter) (Gordeev and Sidorov 1993). Most Arctic rivers exhibit an Arctic nival regime, meaning that their main flow takes place during the period of spring melt. When the melt is completed, water levels in the river drop to base flow only. Some northern rivers display a proglacial regime, flowing throughout the summer due to input from glacial melt. Rivers flowing through wetlands follow a wetland or muskeg regime, with the main flow occurring with the spring snowmelt when the ground is still frozen (Woo 1992).

1.5.3.6. Lakes

The eastern and central North American and western Eurasian Arctic regions were covered with glaciers during the Pleistocene epoch. The glaciation of eastern Asia was less extensive. These large glaciers carved out the land as they moved over it, gouging out topsoil and broken rock. The many depressions left behind filled with water when the glaciers melted, forming lakes.

Following the retreat of the continental glaciers, the land that had been pushed downward by the weight of the ice, began to rise, a process called isostatic rebound. This rebound is still occurring today, and though it is at a much slower rate than when it began, eastern Hudson Bay, for example, continues to rise at a rate of one meter per century. Thus, new lakes are still being formed as gouged land rises up out of the sea. Lakes closer to sea level are often younger than those at higher elevations (Welch and Legault 1986).

Other types of natural lakes exist in the Arctic. Kettle lakes result from the thaw of buried glacier ice (Washburn 1979). Thermokarst lakes are formed in depressions that result from permafrost melting. Ice-dammed lakes are more common in the Arctic than elsewhere and in Greenland all the larger lakes are of this type. These lakes are prone to periodic draining (Mackay and Løken 1974).

There are innumerable small lakes in the Arctic. In the Province of Murmansk in Russia, there are over 100 000 lakes, the largest of which is Lake Imandra with an area of 812 km² and a maximum depth of 67 m (NEFCO 1995). Iceland, Sweden and Finland have approximately 2.7, 5.2 and 5.8%, respectively, of their land area occupied by lakes. Only about 0.5% of Alaska is covered by freshwater (CAFF 1994). The Canadian Arctic contains many lakes, including the large Great Bear (31 326 km²) and Great Slave (28 568 km²) Lakes located on the mainland (Mackay and Løken 1974). In the central Canadian Arctic, 20% of the land surface is covered by water.

1.5.4. Arctic marine environment

1.5.4.1. <u>Geographical area and bathymetry</u>

The Arctic Basin dominates the circumpolar Arctic region. The Arctic marine area within the AMAP boundary includes the Arctic Ocean, the adjacent shelf seas (Beaufort, Chukchi, East Siberian, Laptev, Kara, and Barents Seas), the Nordic Seas (Greenland, Norwegian, and Iceland Seas), the Labrador Sea, Baffin Bay, Hudson Bay, the Canadian Arctic Archipelago and the Bering Sea. This represents an area of approximately 20 10^6 km². The connection with the shallow Bering Sea (and the Pacific Ocean) occurs through the narrow Bering Strait, while the main connection with the Atlantic Ocean is via the deep Fram Strait and the Nordic Seas.

The Arctic Ocean is divided into two deep basins, the Eurasian and the Canadian, by the transpolar Lomonosov Ridge. The Canadian Basin is transected into the Makarov and Canada Basins by a ridge in the north, the Alpha

Cordillera, and reaches depths of more than 3500 m. The continental shelf is narrow off most of Arctic North America, extending only 50 - 100 km from the coast, except in the southeastern Beaufort Sea, where it reaches some 150 km offshore (French and Slaymaker 1993).

The Eurasian Basin is smaller but deeper than the Canadian Basin, reaching depths of 4000 m. It is bisected by the narrow Nansen Cordillera into the Amundsen and Nansen Basins. North of Siberia, the continental shelf is vast and extends up to 900 km from the coast (Sugden 1982, Macdonald and Bewers 1996).

1.5.4.2. <u>Hydrographic conditions in the Arctic</u>

Ocean temperatures within the AMAP area show large variation depending on latitude and the influence of warm Atlantic and Pacific water. In the Arctic Ocean, there are only small variations in temperature between winter and summer. Due to ice coverage, the temperature in this area is close to the freezing point year-round. In the shelf areas, surface water temperatures in winter are close to freezing (just below -1° C), while during summer they may increase to 4 - 5°C due to heating from the sun. In areas influenced by Atlantic and Pacific water, there may be greater seasonal variability, for example in the northeast Atlantic and parts of the Bering Sea. In these areas the temperature remains higher than 0°C throughout the year (USSR Ministry of Defense 1980).

In general, surface water salinity in the Arctic Ocean and the adjacent shelf seas is relatively low compared to other oceans. In the Arctic Ocean itself, surface salinity varies between 30 and 33, and decreases in the area of the shelf seas to below 30. In general, the salinity is lower during summer than winter due to input of freshwater from rivers and ice melt. Close to where the large Russian rivers enter the Kara Sea and the Siberian shelf, the salinity is below 20 throughout the year and drops to as low as 10 during the summer (USSR Ministry of Defense 1980). The average total annual runoff of the Russian rivers is approximately 2100 km³ (Aagaard and Carmack 1989). In the Canadian Arctic, the influence of the Mackenzie River is evident during summer, when the salinity drops to 27 in the surface layer. Salinity in the Arctic Basin increases with depth, reaching levels between 32.5 and 34.5 at 100 m (USSR Ministry of Defense 1980).

1.5.4.3. Ocean currents

In a simplified picture, waters flowing north to the Arctic regions are comprised of warm currents originating from the Atlantic and Pacific Oceans, while cold currents flow out of the Arctic. Atlantic water enters the Arctic Ocean through Fram Strait and the Barents Sea, while Pacific water enters via Bering Strait. Water leaves the Arctic largely via Fram Strait, but also through the Canadian Arctic Archipelago (Macdonald and Bewers 1996). Most of the water in the Arctic Ocean originates from the Atlantic Ocean (79%). The inflow through the Bering Strait is very modest (19%). The main water outflow is via the East Greenland Current (75%) and the outflow via the Canadian straits is relatively small (25%). Inflow from rivers represents only 2% and although this is a small percentage of the total, it is much higher than in other oceans (Sugden 1982).

1.5.4.4. <u>Sea ice</u>

Sea ice forms from ocean water and floats on its surface. It forms when the temperature of the sea falls below the freezing point. The freezing point is dependent on the salinity of the seawater (-1.8° C for a salinity of 33) (Doherty and Kester 1974). The extent of ice cover changes with the seasons, with an average maximum of 15 10^{6} km² in March and an average minimum of 8 X 10^{6} km² in September (Gloersen et al. 1992).

Flaw leads or coastal polynyas occur at the landfast ice border where offshore winds separate the drift ice from the pack ice. Polynyas are open water regions that persist within closed sea ice cover, ranging in area up to thousands of square kilometers. These areas often have a high rate of primary production of phytoplankton and are among the richest marine areas in the world. They represent areas of high-energy exchange between ocean and atmosphere during the winter months when the sea ice cover effectively prevents exchange between water and air. Permanent polynyas have been found near Cape Bathurst (Beaufort Sea), Baffin Bay, northeastern Greenland and a region extending east and west of the northern part of the New Siberian Islands.

1.5.5. Ecological Characteristics of the Arctic

Polar ecosystems exist under extreme environmental conditions, including cold temperatures, large seasonal fluctuations in incoming solar radiation, extensive snow and ice cover, and short growing seasons. These conditions affect the productivity, species diversity, wildlife behavior (e.g., migration), and food chain characteristics of Arctic and sub-arctic ecosystems. These effects have implications on contaminant transfer and storage in Arctic biota, and on the sensitivity of Arctic ecosystems to contaminants and other stressors. This chapter describes ecosystems of the Arctic terrestrial, freshwater, and marine environments as a background for

discussion in later chapters on contaminants and their effects in these ecosystems. It is not intended as a comprehensive coverage of the ecology of the Arctic. Animal species, which are of special interest, for example, those important in the diets of Arctic peoples, such as caribou/reindeer, some fish, and marine mammals, are described in greater detail.

1.5.5.1. <u>Physical-geographical characteristics affecting Arctic ecosystems</u>

Recent glaciations

Over the last 1.8 million years, since the start of the Pleistocene era, polar regions have undergone numerous glaciations. The most recent of these occurred approximately 20 000 years ago, during which time many Arctic and sub-arctic areas were covered with ice. Some regions of the Arctic have been deglaciated for only 3000 years whilst others are still glaciated. Due to these recent glaciations, Arctic ecosystems, in general, are relatively young compared to those farther south (Bliss 1981a, Stonehouse 1989). Despite this short time span for ecosystem development, Arctic ecosystems, including soils and biota, appear to be stable and in equilibrium with the current northern climate (Bliss 1981a).

Cold

The Arctic is synonymous with cold temperatures. In the coldest regions, air temperatures fall below -60° C in winter and reach averages of only 4°C in July (Barry and Hare 1974). Low temperatures slow down chemical reactions and biological processes. Weathering rates and production of dissolved chemicals in Arctic soils are reduced by cold. This is compounded by the presence of permafrost, whereby soils are frozen for most of the year, with only a shallow upper layer of thawed soil during the brief summer. Thus, Arctic soils are immature and low in nutrients (Stonehouse 1989).

The most important factor for the development of life in the Arctic is the length of the growing season (Chernov 1985), which ranges from three to four months in the Low Arctic to as little as one to two and a half months in the High Arctic (Bliss 1981b). This gives little time for growth of plants and represents a short time span during which herbivores have access to quality forage. In addition, the summer provides a short window during which poikilothermic (cold-blooded) animals can develop.

Low temperatures result in extensive ice cover of Arctic freshwater and marine habitats. This results in reduced light penetration and therefore reduced photosynthesis. In some lakes and ocean areas, ice cover lasts year-round and productivity is very limited.

There is a large variety of physiological and behavioral adaptations of Arctic biota to cold. Important among these is the metabolic use of lipids as stored energy and as a source of energy. This has implications for contaminant levels in Arctic wildlife and is discussed further in the following chapters.

Low light levels

The region north of the Arctic Circle receives one-third to one-half of the annual solar radiation compared to that reaching temperate and equatorial latitudes. During the Arctic summer, this radiation is received 24 hours a day, resulting in a relatively large amount of incoming solar energy (Fridriksson 1986, Odum 1983 in Freedman et al. 1994). However, much of this energy is used to melt ice and snow, and over 50% of the total annual radiation is received prior to completion of spring melt, which usually occurs in June. Thus, the growing season typically begins when solar radiation levels are already declining (Courtin and Labine 1977, Fridriksson 1986, Etkin and Agnew 1992). Furthermore, on average, about 90% of the incoming radiation is reflected back to space due to the high surface albedo resulting from snow and ice cover and limited vegetation cover (Stonehouse 1989). In the marine environment, sea ice and snow cover further limit energy input. However, there is some light penetration before all the ice is melted, allowing spring production to begin before the water is open.

Water availability

Arctic ecosystems generally receive little precipitation and much of this is received in the form of snow. Studies have shown that 80 - 90% of the annual runoff in the Arctic can occur in just two to three weeks during snowmelt, followed by an abrupt shift to low runoff during the growing season (Bliss et al. 1984, Bliss 1986). Due to low levels of evaporation under the cold conditions, and the presence of permafrost acting as a barrier to subsurface drainage, the moisture provided by snowmelt and rainfall is largely retained. This is especially evident in the Low Arctic tundra with its numerous wetlands (Chernov 1985, Stonehouse 1989). However, in the polar desert regions of the High Arctic, where the levels of precipitation are extremely low and snowmelt is the key contributor to

water budgets, the cessation of runoff following snowmelt can result in very low availability of moisture during the growing season (Bliss et al. 1984, Bliss 1986).

Anthropogenic stressors

Arctic species are generally not considered 'sensitive', since environmental tolerances of most species are broad. However, the physical environment of the Arctic is sensitive. There are numerous stressors, not directly related to chemical contamination, which do and will continue to affect the Arctic. These could change the effects of contaminants on species or ecosystems. Among these stressors are habitat destruction due to hydroelectric development, increased human settlement and activity, resource extraction, and over-harvesting (Welch 1995). The Arctic terrestrial environment is very susceptible to physical destruction. Evidence of human activity is often still visible after hundreds of years. In some cases, ecotourism has already led to habitat destruction and harassment of animals.

Nearly all populations of large Arctic mammals have been considerably reduced from historic levels. In most cases, the protected or endangered status of Arctic species is due to habitat degradation or excessive harvesting, with species at the higher trophic levels most affected. Some marine mammal stocks are harvested by more than one country, and these countries are not always in agreement about migration patterns, sustainable catches, or the rights of native peoples.

1.5.5.2. <u>General ecological characteristics of Arctic ecosystems relevant to contaminants</u>

Low productivity

Productivity in terrestrial, freshwater, and marine environments is reduced due to limited nutrient availability, low light, low temperatures, ice cover, and short growing seasons. The low productivity in the Arctic results in slowergrowing and longer-lived poikilotherms than in temperate climates. Some Arctic insects, for example, can take up to 14 years to complete their life cycle and Arctic char up to 12 years (Remmert 1980, Kulak and Kevin 1994). Arctic mammals grow at rates similar to temperate mammals of the same size, however, the large mammals (e.g. whales) tend to take a long time to reach maturity.

Bioaccumulation and biomagnification

Levels of some contaminants, particularly metals, in specific tissues and organs of a number of temperate and Arctic species increase with age. This is due to bioaccumulation, i.e., increases in contaminant concentrations in biota with continued exposure over time. Some organic contaminants become further concentrated in animals with each successive step up a food chain, a process called biomagnification.

The burden of contaminants stored in the body of animals usually increases with age, unless they have some mechanism for breaking down or excreting the chemicals. Older animals are thus more likely to have higher levels of some contaminants. The age effect is further pronounced in the Arctic by the fact that predators, including people, are more likely to eat older animals than those that are hunted in more southern climates.

Cyclic annual productivity

Arctic ecosystems are highly cyclic due to seasonal fluctuations in light levels, nutrient inputs, and temperature. Nutrients and contaminants deposited on snow, ice, soil, and plants during the Arctic winter can be mobilized and assimilated very quickly in the spring when sunlight returns and temperatures rise. In freshwater systems, the spring melt carries nutrients and some contaminants into streams, ponds, and lakes. In the Arctic marine environment, a burst of primary productivity occurs under the ice when light levels become sufficiently high in the spring. At this time, nutrients and contaminants can move into, and through, food chains very rapidly.

Cyclic productivity in biota is related to many physiological and behavioral adaptations of animals to their environment. One such adaptation is to consume and store energy and nutrients when food is available, and metabolize these when food is lacking. Another adaptation is to migrate to superior over-wintering, feeding or spawning habitats. Migratory species include: small birds that may migrate over two continents, foraging mammals such as caribou that move from the boreal forest to summer grazing areas on the tundra, fish that travel to find favorable spawning sites, and whales that move in search of food. This means that contaminants in some species, and also in the predators that consume them, may not relate to contaminant deposition in the Arctic, but to levels in other parts of the world.

Low species diversity

The low species diversity in the Arctic is a consequence of low absolute productivity and recent glaciations. In contrast, the Antarctic marine environment, which has not experienced such glaciations, has considerably higher biological diversity and an accompanying higher degree of specialization (Dunbar 1986). Although listings of Arctic species may appear substantial, the number of species in any particular area is usually very limited. Because of this low biological diversity, some food chains may be very simple, for example, the lichen-caribouwolf food chain in Arctic Canada. The complexity of food webs increases as Arctic ecosystems grade into temperate systems.

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 of most Arctic species adjust their feeding habits, growth rates, 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 will not have a well-defined position in the food web. For example, freshwater Gammarus can be herbivorous, but is carnivorous if possible; a few individuals in a population of freshwater fish may be cannibals; and walrus may eat seals if alternative foods are lacking. Feeding strategies may also depend on the age and experience of an animal, and may differ from one year to the next.

1.6. PATTERNS OF DEVELOPMENT/SETTLEMENT

The sources of most contaminants of interest to AMAP generally lie outside of the Arctic region. In addition, there is little agricultural activity within the boundary of the Arctic as defined for this report. Therefore, this section will not discuss urban, agricultural and industrial complexes but will serve as an introduction to the inhabitants of the Arctic. The information is intended to help understand how contaminants may affect Arctic residents. The impacts that both contaminants and, more insidiously, the fear of contaminants have on, in particular, indigenous peoples and cultures demonstrate the need for effective communication and for preventing contamination that may lead to adverse effects on Arctic peoples.

A primary focus of this report is the indigenous peoples of the Arctic. This is for two reasons. First, indigenous peoples have been living as part of the Arctic ecosystem for millennia, and in most areas, continue to do so. As consumers of local resources, they are in some ways the end recipients of the types of pollution that are transported long distances, as described throughout this report. Many of the effects of environmental contamination are likely to be most pronounced among indigenous peoples. Second, the cultures and traditions of Arctic indigenous peoples are found nowhere else. Most of these groups continue their patterns of resource use, maintain their cultural traditions, and fight for their rights to continue to do so. Their connection to the Arctic environment is unique and multifaceted. This relationship is everywhere affected, and in some cases at risk of disappearing, due to dislocation of people and traditional activities. In other cases, the very existence of some indigenous groups is at risk. Some have become extinct, even within the twentieth century. Arctic indigenous peoples are the most fragile elements of human society in the Arctic and the most susceptible to environmental change. As such, they deserve special attention to their ways of life, living conditions, and prospects for the future.

In addition, of course, large numbers of immigrants have moved to the region. In the Nordic countries and Russia, this has taken place over several centuries. In North America, only in the past century have a significant number of new arrivals taken up permanent residence. While most of these immigrants have come from European cultures, other peoples are moving to the North as well, increasing the ethnic, racial, and cultural diversity of the Arctic population. Throughout the Arctic, the non-indigenous population is growing, and in many areas is larger than the indigenous population. Although some mutual assimilation and acculturation take place, cultural, social, economic, and other differences remain between the two groups, and their activities, diets, and other routes of exposure to contaminants may vary as well. Some members of the general populations depend upon the Arctic environment, and are sensitive to environmental change. Their concerns must be considered along with those of indigenous peoples.

Iceland and the Faeroe Islands occupy an ambiguous place in the dichotomy between indigenous and nonindigenous. The settlers who arrived before year 1000 found no inhabitants, and their descendants continue to live in the islands, with distinct languages and cultures. Their societies are susceptible to environmental change in their lands and seas. The great diversity of human society in the circumpolar north confounds our attempts to summarize the characteristics of the Arctic population. The Arctic comprises eight nationalities and about three dozen, distinct, indigenous cultures. Statistical averages may conceal widely ranging figures, and demographic categories appropriate to societies in the temperate zone may obscure important facets of life in the Arctic.

Despite these cautions, there are certain broad similarities across the countries and regions of the Arctic. The Arctic population is, in general, young. In many regions, birth rates are high, and infant mortality is low or declining. The exception is Russia, where the economic situation has caused extensive emigration, and declining health care has led to increased mortality of all age groups.

Arctic populations typically live in close contact with the local environment. This is especially true of indigenous peoples, but also applies to many of the other inhabitants of the region. While Iceland and the Faeroe Islands have no indigenous peoples, local foods remain important dietary and cultural resources. Use of local and indigenous foods is a common characteristic of indigenous groups in the Arctic, and a wide range of animals and plants are used in this way. Meats and other animal products, especially from reindeer or caribou, marine mammals, and fish, account for the vast majority of indigenous foods.

As an indication of overall health, life expectancies in the Arctic areas of each country are lower than the national averages. This is largely due to high accident rates, as well as the physical and psychological stresses of the Arctic climate. Specific diseases vary, often by ethnicity. While this may be due to genetic factors, lifestyles and diets are also important determinants of cancer, heart disease, diabetes, and other major illnesses.

Health care in the Arctic is improving, but sparse populations and long distances limit the capacities of hospitals and clinics to provide medical care at the levels expected in less remote or more densely populated areas. Again, Russia is an exception to the overall trend, as disruptions to supply lines and removal of incentives to live in the north have reduced health care capacities below their 1980s levels.

Two conclusions emerge from this brief summary. First, while we have a basic understanding of the living conditions and lifestyles of Arctic residents, many data are lacking, prohibiting study of trends or conditions among Arctic residents that might help illuminate the potential and actual impacts of environmental contaminants. This is especially true of dietary and health data.

Second, effectively communicating information about contaminants is critical, especially in the absence of quantitative data on exposure and effects at low levels. This is a two-way process, for not only do Arctic residents have the right to good information about their lands, seas, and their bodies, they also have knowledge that may assist our overall understanding of environmental change. While many facets of contaminants in the Arctic are not well understood, gaining knowledge requires sound research and effective partnerships between researchers, policy makers, and the residents of the region.

Figures alone, however, do not always show why the future of Arctic peoples depends on preventing the degradation of the Arctic environment. These are intangible or elusive matters, but they also lie at the heart of the global debate about pollution, climate change, and sustainable development. For the peoples of the Arctic, these are crucial questions today. Their lives and cultures are affected, and their daily decisions often include such previously arcane questions as the levels of industrial and agricultural chemicals in local foods such as seal and reindeer. These peoples, these societies, these cultures must be at the forefront of research and policy addressing the Arctic.

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2. SOURCE CHARACTERIZATION

2.1. BACKGROUND INFORMATION TO PTS SOURCES

There can be some confusion about the term 'source'. For example, is the source of a pesticide the factory where it is made or the field upon which it has been sprayed and, therefore, introduced into the environment? The answer depends on whether one wishes to abate the production of the material or to determine the pathway of a pesticide in or into the Arctic ecosystem. In the later case, one needs to know the location of release not the location of manufacture. The context should make the intended meaning clear.

The sources of most contaminants of interest to AMAP lie outside of the Arctic region. Hence, the levels of contaminants in the Arctic depend to a large degree on the effectiveness of transport mechanism. These mechanisms are discussed in Chapter 4. In addition, since large sources external to the Arctic will be discussed in detail in other regional reports, the focus in this report will be the Arctic sources though, in some instances, they may be relatively less important than the 'foreign' sources. A brief description of the main Arctic sources follows.

Perhaps the largest single source of anthropogenic contaminants are the mines, where the presence of sulfides requires roasting prior to the cyanide process. The fumes produced from the roasting process can contain significant amounts of arsenic trioxide and sulfur dioxide, some of which are released to the atmosphere (Thomas et al. 1991).

Military installations in the Arctic areas of North America and probably elsewhere, have used DDT mixtures, as well PCB-containing devices (electrical transformers and capacitors) widely. These PTSs remain in the Arctic.

The potential for agricultural activities to act as sources for pesticides to the Arctic environment is real. While there is little agricultural activity within the boundary of the Arctic as defined for this report, the agricultural lands of the entire northern hemisphere are potential sources for atmospheric delivery of contaminants. As well, the large rivers in Canada and Russia which drain to the Arctic, often have drainage basins extending far outside of the Arctic that contain extensive agricultural activities, which represent a potential source of contaminants.

In the case of unintentional by-products, the most important sources of these chemicals are local.

Local development has a direct influence on water quality. In addition to the effects on the environment from land clearing, construction, and decreased permeability, the quality of water returned to the basin is usually diminished even after treatment. The disposal of industrial and municipal wastewaters is always a concern. Storm and melt water runoff, which is usually routed directly to receiving water bodies without treatment, may be highly contaminated due to spills and localized atmospheric fallout. These factors can exacerbate the problems of providing clean water to basin residents. This and the subsequent section provide some examples of the industrial and municipal sources of contamination within the Arctic.

Contaminants are dumped at sea in designated dumping grounds. The 1972 London Dumping Convention regulates all dumping at sea. Both shipping and oil production contribute to the input of petroleum hydrocarbons to the sea. Municipal and industrial wastes, often untreated, are discharged directly to the marine environment. Records of the chemical nature of these discharges are frequently not available.

Large volumes of used oil-drilling fluids have been released into Arctic wetlands during exploration. Increased concentrations of common and trace metals and organic hydrocarbons have been found in ponds both near and at a distance from drill site sumps (Garland et al. 1988).

2.2. PRODUCTION, USE AND EMISSION

POPs are not found naturally in nature. They are produced / manufactured intentionally or unintentionally. In the case of pesticides and industrial products, we are interested in the location of the production facilities and the quantity of materials manufactured, as well as where they are used or stored in the Arctic. In the case of unintentional by-products, we need to know where they are produced.

Sources of POPs in the Arctic are not well documented in most cases, but are 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. Finally, pesticides have been deliberately used for insect control.

2.2.1. Pesticides - Production and Use

2.2.1.1. <u>Global</u>

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 1.

Pesticides	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 29000	1980 1990	Li et al. 1996				
Technical lindane	5900 4000	1980 1990	Li et al. 1996				
α-НСН	28000 20400	1980 1990	Li et al. 1996				
ү-НСН	11900 8400	1980 1990	Li et al. 1996				
Interpolated							
DDT	2600000 990000	1950-1993 1970-1993	Voldner and Li 1995				
Toxaphene	1330000 670000	1950-1993 1970-1993	Voldner and Li 1993				

Table 1: Global cumulative usage of selected pesticides for various periods of time (Barrie et al. 1997)

2.2.1.2. Regional

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). 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.

2.2.2. Industrial Chemicals

2.2.2.1. Regional - Use and Restrictions

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. 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.

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.

2.2.2.2. Local

2.2.2.2.1. 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 not documented.

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.

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, snow pack, 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 km² 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 defence sites (FUDS), including 49 DEW Line sites, have been identified in the state of Alaska for possible environmental assessment and remediation (US Department of Defence 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.

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.

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

Larger urban settlements, particularly those along the Arctic coasts and major Arctic rivers discharge wastewater 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, PCBs 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 PCBs 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 PCBs (SFT 1996).

Another potential source of PCBs is transformers on oil-drilling platforms. The oil well platform Piper Alpha released 5 tonnes of PCBs 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. PCBs 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.

2.2.3. Unintentional by-products

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.

2.2.3.1. <u>Global</u>

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 wood burning. 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.

2.2.3.2. Regional /Local

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. 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.

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.

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, Canada, 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/Fs 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.

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 (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.

2.2.4. Metals Sources

Metals are emitted to the atmosphere from both natural and anthropogenic sources. Very few of the sources that directly result in the contamination of the Arctic environment are located in the Arctic. Metals released to the environment outside the Arctic are transported to the Arctic via air currents, rivers, and ocean currents. This section reviews the sources and fluxes of metals within the Arctic as well as their sources outside and pathways to the Arctic.

2.2.4.1. Natural sources

An accurate inventory of heavy metal sources and emissions to the atmosphere from natural processes is needed to make a complete assessment of the extent of regional and global pollution by heavy metals in the Arctic. It is generally presumed that the principal natural sources of heavy metals include wind-borne soil particles, volcanoes, sea salt spray, and wild forest fires (Nriagu 1989). Recent studies have shown, however, that particulate organic matter is the dominant component of atmospheric aerosols in non-urban areas (Talbot et al. 1988, Artaxo et al. 1988) and that over 60% of the airborne heavy metals in forested regions can be attributed to aerosols of biogenic origin (Zoller 1983).

A global assessment of natural sources of atmospheric heavy metals has been made by Nriagu (1989). Biogenic sources can account, on average, for over 50% of the Se, Hg, and Mo, and from 30 to 50% of the As, Cd, Cu, Mn, Pb, and Zn, released annually to the atmosphere from natural sources. Volcanic emissions can account for 40-50% of the Cd and Hg and 20-40% of the As, Cr, Cu, Ni, Pb, and Sb released annually from natural sources. Sea salt aerosols seem to account for < 10% of atmospheric heavy metals from natural sources. Finally, soil-derived dusts can account for 20-30% of the Cu, Mo, Ni, Pb, Sb, and Zn released annually to the atmosphere. As the accuracy of emission estimates for natural sources is low, these percentage contributions should be considered as approximations only.

The natural sources of heavy metals, which influence the freshwater, terrestrial, and marine environment, are even more difficult to assess than the atmospheric sources. In general, soils and sediments tend to reflect the composition of their parent material. Soils and sediments in mineralized areas, therefore, usually have the highest concentrations of the corresponding metals. For example, rocks with high Hg content usually occur in areas of crustal instability where volcanic and geothermal activities are high.

It is also very difficult to assess the extent to which emissions from natural processes affect the contamination of the Arctic environment. In general, fluxes from these processes within the Arctic are regarded as less significant than anthropogenic releases, both within and outside the Arctic. However, very long-range transport within air masses of soil particles from deserts in Asia and Africa to the High Arctic has been postulated by Pacyna and Ottar (1988). A series of haze bands over Barrow, Alaska in April and May 1976 was found to consist of dust (Rahn et al. 1981). The bulk elemental composition of the particles was crustal or near-crustal and their massmedian radius of about 2 μ m indicated that they could have originated more than 5000 km from Alaska. Trajectory analysis showed that these particles could have passed over the arid and semi-arid regions of eastern Asia during intense dust storms that had occurred there. This hypothesis has been confirmed by measurements in the Norwegian Arctic (Pacyna and Ottar 1989) and in the Canadian Arctic (Welch et al. 1991). The origin and evolution of dust clouds in central Asia has recently received consideration.

Although the existence of natural constituents in the Arctic aerosol originating in central Asia was explained by long-range transport of eroded dust from the deserts in Asia and Africa during dust storms, no quantitative assessment has been made of how much of the eroded dust and attached heavy metals are transported from the Asian and African deserts to the Arctic.

2.2.4.2. <u>Anthropogenic sources</u>

High temperature processes generate various metals. These processes include coal and oil combustion in electric power stations and heating and industrial plants, gasoline combustion, roasting and smelting of ores in non-ferrous metal smelters, melting operations in ferrous foundries, refuse incineration, and kiln operations in cement plants. The metals enter the atmosphere and the aquatic and terrestrial ecosystems; virtually every industry discharges metals into these ecosystems. The assessment presented here is focused on the principal industrial and commercial users of raw materials and water, and on producers of solid wastes.

2.2.4.2.1. Atmospheric Sources and fluxes on a global scale

The first quantitative worldwide estimate of the annual industrial input of 16 heavy elements into air, soil, and water was published by Nriagu and Pacyna (1988). Pyrometallurgical processes in the primary non-ferrous metal industries are an important source of Pb and Se. Combustion of coal in electric power plants and industrial, commercial, and residential burners is the major source of anthropogenic Hg, Mo, and Se. Combustion of leaded gasoline is still the major source of Pb. Little information is available on the emission of heavy metals from various diffuse (fugitive) sources. A comparison of the median values of worldwide emissions of heavy metals from natural and anthropogenic sources suggests that human activities generate emissions of heavy metals that exceed those from natural sources. Therefore, anthropogenic emissions result in significant alterations of the natural biogeochemical cycling of many heavy metals in the global environment.

Recently, a revision of the global emission inventory of Pb was prepared for the reference year 1989 (Pacyna et al. 1993b) as a part of the Global Emission Inventory Activities (GEIA) operated within the IGBP International Global Atmospheric Chemistry (IGAC) program. The results show that in 1989 the maximum emission was about 209 000 tonnes of Pb, of which 62% came from gasoline combustion, followed by 26% from non-ferrous metal production (Pacyna et al. 1995).

An estimate of global anthropogenic emissions of Hg has also been completed for AMAP (Pacyna and Pacyna 1996). The preliminary data suggest that between 1300 and 2150 tonnes of Hg are emitted annually to the atmosphere at the present time. The major sources of these emissions are combustion of coal to produce electricity and heat (60%), followed by gold production and waste disposal.

2.2.4.2.2. Aquatic Ecosystems: Global Sources and Fluxes

For the metals of interest in this assessment, Hg, Pb and Sn, the major sources of anthropogenic metal contamination of aquatic ecosystems (including the ocean) include coal-burning power plants (As, Hg, and Se in particular), non-ferrous metal smelters (Cd, Ni, Pb, and Se) and the dumping of sewage sludge (As, Mn, and Pb). The atmosphere is the major route of Pb entry into natural waters. The results of the worldwide assessment of anthropogenic inputs of 13 metals into the aquatic ecosystem have been reported by Nriagu and Pacyna (1988).

For most of the heavy metals, the annual anthropogenic inputs into water exceed the quantities emitted to the atmosphere. If it is assumed that only 25% of the industrial effluents are discharged into lakes and rivers, the average concentrations in these waters should reach levels several-fold higher than those in unpolluted lakes and rivers. In other words, the current rate of worldwide industrial inputs greatly exceeds the baseline burdens of heavy metals in the average lake and river. Most of the effluent discharges occur in Europe (including Russia), North America, and some Asian countries, implying that the contamination of the freshwater resources in these regions may be much more severe than is generally realized.

2.2.4.2.3. Terrestrial Ecosystems: Global Input and Output

The first quantitative assessment of worldwide fluxes of heavy metals into soils was prepared by Nriagu and Pacyna (1988) for the reference year 1983. The estimates suggest that soils are receiving large quantities of heavy metals from disposal of a variety of industrial wastes. The two principal sources of heavy metals in soils worldwide are the disposal of ash residues from coal combustion and the general breakdown and weathering of commercial products on land. Urban refuse represents an important source of Cu, Hg, Pb, and Zn with notable contributions of Cd, Pb, and V also coming via the atmosphere. The large volumes of wastes associated with animal husbandry, logging, and agricultural and food production can significantly affect the heavy metal budget of many soils. Although municipal sewage sludge may not be particularly important source on a global scale, it can be one of the most important sources of metal contamination of soils on a local scale.

If the estimated metal inputs are distributed uniformly over the cultivated land area, the annual rates of metal application are not very significant because of the large background reservoir of heavy metals. Nevertheless, each soil has a limited retention capacity for heavy metals and there is growing concern that, at the current rate of anthropogenic input, many soils in various parts of the world (e.g., central Europe and Japan) either have become or will soon become overloaded with heavy metals (Kabata-Pendias 1984, Asami 1988).

Soils, like waters, can be a source of atmospheric contamination of some heavy metals, particularly Hg. These emissions result from various out-gassing of Hg laden rock and from volatilization of Hg from soils, vegetation, and water bodies. Current data suggest that these emissions of Hg are of the same order as emissions from anthropogenic sources (Pacyna and Keeler 1994).

It should be noted that emissions from natural sources are difficult to distinguish from so-called secondary emissions and diffusive re-emissions from anthropogenic sources. These include re-emissions of previously deposited Hg as well as emissions resulting from discharge into water bodies and from contaminated soils. Hence, it is more appropriate to differentiate between pre-industrial and post-industrial diffuse sources (Lindqvist 1991) than between natural and anthropogenic re-emission.

2.2.4.3. Emission inventories for sources within and outside the Arctic

Obviously, only a part of worldwide emissions is responsible for the contamination of the Arctic environment by heavy metals. It is critical to identify sources important to the Arctic and to quantify the amount of emissions from these sources that reaches the Arctic region. The results of source-receptor studies, summarized in Pacyna (1991), indicate that emissions from sources in Eurasia contribute more than half of the air pollution measured in the Arctic. The major source regions include the Urals, the Kola Peninsula, the Norilsk area, and the industrial regions in Central and Eastern Europe (Rahn and McCaffrey 1980, Rahn and Lowenthal 1984).

The contributions of European and North American emissions to Arctic air pollution seem to be smaller than the contribution from the Russian sources. European and North American emissions are, however, major contributors to the contamination of the sub-arctic regions, such as northern Scandinavia (European emissions) and the northern part of Canada (North American emissions).

These sources, located outside the Arctic, are important in discussing the contamination of the High Arctic environment, which remains largely unaffected by local industrial activities. It is necessary, therefore, to review the emissions from these outside sources in order to assess quantitatively their contribution to Arctic contamination.

2.2.4.3.1. Atmospheric emissions from sources outside the Arctic and their trends

The first attempt to estimate atmospheric emissions of heavy metals from anthropogenic sources in Europe was completed at the beginning of the 1980s (Pacyna 1984). This European survey has since been updated, completed, and emission gridded (Axenfeld et al. 1992, a review by Pacyna 1994). National emission inventories have only recently become available in the European countries. These emission data, together with the international expert estimates, were used to compile current European emission estimates for Hg, Pb, As, Cd, and Zn. A spatial distribution of the heavy metal emission estimates in Europe is available within the EMEP 150 km by 150 km grid system (Axenfeld et al. 1992).

Changes of heavy metal emissions to the atmosphere from sources in Europe from the 1950s until present have also been studied (Olendrzynski et al. 1995). A decreasing trend of Hg levels in atmospheric deposition in Scandinavia has been observed during the last few years (Munthe et al. 1994). This trend was related to a possible decline of Hg emissions, particularly in Central and Eastern Europe. These emissions can be expected to decrease by up to 30%. The economic decline in this part of Europe at the beginning of the 1990s, related tithe transition from centrally planned economies to market oriented ones, was suggested as the major reason for the possible changes in Hg emissions. In addition, the lower consumption of fuels in Eastern Europe in recent years is an important factor, which has caused the decrease of Hg emissions.

Pursuant to the requirements of the 1990 US Clean Air Act Amendment, an interim toxic emission inventory has been developed for the continental United States. Emission estimates for As, Pb (both after Voldner and Smith 1989) and Hg (after US EPA 1993b) from major source categories in the United States are available.

Environment Canada has initiated several projects on emission inventory development for heavy metals in Canada. The 1982 emissions of Hg, Pb, As, Cd, Cr, and Cu from major source categories have been determined, based on data from Jacques (1987). Emission estimates for Hg and Pb have been revised to account for major changes in consumer patterns in recent years. It is believed that the reliability of emission data from European North America decreases in the following order: Pb > Hg and Cd > remaining heavy metals. An accuracy of <25% was suggested for the emission estimates of Pb, 50% or less for Hg, and 100% for the rest of the metals (Pacyna 1994).

2.2.4.3.2. Atmospheric emissions from sources within the Arctic

In addition to outside sources, there are also sources of heavy metals within the Arctic.

Preliminary estimates of atmospheric emissions of Pb, As, Cd, Cu, Cr, Mn, Ni, Sb, Se, V, and Zn for major source regions in the former Soviet Union, including the areas outside of the Arctic, have been prepared by NILU (1984)

on the basis of emission factors and statistical data for the reference year 1979/80. Emissions from non-ferrous and ferrous metal production, fossil fuel combustion, and gasoline combustion were estimated to dominate the total emissions in those regions entirely or partly located within the Arctic, such as the Kola Peninsula, the Norilsk area, and the northern Urals. The inventory of sources in the European part of the former Soviet Union is included in the European emission survey noted above. A list of major point sources of heavy metal emissions to the atmosphere in the Urals and the Asian part of the former Soviet Union has been produced by (Pacyna et al. 1993b).

2.2.4.3.3. Aquatic discharges from sources outside the Arctic

Compared with atmospheric emissions, much less information is available on emission inventories reporting discharges of heavy metals to the aquatic environment. In one of very few approaches in Europe, discharges of Pb, Cu, Ni, and Zn from major point sources were estimated for the Commission of the European Communities (EC) (Daamen et al. 1990). The manufacture of ceramics is by far the most important industry discharging Pb to the aquatic environment.

2.2.4.3.4. Aquatic discharges from sources within the Arctic

In Greenland, mining has been an important source of local heavy metal pollution of the sea. Asmund et al. (1991) have made an estimate of quantities of Pb, Zn, and Cd released to the sea from two mines in Greenland. One of these mines, the Black Angel Pb-Zn mine in Uummannaq, West Greenland, operated from 1973 to 1990. During production, it was estimated that 6-12 tonnes of Pb were released annually to seawater. Dumping of waste rock at the time the mine was closed in 1990 added about 1 tonne of Pb to this total (Asmund 1992a, 1992b). After mine closure, the amounts released have declined drastically, to 10% or less of the amounts measured during production (Asmund 1992a, 1992b). At a closed cryolite mine in Ivittuutin south Greenland, Asmund et al. (1991) estimate an annual input to the sea of 0.4-1 tonnes of Pb from deposited waste rock in the intertidal zone. No estimates are available from a closed Pb mine at Mestersvig in East Greenland, which operated from 1956 to 1963, but environmental studies of seaweed, mussels, and fish indicate that significant amounts of Pb and Zn have entered the marine environment (Johansen et al. 1985). A comparison of the Greenland mines with the Pb-Zn mine at Nanisivik in the Canadian Arctic shows that the Canadian mine has impacted the sea to a much lesser extent (Asmund et al. 1991). Information on the releases of metals from the Red Dog Pb-Zn mine in Alaska and their impaction the environment is not yet available.

2.3. DATA GAPS

In some locations in Russia, there are high HCH levels in lake water and high PCB and DDT levels are now seen in snow, rivers, seawater, coastal sediments, and the few samples of invertebrates, fish, reindeer, lemming, seabirds, seal, and beluga. These findings indicate possible fresh releases or improper disposal. These high levels must be verified.

Military and civilian sites and dumpsites that contain significant amounts of electrical equipment may be sources of PCB-contamination. Local PCDD/Fs contamination in the vicinity of a smelter has also been demonstrated. Surveys of all such sites within the Arctic should be made to determine the circumpolar scope of the problem.

Numerous local sources of other POPs also exist, but have not been studied. Surveys of local sources of contamination by POPs within the Arctic are needed to quantify the emissions and leakage.

2.4. SUMMARY OF HOT SPOTS AND MOST SIGNIFICANT REGIONAL SOURCES

Over much of the Arctic, the levels of POPs cannot be related to known use and/or releases from potential sources within the Arctic and can only be explained by long-range transport from lower latitudes. For the metal contamination in the Arctic, industrial sources in Europe and North America account for up to one-third of the deposition, with maximum input in winter. These sources PTSs are discussed in the UNEP reports from other regions.

The following sources are the most important ones in the Arctic, though they may be less important than sources outside the Arctic (see Annex I).

2.4.1. POPs

• 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 not documented.

- The Barents region of the Russian Arctic is the most highly populated and industrially developed part of the circum-polar Arctic. The province of Murmansk has over one million inhabitants with many in major industrial cities, including Murmansk, Apatity, Kirovsk, Kandalaksha, and Monchegorsk. To the east of Murmansk is the province of Archangel, which, including the Nenets autonomous area, in 1993 had a total population of 1 561 000 inhabitants with over 70% in major cities (including the cities of Archangel and Severodvinsk). In highly urbanized areas, surface runoff waters, including organized industrial and communal wastewater discharges, can contribute up to 40-50% of total pollution entering the water bodies (WHO/UNEP).
- Aside from the PTSs that are found on military establishments, there are a large number of other sites where PTSs, notably PCBs may occur in high concentrations. These sites include:
 - Electrical capacitors and fluorescent light ballasts, manufactured prior to 1979, contained small quantities of PCBs. These may have been disposed of at municipal landfills, as equipment was replaced.
 - PCBs in sealants in buildings
 - o Leakage from solid waste disposal sites at the coal-mining settlements
 - Hydroelectricity power plants built during the 1960s and early 1970s whose transformers most probably contained PCBs at one time
 - Effluents from oil drilling sumps and tailings effluent from metal ore and coal mines, which in the past may have used hydraulic and drilling fluids containing PCBs.
- Combustion, especially of municipal garbage, is a common sight in the Arctic and could be a source of PCDD/Fs and HCB.
- Pulp and paper mills employing elemental chlorine for bleaching wood pulp are potential sources of local PCDD/F contamination.
- Overall, metal smelting and other combustion-related processes within the Arctic could be important contributors to PCDD/F contamination.

2.4.2. Metals

- Combustion of fossil fuels to produce electricity and heat is one of the major heavy metal source categories present in the region.
- Non-ferrous and ferrous metal production facilities, particularly in the Russian Arctic (Kola Peninsula and at Norilsk), are important heavy metal sources of pollution to the air and aquatic sectors.
- Although municipal sewage sludge may not be a particularly important metal source on a global scale, it can be one of the most important sources of contamination of soils on a local scale. Near point sources such as mine sites and some Russian estuaries, heavy metals exceed background levels up to 30 km from the source.

2.5. **References**

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3. ENVIRONMENTAL LEVELS, TOXICOLOGICAL AND ECOTOXICOLOGICAL PATTERNS

The first sections of this chapter deal with the environmental levels and trends. Here the relative spatial and temporal variations in environmental concentrations of PTSs are described. The following section, Toxicology, addresses human health in the context of a variety of standard and thresholds. General toxicology and effects are discussed first, followed by the levels and trends of PTSs based on national surveys. Section 3 reports on the effects of PTSs in the environment. The compartments are defined descriptively; air, fresh water, etc.

3.1. LEVELS AND TRENDS

3.1.1. POPs

3.1.1.1. Abiotic Media

3.1.1.1.1. 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.

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 that 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 DDT and PCB 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 that 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.

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. 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/m³ at Lista (S. Norway) compared to levels of 2-38 pg/m³ 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 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.

3.1.1.1.2. Terrestrial Environment

Soils and humus, contaminated with OCs, have been investigated only in a limited number of regions with the majority of samples originating near military radar facilities.

3.1.1.1.3. Freshwater Environment

Recent studies of contaminants in water, sediments, and biota from Arctic lakes and rivers have greatly expanded the information available on OC levels.

Data on PCBs and DDT in Russian rivers are problematic. PCB and DDT 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 DDT 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 HCH because most of the OCs were in the dissolved (or dissolved organic carbon associated) phase.

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., $\log_{Kow} < 5.5$). 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.

HCH 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 DDT and PCB 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 PCBs in Arctic lake waters in Canada exceeded the USEPA Great Lakes Water Quality guideline for protection of aquatic life of 17 pg/L, but DDT did not exceed the guideline limit of 870 pg/L. Levels of PCB 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 freshwaters, 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 PCBs and DDT 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 PCBs and DDT in Arctic-lake, surface sediments generally do not exceed guideline limits for effects on aquatic life. 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 PCB, 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 DDT of 1.6 ng/g dw was exceeded at about ten locations, 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. 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 melt-water does not mix and net sedimentation rates are generally low (Diamond 1994, Semkin 1996).

3.1.1.1.4. Marine Environment

The information now available on POPs in seawater, ice, snow, suspended sediment, and bottom sediments is 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 \approx toxaphene > chlordanes \approx PCBs > DDTs, as described by Bidleman et al. (1990). An exception seems to be the Russian Arctic seas, where the order is reversed, Σ PCBs > Σ DDT> Σ HCH> Σ CHL> chlorobenzenes. Σ PCB concentrations in some seawater samples are so high, for example, 15 ng/L, 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 PCBs 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 Σ HCH in the world's oceans are found in the Arctic Ocean, especially in the Beaufort Sea and Canadian Arctic Archipelago. Σ HCH levels measured in the late 1980's to early 1990's 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, PCBs, 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 Σ PCB 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.

While evidence for transfer of OCs via ice may be lacking, there is evidence that the elevated concentrations of Σ HCH, Σ DDT, and Σ PCB in Russian rivers (on SPM or in whole water) and near-shore 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 Σ PCB 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 associated with low probability of effects (Long et al. 1995).

A distinct difference between offshore and near-shore sediments is evident at locations along the coast of Norway between Bergen and Tromsø and along the Russian coast. Relatively high levels of Σ HCH, Σ PCB, 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 near-shore 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.

3.1.1.2. Biotic Media

3.1.1.2.1. 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). However, the present coverage is not adequate in terms of regions studied. Spatial coverage of OC levels in major species is good only for caribou/reindeer (Rangifer), 40

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.

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 $\Sigma PCBs$, 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 either to 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 \rightarrow caribou \rightarrow 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 reflect, therefore, 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 Σ DDT and Σ PCB 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 are 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.

3.1.1.2.2. Freshwater Environment

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, 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 fresh-water 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, 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 $\Sigma PCBs$ and ΣDDT when compared to levels in the Great Lakes. Lake trout from smaller remote lakes in Alberta and northwestern Ontario, which receive contaminants solely from the atmosphere, have PCB 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), which is highly impacted by industries and towns

The Σ PCB 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). 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 g/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.

3.1.1.2.3. Marine Environment

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/Fs 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 Σ DDT in biota from Russian waters. Evidence for this includes higher Σ PCB, Σ DDT, and chlorobenzene levels in bivalve mussels from the Ob and Baydaratskaya Gulfs (southern Kara Sea) than in samples from Iceland or Greenland; higher Σ PCB 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 Σ PCBs 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 Σ DDT 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 Σ PCB levels were found in Greenland halibut muscle from the Norwegian and Barents Sea. Σ PCB concentrations in Greenland halibut muscle do not exceed guidelines of 2 µg/g for import of fish in the European community.

Berg et al. (1997) 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. PCBs and DDT 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 Σ PCBs and Σ DDT in Arctic ringed seals were found in samples from the Yenisey Gulf. PCB 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 Σ PCB and Σ DDT than those in the central Canadian Archipelago and western Greenland. However, ringed seals from eastern Greenland (Scoresbysund), Svalbard, and northern Norway had similar Σ PCB 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 near-shore 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 Σ HCH 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 Σ HCH (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 Σ HCH are consistent with higher Σ HCH 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.

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 Σ PCB 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 Σ PCB 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). Σ PCB 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 Σ PCB 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 $\Sigma PCBs$ 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 μ g/g in fat of males and up to 36.7 μ g/g in females) were found in some adults from Svalbard. Σ PCB levels found in adult males were significantly higher than in young and adult female bears.

The highest Σ CHL (Chlordane) levels were found in young (3.38 µg/g in fat) and subadult bears (3.44 µg/g). Levels of CHL 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 Σ CHL 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. 1997), 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) shows generally increasing OC concentrations from west to east, similar to that observed in several other marine species. Higher Σ PCB levels were found in bears from eastern Greenland and Svalbard, in agreement with results of Bernhoft et al. (1997) 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.

 Σ CHL 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 Σ CHL and HCB in air and seawater in the northern latitudes than in tropical areas (Iwata et al. 1993). The lower Σ CHL levels in polar bears from Wrangel Island and the Alaskan Beaufort Sea area indicate that Σ CHL 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 Σ CHL levels are found in the Canadian Archipelago and Barents Sea compared with the Chukchi Sea.

Concentrations of Σ PCB and Σ CHL 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 μ g/g lw) are in the same range as observed in polar bear, and comparable or higher than in other marine mammals.

Biomagnification factors (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 Σ DDT 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 Σ PCB and Σ CHL 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 Σ PCB and five-fold for Σ CHL at each level of the food web. Σ DDT also increased about seven-fold with each level of the food web, except between seals and bears.

3.1.1.3. <u>Summary of Temporal Trends</u>

The results of a number of temporal trend studies of Arctic biota indicate that Σ PCB and Σ DDT 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, Σ CHL, 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 Σ HCH 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 Σ PCB concentrations in mosses from both coastal and inland areas over the period 1977-1990. Over the same time period, PCBs 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 Σ PCB 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 sub-arctic (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 sub-arctic 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 Σ PCB 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 pre-industrial 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 Σ PCB and Σ DDT 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. PCBs (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 Σ DDT, Σ PCB, and HCB were noted in these Arctic locations. For DDT and PCBs, 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 Σ PCB and Σ DDT have been small, there is no indication that concentrations are leveling off. Levels of Σ PCBs in pike in sub-arctic Finnish lakes also decreased 5- to 10-fold from the early 1970s to the early 1990s. Toxaphene, Σ HCH, 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 parallel observations in lake trout in the Great Lakes (DeVault et al. 1995). The decline in concentrations of Σ DDT 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 Σ PCBs 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 Σ DDT and Σ HCH 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/ Σ DDT as well as in total concentrations of Σ DDT (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 over-winter. 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, Elliott 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 (1995) concluded that, between 1972 and 1991, PCB concentrations in blubber declined about five-fold, while Σ DDT 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 Σ DDT, Σ PCB, Σ CHL, 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 Σ DDT 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.

3.1.2. Metals

3.1.2.1. Abiotic Media

3.1.2.1.1. Air

Median winter concentrations of heavy metals and other compounds have been measured in 1983, 1984, 1986, 1987, and 1989 at Ny-Ålesund, Spitsbergen (after Maenhaut et al. 1996). The median values in the 1989 campaign are generally of the same order as those in the previous winter campaigns, but for some heavy metals having strong anthropogenic sources (such as Pb, V, Mn, Ni, Zn, As, Mo and Sb), they are clearly lower.

Much less information exists on heavy metal concentrations during summer. A comparison of median concentrations measured at Ny-Ålesund and Alert in the Canadian Arctic in winter and summer of 1984 has been made (Maenhaut et al. 1989). The ratios of winter concentrations to summer concentrations range from 3 to 15. The ratios for individual metals are generally similar for the Alert and Ny-Ålesund locations.

A comparison has been made of the winter air concentrations of several heavy metals (Pb, As, Zn, Cu, Mn, V and Ni) in the Norwegian Arctic, Alaska, Greenland, the Canadian Arctic, and Severnaya Zemlya and Wrangel Island in the Russian Arctic in the 1980s and the 1990s, together with European emissions of Pb, As and Zn during the same periods. Concentrations at the beginning of the 1980s are somewhat higher than those at the beginning of the 1990s. At the same time, the emissions of Pb, As and Zn are significantly higher in 1980 than in 1990.

Although there have been a number of successful studies directed at the origin of Arctic air pollution by heavy metals, a quantitative assessment of the portion of the pollution load actually deposited in the High Arctic has not been made.

Rainwater composition was measured at eight Arctic catchment areas in northern Europe: four in Russia, three in Finland, and one in Norway (Reimann et al. 1997). Close to industrial sources in Russia, most of the heavy metals measured show enrichment of two to three orders of magnitude in their median levels, compared with background levels measured in Finland.

The 1980s deposition of heavy metals on the Kola Peninsula was at least one order of magnitude higher than the deposition in the 1960s (in Kryuchkov and Makarova 1989), with the major change occurring in the 1970s.

3.1.2.1.2. Terrestrial Environment

Somewhat limited data for metals in soils are available for Greenland, Norway, Sweden, and Russia. General information on concentrations of Pb, Cu, Cr, Ni, V, and Zn in the soils of northern Fennoscandia is available.

Large amounts of data have been collected in the Kola Peninsula and in neighboring areas of Fennoscandia in an attempt to quantify the effects of emissions from the metallurgical complexes in the area (Pechenganikel in Nikel and Zapolyarnyy, and Severonikel in Monchegorsk) and to study the mobility of metals in soils in areas influenced by acidification.

Metal concentrations in Greenland soils from four remote locations were: < 12-37 μ g/g, < 12-13.8 μ g/g, 8-13 μ g/g, <0.04-0.10 μ g/g, and < 0.01-0.03 μ g/g for Cu, Zn, Pb, Cd, and Hg, respectively. These concentrations probably represent background conditions.

3.1.2.1.3. Fresh Water Environment

Data on the background metal content of freshwater in the Arctic are very limited both temporally and geographically. Erickson and Fowler (1987) report data for 1985 and 1986 in various channels of the Mackenzie River Delta during winter. Mean values were approximately 1.1-1.4 μ g/L, 0.5- 2.5 μ g/L, 0.15-0.99 μ g/L, 10-123 ng/L and 5-8 ng/L for Cu, Zn, Pb, Cd, and Hg, respectively. Average values published for the same metals during the period 1960-1974 (Environment Canada 1981) were 5, 8, 2, 1, and 0.005 μ g/L, respectively. Rovinsky et al.

(1995) report values for Russian rivers in generally the same ranges as noted for the Mackenzie River, namely 0.3-3 μ g/L, 1.8-2.6 μ g/L, 0.15-0.99 μ g/L, 20-290 ng/L, and 20-66 ng/L for Cu, Zn, Pb, Cd, and Hg, respectively. Recent data for 11 rivers in Arctic Canada indicate average concentrations of < 1-2 μ g/L, < 1-2 μ g/L, <0.7-1.3 μ g/L, < 0.2 μ g/L and <0.2 μ g/L for Cu, Zn, Pb, Cd, and Hg, respectively (Jeffries and Carey 1994).

There are numerous examples of localized enrichment of metals in fresh water in the Arctic. Most of these enriched areas are associated with mining operations. Garrow Lake, NWT is used as a tailings pond at a Pb-Zn mine in the Canadian Arctic and concentrations of Cu, Zn, Pb, Cd, and Hg are typically in the order of 28 μ g/L, 360 μ g/L, 1.8 μ g/L, 0.7 μ g/L and 0.03 μ g/L, respectively (INAC 1994).

Skotvold et al. (1996) studied heavy metals in sediments from 91 lakes in northern and Arctic regions of Norway. They concluded that Pb, Cd, Zn, and Hg deposition was primarily from long-range air transport sources, whereas Cu was the result of local sources. On the northern Norwegian mainland, gradients for Pb, Cd, and Hg decreased south to north, while Cu and Zn had the opposite trend. The range of mean values for Cu, Zn, Pb, Cd, and Hg were 25-110 μ g/g, 80-175 μ g/g, 17-85 μ g/g, 0.45-0.75 μ g/g, and 0.06-0.20 μ g/g, respectively. On Spitsbergen and Bear Island, the average concentrations of Pb (25- 90 μ g/g) and Cd (0.5-1.3 μ g/g) were generally higher than those found on the northern Norwegian mainland, but the mean concentrations of Hg (0.062-0.098 μ g/g) were lower.

Johansson (1989) reported similar trends for metals measured in 54 lakes in Sweden (14 were located within the AMAP region). Gradients for Pb, Zn, Cu, and Cd all decreased south to north. Concentrations in the surface layer were 3-25 μ g/g, 5-40 μ g/g, 20-65 μ g/g, and 0.07-1.3 μ g/g for Pb, Cu, Zn, and Cd, respectively. In the south, vertical gradients of the metals along cores suggested enrichment factors of about 50, 7, 4, and 2 for Pb, Cd, Zn, and Cu, respectively. In the north, Zn and Cu showed no vertical gradient and the enrichment of Pb and Cd in the surface sediment was only about a factor of 2.

Verta et al. (1989) provide profiles for Pb, Zn, Cd, and Hg in sediment cores from 18 small headwater drainage and seepage lakes in Finland. The trends are in general agreement with the data of Skotvold et al. (1996) and Johansson (1989), namely a gradient decreasing south to north.

Lockhart et al. (1995) provide data for Hg in dated cores from ten lakes in Arctic Canada. Recent fluxes of Hg varied from 5 μ g/m²/y to approximately 50 μ g/m²/y compared with fluxes of 0.7 to 31 μ g/m²/y estimated from the deepest portions of the cores. The enrichment factors for Hg (ratio of concentration of Hg at top of each core divided by that at the bottom) ranged from 1.1-7.0 (mean, 2.4). These values are consistent with data reported in temperate areas of North America.

Viewed as a whole, the data suggest a widespread and continuing input of Hg into Arctic sediments. Indeed, the results show that for Finnish and Canadian sediment cores are remarkably similar despite the wide geographical separation between the sampling areas. This may be related to the rather unique properties of Hg as a metal. Specifically, it is highly mobile, particularly in the gas phase, and is capable of being reemitted from sediment and water. Successive deposition/reemission cycles combined with decreasing average temperatures at higher latitudes could lead to enrichment of Hg in Arctic regions.

Rognerud et al. (1993) provide data on historical trends on accumulation rates of Cu, Cd, Co, Ni, Pb, and Zn for two Norwegian lakes (Dalvatn and Durvatn) from cores collected during a joint Norwegian – Russian expedition in 1992. The lakes are located downwind of Russian smelters. Rognerud et al. (1993) showed that the sediment accumulation rates for each lake were constant during the past 50-60 years. Consequently, any changes in the rate of accumulation of individual metals should be attributable primarily to changes in atmospheric deposition. The ratio between the accumulation rates of metals in the most recent settled sediment and a reference sediment from the 1920s prior to the start of smelting operations indicate that the accumulation rates of Ni, Cu, and Pb have increased by factors of 2-4. The increase in Pb accumulation rates are thought primarily to be related to the general increase of Pb in the atmosphere of the northern hemisphere (Norto and Kahl 1991), but a small contribution is related to the smelters (Hagen et al. 1991).

Dahl-Hansen and Evenset (1995) measured relatively low concentrations of Cu (4.4-12 μ g/g), Zn (76-114 μ g/g), Hg (0.026-0.092 μ g/g), Cd (0.11-0.89 μ g/g), and Pb (2.9-8.5 μ g/g) in Lakes Nyulay, Kotyol, and Kapylty in Arctic Russia. Far greater pollution by metals is seen in areas of Arctic Russia influenced by metallurgical complexes such as those on the Kola Peninsula and around Norilsk. In these areas, the concentrations of Ni, Cu, Co, Cd, and Hg in the surface sediments of lakes located up to 40 kilometers from the source of pollution exceed

background values by 10-380 times (ACOPS 1995). As lake sediments are excellent storage reservoirs for metals, enhanced metal concentrations are likely to exist in these areas for many decades.

3.1.2.1.4. Marine Environment

3.1.2.1.4.1 Water

Contaminants in seawater, including heavy metals, have been given low priority in the AMAP programs because the concentrations of most pollutants in seawater are close to the detection limits of most laboratories and are very costly to measure.

Mercury: Very little information is available on the concentrations of Hg in Arctic Ocean water. Weiss et al. (1974) reported mean Hg concentrations of 11-22 ng/L (0-400 m) for the southern Beaufort Sea. Thomas (1983) reported background values of 1-15 ng/L for total Hg in samples from the Beaufort Sea shelf. The dissolved Hg concentrations were strongly and negatively correlated with the concentrations of particulate organic carbon. In Puget Sound (Washington, USA), the Hg concentration was 2-10 ng/L (Bothner and Robertson 1975).

By comparison, the average Hg concentration was 4.1 ng/L in the Gulf Stream (over a depth of 250-4500 m), 8 ng/L (0-750 m) in the Sargasso Sea, and 3-4 ng/L in waters around the United Kingdom (Mukherji and Kester 1979). In the temperate zone in the North Pacific, total Hg was 14 ng/L in surface waters and 2 ng/L between 500 and 5000 m (Miyake and Suzuki 1983). Off the Swedish west coast, the values ranged from 5 to 12 ng/L (Gustavsson and Edin 1985). There appears to be no great difference in the concentration of Hg in oceanic water around the world, except in polluted waters where concentration can exceed the values noted above by an order of magnitude or more.

The older data have, however, been questioned in a recent NATO workshop (NATO-ARW 1996). Current consensus is that total dissolved Hg in unpolluted ocean water is between 0.2 and 1 ng/L with somewhat higher concentrations in coastal waters and in density interfaces where accumulation due to particulate dissolution is enhanced (up to 2 ng/L). Methylated Hg (when it can be measured) is generally less than 10% of the total Hg. Dimethylmercury is often the major methylated species.

Lead: Mart and Nurnberg (1984) reported Pb concentrations in eastern Arctic Ocean water of 15 ng/L at the surface and 3-4 ng/L at 1500-2000 m. Background concentrations for Pb in Beaufort Sea shelf waters appeared to be in the range of <20-40 ng/L (Thomas et al. 1982). Recent seawater analyzes from the Pechora, Kara, and Laptev Seas in Russia show very high Pb levels in the range 160-500 ng/L (Rosgidromet 1995), but these data should be confirmed as they may be unrepresentative. By comparison, Pb levels ranged from 29 to 41 ng/L in the surface waters of the North Atlantic and the Norwegian Sea (Mart and Nurnberg 1984). In the North Pacific between Hawaii and California, Pb in surface water ranged from 5 to 15 ng/L, decreasing with depth to approximately 1 ng/L (Schaule and Patterson 1981). Lead concentrations are consistently higher in surface water than in deeper layers. In coastal waters, concentrations can reach 50 ng/L, and can be much higher in heavily polluted waters (Burnett et al. 1980).

Probably more than any other metal, Pb has been enriched, particularly in the northern hemisphere, because of anthropogenic inputs. An indirect estimate of 0.6 ng/L of Pb in prehistoric oceanic surface water has been reported (Schaule and Patterson 1981). Surface waters of the North Pacific contain ten times more Pb now, and those of the Arctic even more, than in prehistoric times. In the Greenland Ice Sheet, modern Pb concentrations are 300 times higher than those that are characteristic of prehistoric times (Ng and Patterson 1981). However, a recent decrease has been observed due to the use of unleaded gasoline (Boutron et al. 1991, 1995, Hong et al. 1994).

There is clear evidence that mining in the Arctic has increased local Pb concentrations in seawater. At the Black Angel Pb-Zn mine at Maarmorilik in northwest Greenland, which operated from 1973 to 1990, Pb levels in seawater as high as 200 000 ng/L were reported in bottom fjord water, where mine tailings were discharged (Asmund 1992a, 1992b). In surface waters close to the mine site, Pb concentrations up to 42 000 ng/L were measured (Asmund et al. 1991). Close to a cryolite mine in Ivittuut in south Greenland (closed in 1986), Pb concentrations in surface waters reached 18 000 ng/L (Asmund et al. 1991). Lead concentrations in the range of 40-100 ng/L were reported by Thomas et al. (1984) for the waters of Strathcona Sound, a northern Baffin Island fjord at the Nanisivik Pb-Zn mine; these concentrations were approximately one to two orders of magnitude higher than open ocean background concentrations.

3.1.2.1.4.2 Sediments

Mercury: Reported values for the concentration of Hg in Arctic sediments range from below the detection limit of 0.01 mg/kg in subsurface sediments from Roroy and Skrova, Norway, and 0.009 ± 0.004 mg/kg in sediments from Uummannaq, Greenland to 0.243 ± 0.043 mg/kg in southern Beaufort Sea continental shelf sediments and 1.6 ± 1.2 mg/kg in sediments from Strathcona Sound, Canada collected in 1975. The latter very high value may reflect the influence of the nearby sulfide ore body (the Nanisivik mine started operation in October 1976). No dependence of Hg concentration on source rock geology was detected from Greenlandic sediments (Loring and Asmund 1996), where the average concentration was 0.045 ± 0.045 mg/kg.

Hg concentrations generally correlate negatively with sediment grain size. On the Beaufort Sea shelf, Hg ranges from 0.01 to 0.1 mg/kg in sediments containing 0% to 100% mud (Macdonald and Thomas 1991). Similarly, Loring (1984) reported a grain size-Hg dependence for sediments from the Canadian Arctic in the concentration range 0.04±0.014 to 0.07±0.039 mg/kg. There are several data sets that indicate widespread accumulation of Hg in surficial Arctic sediments (Lockhart unpubl., Dietz et al. 1997b, OSPARCOM/NIVA unpubl. data, Gobeil and Macdonald unpubl.). The enrichment of Hg occurs in the upper 2 to 10 cm of the sediments, even at the North Pole. This phenomenon could indicate global scale input of Hg to the marine environment in recent times. More comprehensive investigations of Arctic sediments are required, however, before definite conclusions can be drawn about the nature and source of the observed enrichment.

Lead: The lowest reported levels are $1.4\pm0.2 \text{ mg/kg}$ (Thomas et al. 1983, cited in Muir et al. 1992) in 124 sediment samples from the southern Beaufort Sea continental shelf, and 4.17 ± 0.72 in 26 sediment samples from Hudson Bay. In sediments from the Beaufort Sea shelf, concentrations ranged from 9.1 to 20.1 mg/kg, corresponding to 0% and 100% mud, respectively. The highest concentration of Pb in Arctic sediments far from point sources is $53.0\pm2.2 \text{ mg/kg}$ in the surface layer of three sediment cores from Ålesund, Norway. This is probably of anthropogenic origin as the same cores had a concentration of only 22 mg/kg at a depth of 40-45 cm. The highest reported concentration in sediments believed to be natural is 43.5 ± 2.2 in sediments from southern Beaufort Sea continental shelf. The wide range of values evident in the southern Beaufort Sea is probably related primarily to sediment texture and mineralogy. The data for the Russian Arctic (7.7 ± 1.5 to 17 ± 5 mg/kg) could be low by 0% and 61% based on the results of the Norwegian-Russian laboratory inter-calibration exercise (AkvaplanNIVA 1996).

Loring and Asmund (1996) found that the concentrations of Pb in Greenlandic sediments from areas containing tertiary volcanic rocks and from the Nagssugtoqidian mobile belt were lower (12-14 mg/kg) than in sediments from other geological provinces (21-24 mg/kg). In contrast, Loring (1984) found no dependence of concentrations on mud content for a suite of Arctic sediments that contained 18 ± 6 to 21 ± 8.5 mg/kg of Pb. The lack of enrichment of Pb in the upper one or two centimeters of Arctic marine sediments suggests that long-range transport of aerosol particles is not an important process in the Arctic.

3.1.2.2. Biotic Media

3.1.2.2.1. Terrestrial Environment

No data were found for the metal content of microorganisms in the Arctic terrestrial ecosystem. The most abundant data on the metal content of vegetation in the Arctic exists for mosses and lichens, particularly in Scandinavia. However, a limitation of the technique is that factors other than air pollution contribute significantly to the element distribution observed in mosses, thus posing the greatest difficulty for interpreting and comparing results. Therefore, these data are not included here.

There is only a limited database on metal concentrations in terrestrial bird species in the Arctic.

The most comprehensive data set for metals in Arctic mammals exists for caribou/reindeer (*Rangifer tarandus*). To lesser extents both spatially and temporally, measurements are compiled for a variety of other mammals.

The only substantial temporal data set examining metal concentrations in caribou is provided by the Swedish Environmental Monitoring Programme (SEMP 1995) through which samples of reindeer have been collected continuously since the early-1980s in three districts along the Swedish easternmost mountain chain, encompassing the Saami villages of Gabna, Laevas, Girgas, Iran, Ran, Handölsdalen, and Mittådalen. Analysis of the accumulated data indicates that during the past ten years, Cd, Pb, and Hg concentration in the muscle and liver of Swedish reindeer show no significant log-linear or linear change. Furthermore, SEMP (1995) calculated that the

number of years required to detect an annual change of 5% ranged between 10 and 21 years depending upon element and tissue.

3.1.2.2.2. Fresh Water Environment

In general, there are few, or no, data on the content of metals in fresh-water micro-organisms, algae, plants and invertebrates.

3.1.2.2.2.1 Fish

There are few instances of comprehensive data for a given fish species that will allow comparison of values across the Arctic or as a time series. A possible exception is Hg, for which the largest set of values exists. This emphasis on Hg in the environment and in the food chain reflects the fact that it clearly bioaccumulates and that the effects Hg has on fish and on human consumers are well documented.

Data for Hg in the muscle tissue of Arctic char (Salvelinus alpinus), whitefish (Coregonus spp.), and burbot (Lota lota) are relatively consistent for all Arctic regions.

The recent analysis of archived samples of pike (Esox lucius) muscle tissue in Sweden under the Swedish Environmental Monitoring Programme provides the most comprehensive time series for Hg in Arctic freshwater fish. A total of 237 samples of pike caught in 21 different years between 1968 and 1996 were analyzed. Geometric mean values (weight adjusted) varied between 194 and 459 ng/g ww. No clear temporal trend is evident in the data; variations around a mean for the entire dataset of 306 ng/g ww probably represent natural year-to-year fluctuations and analytical variability (Olsson pers. comm.).

The highest concentrations of Hg in fish are not necessarily correlated with the presence of high Hg concentrations in sediments. There are several reasons for this. First, Se is known to reduce the uptake of Hg by fish (and biota in general) so the Se content of sediments must be factored into any model involving correlation. Second, the concentration of calcium and magnesium affect the bioaccumulation of Hg by altering competition equilibria. Finally, pH and buffer capacity have an impact on the bioaccumulation of Hg by fish because humic substances can form strong complexes with Hg thereby effectively reducing Hg availability (bioaccumulation of metals other than Hg is usually enhanced in acidified low calcareous lakes).

The fish from Arctic Canada generally have the highest body burdens of Hg found in the Arctic. This is thought to reflect the widespread naturally elevated baseline for Hg in the Canadian Arctic and may be related to Hg-organic matter associations). Fish exceeding the guideline level of 0.5 μ g/g for human consumption occur most frequently in Arctic Canada and Greenland (range of mean muscle values 0.17-1.0 μ g/g (n = 86)).

3.1.2.2.2.2 Birds

Based upon a limited number of measurements, the bioaccumulation of metals by aquatic birds appears to be at least an order of magnitude less than that of terrestrial birds. The distribution among tissues is the same as for terrestrial birds, however: kidney > liver > muscle. There are too few data to delineate any spatial, temporal, or species trends. The largest dataset for metals in aquatic birds is for a group of ten species at various locations in Arctic Canada (NWT, Yukon, Quebec, and Labrador, CWS 1996). For Cd, Pb, and Hg, many values were at or near the detection limit. Ranges for values exceeding the detection limit in breast muscle tissue were as follows: Cd, 0.02-0.87 μ g/g ww (n = 134); Hg, 0.2-1.93 μ g/g ww (n = 158); and Se, 0.04-2.1 μ g/g ww (n = 418).

3.1.2.2.2.3 Mammals

The data for metals in freshwater mammals consist of Pb, Cd, and Hg measurements in the hair of ringed seals living in Lake Saimaa, Finland (Hyvärinen and Sipilä 1984). Although Lake Saimaa is located just outside the AMAP assessment area, it is nonetheless relevant to include this dataset here because the boreal environment of this lake is similar to that for lakes in the AMAP area. Ringed seals exist in the Arctic and the data provide one of the very few observations of possible links between exposure to metals and biological effects.

Hair samples collected around 1983 had the following ranges of concentration means (n=32): 0.5-0.7 μ g/g dw, 3.6- 8.5 μ g/g dw, and 3.2-20.7 μ g/g dw for Cd, Pb, and Hg, respectively. According to data obtained for museum samples, the concentration of Hg in the hair of adult ringed seals was about the same as found by Hyvärinen and Sapilä (1984) in yearlings. In 1965, the concentration in hair was approximately 50 μ g/g. The authors speculated that the sharp decline in the ringed seal population of Lake Saimaa during the 1960s and 1970s could be related to

the insufficient availability of Se in the lake, which made the seals more susceptible to the toxic effects of Hg and premature (still) births.

3.1.2.2.3. Marine Environment

Few data have been published on heavy metals in Arctic algae. Data, primarily for Cd and to a lesser extent Pb, Hg, and Se, are available for five intertidal species in Greenland, but the geographical extent is very limited. Additional data in algae are available only on Pb for Canada and Norway.

3.1.2.2.3.1 Invertebrates

Data are available mainly for bivalves, amphipods, and decapods.

Mercury: Most data are for bivalves from Greenland and Norway and for decapods from Greenland. In bivalves from Greenland, Hg levels are similar among species and regions. In soft tissue, the range is from 0.011 μ g/g in a cockle to 0.020 μ g/g in an Iceland scallop. The 1994 AMAP data from Greenland fit well into this range with levels from 0.014 to 0.017 μ g/g, and similar values are found in blue mussels from Iceland. Concentrations of Hg in bivalve samples from Norway are similar (< 0.009-0.033 μ g/g) to those found in the Greenland samples.

Although tissues and species are not similar, Hg values for bivalves from Russian and Canada are similar to those found in Greenland and Norway. In general, Hg concentrations in bivalves are low compared with those found in fish, seabirds, and marine mammals from the Arctic. Although higher than in bivalves, Hg levels are low in crustacea. In decapods from Greenland whole animal values range between 0.023 and 0.258 μ g/g, with the lowest levels occurring in small Bikini prawns from Avanersuaq and the highest in large deep-sea prawns from Baffin Bay.

Lead: In bivalves, most data are available for the blue mussel, which has been selected as an indicator species in the AMAP program. Data from Greenland show that the Pb concentration in blue mussels increases slightly with increasing shell length (Riget et al. 1996). Older data (1983-1990) from southwest Greenland show Pb concentrations from 0.142 to 0.476 μ g/g. Recent AMAP data from Greenland are similar to the lowest value of the older Greenland data, ranging from 0.072 to 0.155 μ g/g at Qeqertarssuaq (central West Greenland) and from 0.124 to 0.188 μ g/g at Nanortalik (south Greenland). Norwegian samples of blue mussels collected from 1984 to 1993 are in the range of 0.101-0.483 μ g/g, which is similar to the range observed in the same period in Greenland. Old Icelandic data (1978) are relatively high (0.56-0.805 μ g/g), whereas Icelandic data from 1990 to 1992 are lower (range 0.019 to <0.195 μ g/g). Older Greenland data (1984-1985) for other bivalve species are in the same range (0.069-0.186 μ g/g) as the 1994 AMAP data for blue mussels.

At the Black Angel mine in northwest Greenland, Pb concentrations in blue mussels were elevated over background levels in a wide area (up to 30 km from the mine), with values exceeding 1000 g/g nearest the mine (Johansen et al. 1991). Local elevations were also observed in blue mussels at the cryolite mine in south Greenland (Johansen et al. 1985), in three bivalve species at the Pb-Zn mine in East Greenland (Hansen and Asmund 1986), and in clams at the Nanisivik Pb-Zn mine at Strathcona Sound in the Canadian Arctic (Asmund et al. 1991). At the Black Angel mine, local elevations of Pb were also observed in the deep-sea prawn (Johansen et al. 1991).

3.1.2.2.3.2 Fish

Mercury: Data are available for Hg in muscle, liver, kidney, bile, spleen, and gonads of fish; most data are for muscle and liver. In muscle, the concentrations of Hg are generally in the range of 0.01-0.1 μ g/g. In some species, such as Greenland halibut, two eelpout species, Arctic cod, and shorthorn sculpin, Hg concentrations are higher, between 0.1 and 0.2 μ g/g. The highest values in fish muscle were found in Arctic cod from Kong Oscars Fjord in East Greenland (0.882 μ g/g). Concentrations in liver were lower than in muscle, between 0.01 and 0.06 μ g/g for most species. The highest value of Hg in liver tissue was found in samples of Arctic cod from Upernavik (0.192 μ g/g).

Geographical differences: There are very few fish species that have been sampled on a circumpolar scale and analyzed for Hg. An exception is Arctic cod for which the concentration of Hg in muscle tissue is available for the eastern Canadian Arctic, West and East Greenland, Jan Mayen, the Greenland Sea, and the Barents Sea. The concentrations of Hg in fish from the Barents Sea and Greenland Sea are lower than in the other areas; however, the sampling periods differed considerably among areas, which may have influenced the levels found. Some of the

highest values were found in fish from the northern areas of eastern Canada as well as western and eastern Greenland.

Lead: Generally, Pb concentrations in fish muscle are very low (< $0.002-0.05 \ \mu g/g$). Where muscle and liver have been analyzed, the levels in liver are higher than those in muscle – up to 0.2 $\mu g/g$, mainly in fish species from Orkdalsfjorden/ Trossavika, Norway that may be contaminated.

3.1.2.2.3.3 Seabirds

Mercury: Hg concentrations generally decrease in the order liver > kidney > muscle. Levels in liver range from 0.1 to 3 μ g/g. Kidney tissue levels were intermediate between the values in liver and muscle, ranging from 0.053 μ g/g in one-year-old kittiwake to 2.33 μ g/g in older glaucous gulls.

Lead: Several seabird species were analyzed for Pb, mostly from Greenland, Canada, and Norway. Few data are available from Iceland and Russia.

Pb values are below 0.4 μ g/g, except for one specimen of king eider (Somateria spectabilis) from Holman Island, Canada that contained 1 μ g/g of Pb. The levels in liver and kidney ranged from <0.009 to 0.8 μ g/g. The highest concentrations in kidney are found in seabirds from Svalbard.

3.1.2.2.3.4 Marine Mammals

A substantial amount of data is available on the concentrations of metals in Arctic marine mammals. Because Cd and Hg bioaccumulate strongly and because Hg biomagnifies, much more emphasis has been placed on obtaining body burdens of metals in biota of higher trophic levels.

Lead: Data on tissue concentrations of Pb in marine mammals have been produced for ten species including polar bear. Most data are from Canada and Greenland, some are from Alaska and Norway, and almost none are from Iceland and Russia. Concentrations of Pb range from below detection (< 0.010 μ g/g) to 0.083 μ g/g in tissues of ringed seals. For other pinniped species (harp seal, Pagophilus groenlandicus; fur seal, Callorhinus ursinus; and walrus), they range between 0.010-0.328 μ g/g. A recent study by Wagemann et al. (1996) includes data for ringed seals, beluga whales (Delphinapterus leucas), and narwhals and shows no Pb levels higher than 0.083 μ g/g.

Mercury: Mercury data from eight seal species, eight whale species, and polar bears have been compiled, showing, as for Cd, considerable differences among tissue types. Concentrations generally decrease in the order liver > kidney > muscle. In polar bears, however, the highest Hg levels are found in the kidney.

Concentrations of Hg in seal and whale muscle frequently exceed 0.50 μ g/g ww, particularly in older individuals because of the accumulation of Hg with age. The highest mean Hg concentrations (0.72 μ g/g in muscle, 32.6 μ g/g ww in liver) are found in ringed seals in the western Canadian Arctic (Wagemann et al. 1996). Up to 219 μ g/g have been measured in the liver tissue of a ringed seal from Sachs Harbour. This is also the population where the highest mean values have been recorded (Wagemann et al. 1996). Very high levels, 143 μ g/g on average, have been reported for bearded seal from the Amundsen Gulf (Smith and Armstrong 1975, 1978). Mercury can be transferred from the mother to the fetus during gestation, which is apparently not the case for Cd (Wagemann et al. 1988). Almost all the Hg in muscle tissue is present as methylmercury, whereas in liver tissue methylmercury seldom exceeds 2 μ g/g, even when the total Hg concentration is high (Dietz et al. 1990). In the kidney tissue of adult seal and whale, organic Hg is 10-20% of total Hg, whereas in polar bears it is 6% (Dietz et al. 1990). This fraction is consistently higher, up to 70%, in young seals and walrus (Born et al. 1981, Wagemann et al. 1988, Dietz et al. 1990). The concentration of Hg increases in marine mammals with age.

Geographical trends: The increasing trend of Hg in ringed seal liver from eastern to western Canada is supported by a recent study of Wagemann et al. (1996), but no similar trend was found in kidney and muscle. Juvenile ringed seals from Jarfjord in Norway had lower Hg concentrations in liver and kidney than did ringed seals from Canadian and Greenland waters. Ringed seals from the Gulf of Bothnia and Gulf of Finland, on the other hand, exceed even the highest values reported for the Arctic (Helle 1981, Perttilä et al. 1986, Frank et al. 1992). Higher levels in seals from northwestern European waters than in those from the Arctic are also indicated by studies involving grey seal (Halichoerus grypus) and harbour seal (Phoca vitulina) (Law et al. 1991, Frank et al. 1992), and in harbour porpoise from the two areas (Paludan-Müller et al. 1993). In seals found dead off the coast of the Netherlands, the Hg concentration in liver was extremely high, 257-326 μ g/g (Koeman et al. 1972), most likely due to anthropogenic sources. The high Hg levels in the western part of the Canadian Arctic have also been documented in beluga whale tissue (muscle, liver, and kidney) and are most clearly seen in liver. The observed geographic trend has also been documented for Hg levels in polar bear hair (Eaton and Farant 1982, Renzoni and Norstrom 1990, Born et al. 1991). Norstrom et al. (1986) suggested that the differences found between western Arctic Canada and eastern Arctic Canada were most likely caused by higher Hg levels in the ringed seal food chain caused by higher natural levels in the sediments (and consequently in the lower food chain) of the Melville Island area. The evidence presented above indicates a geographic trend in ringed seal liver, but these differences are not apparent in kidney and muscle. Too few data are available to evaluate geographical trends in ringed seal blubber, which is the preferred food of polar bears.

3.1.2.3. Summary of Trends

3.1.2.3.1. Spatial trends

The concentrations of heavy metals in surface deposition around the sources, e.g., on the Kola Peninsula, decrease by between one and two orders of magnitude within 10-100 km from the emission source. The concentrations within the large area of the High Arctic are uniformly distributed, varying by a factor of 2-3.

The concentrations of trace elements in marine sediments are dependent on local geology, particle size, the amount of organic matter, and anthropogenic influence. The back ground geographical distribution of Pb, Cd, Hg, and Cu in marine sediments is related to the geological provinces of the Arctic.

Regional geographical differences in metal concentrations of benthic flora and fauna as well as in those of fish are not very apparent. Seasonal and local trends for some metals for some stationary marine species are larger than regional differences in baseline data. No geographical differences can be observed in fish species. Mercury levels for ringed seals, beluga whales, and polar bears, have proven to be highest in the western Canadian Arctic, decreasing toward south and east. Geology, food constitution, and growth processes linked to temperature are possible explanations for these differences.

3.1.2.3.2. Temporal trends

The concentration of heavy metals measured in sub-arctic air has decreased during the last two decades. All the heavy metals show strong seasonal variation in the High Arctic.

Mercury in Arctic sediments shows an increase over time, indicating a widespread regional process. As the anthropogenic fluxes does not show the same pattern, further investigations are needed before firm conclusions can be drawn.

Temporal trend data are scarce in Arctic biota. There is some evidence of Hg increasing by a factor 2-3 in some marine mammals within the last two decades. Only liver, and in certain cases kidney, shows such increase. It remains uncertain, however, whether this is a real increase or reflects year-to-year variation. Mercury concentrations in human and seal hair from the 15th century are 2-3 times lower than present-day samples.

3.2. TOXICOLOGY

This section on human health describes the levels of contaminants in humans and their effects on human health. It has not been written as an overview of the general health of the peoples of the Arctic. Rather, the section is an evaluation of the current knowledge of the consequences to Arctic peoples of environmental exposure to priority contaminants. The material is presented in two parts, first an overview of harmful effects followed by specific information from each country, National Reports.

Many factors contribute to health and illness of human populations living in the Arctic; these include socioeconomic conditions, availability of health services, societal and cultural factors, individual lifestyles and behaviors, and genetics. Environmental contaminants, although just one of these factors, can have a significant influence on human health. There are numerous population groups in the Arctic of which many are indigenous. These groups may be more threatened by contaminants than immigrants from the south since they generally live their whole lives in the Arctic and subsist largely on local food. This chapter, however, deals with the contaminant-related health implications for all Arctic inhabitants.

Observations from most parts of the Arctic ecosystem indicate that it is far from pristine. Pollution, both local and due to long-range transport of contaminants, is prevalent at levels which pose a concern for human health. For some indigenous populations of the Arctic, blood mercury levels and concentrations of persistent organic

pollutants in blood and fat are 10-20 times higher than those found in most temperate regions. Although there are currently no confirmed diagnoses of illness in these populations, which can be causally linked to contaminants, the high concentrations of toxic substances are a cause for concern.

Human exposure to pollutants through the diet is of central concern in the Arctic. Many of the relevant pollutants, including mercury and persistent organic pollutants, are biomagnified through Arctic food chains. Because humans in the Arctic traditionally feed on other carnivores such as seals and whales, they are at the top of food chains that include other top-level predators. Thus, indigenous peoples consuming traditional diets are more likely to be exposed to higher concentrations of these pollutants than populations elsewhere in the world. However, the traditional diet is often important for the social, spiritual, and cultural identity of indigenous peoples. The negative attitude that can develop because of, e.g., bans on consumption of local foods, can disrupt this identity and can have other effects on health. Furthermore, the traditional diet including marine mammals, birds and fish is rich in vitamins, minerals, protein and fatty acids. The n-3 fatty acids that are widely marketed in the south as 'fish oils' are known to prevent arteriosclerosis. This component of the diet is probably one of the reasons for the very low mortality from ischemic heart disease among indigenous Arctic populations.

The effects of environmental pollutants on health are most often subtle, since they usually occur at concentrations that are not expected to result in acute toxic symptoms. What might be expected are long-term, possibly transgenerational, effects on the neurologic and reproductive systems. These effects are confounded by many other factors and can be difficult to identify even in long-term epidemiological studies in large populations. In the small Arctic populations, it may prove impossible to conduct epidemiological studies with enough power to detect the effects of concern. It is therefore important to recognize that the paucity of health data or the absence of overt illness or malfunction does not imply that the exposure of peoples in the Arctic to contaminants is without effects. Even biochemical changes in blood and tissue must be regarded as undesirable effects of pollution.

3.2.1. Overview of Harmful effects

Availability of information on contaminant levels in the tissues of northern residents is very recent, and the quantity is increasing rapidly. While trends are difficult to determine, there is a clear indication in the National Reports and in the comparison of data from an international study that a number of persistent substances are significantly elevated in the tissues of several Arctic ethnic groups.

Human populations are always exposed to mixtures of POPs in the ecosystem, never to single compounds. Hence, toxicological risk assessments that make use of animal test data on individual chemicals rather than mixtures of chemicals, and their applicability to humans, is frequently in question. The actual levels of individual contaminants in the mixture of POPs to which populations are exposed vary by region (because of differences in environmental occurrence and food consumption patterns), making comparisons of possible effects between populations very difficult. Human exposures to POPs are usually to lower levels than those chosen for animal studies. Humans are also typically exposed to these contaminants over their entire lifetime, commencing with conception, and not merely for the limited life stages chosen for most animal studies. Finally, confounding factors of lifestyle, diet, age, reproductive status, gender, and general health also affect how individual POPs will influence the onset of disease or adverse effects.

Metals can occur in ecosystems in organic and inorganic form as well as in different oxidation states. These factors will affect the absorption, metabolism and toxicity of metals, making information on their form and speciation crucial for realistic risk estimates.

The toxicity of metals is often due to their interference with important sites in cellular biochemical systems, such as the sulfhydryl groups in enzyme systems. This interference often results in cell death. Metals may also compete with essential elements as enzyme co-factors, creating a toxic response that is manifested as a deficiency of an essential metal. For this reason, it is also important to consider the status of essential elements when evaluating an exposure to the toxic metals. There is an abundance of data on such interactions (for a review see Nordberg et al. 1986). Mercury, lead, and to a lesser degree cadmium, can all cross the placenta and can affect the developing fetus.

3.2.1.1. <u>DDT/DDE/DDD</u>

DDT and its metabolites are stored in fatty tissue and are excreted very slowly, primarily via urine and feces. Because DDT and its metabolites are found in breast milk, 'excretion' also occurs during breast-feeding. DDT can readily cross the placenta.

Acute lethal (LD₅₀) oral doses in test animals range between approximately 115-800 mg/kg bw (body weight). Noobserved-adverse-effect levels (NOAELs) for chronic exposure to DDT, DDE and DDD for most mammalian test species range between 10-100 mg/kg bw/d for respiratory, cardiovascular, gastrointestinal, hematological, hepatic and renal outcomes. However, the more serious effects (neurological, developmental, reproductive and carcinogenic) have lowest-observed-adverse-effect level (LOAEL) values more in the 8-50 mg/kg bw/d range for chronic exposure (ATSDR 1994a). Some forms of DDT are considered to have weak estrogen-like responses with potencies relative to estradiol of between approximately 0.01 and 0.0001 (Soto et al. 1992). This estrogenic effect is probably responsible for DDT impacts on reproduction in animals (ATSDR 1994a). DDE has recently been shown to be a potent androgen receptor antagonist (Kelce et al. 1995).

In humans, acute lethal exposures to DDT are probably greater than 250 mg/kg bw. Very little data on the effect of chronic exposure is available. Long-term exposure of volunteers to amounts up to approximately 0.6 mg/kg bw/d did not lead to any observable neurological signs (Hayes et al. 1956). The World Health Organization (WHO) has proposed a Tolerable Daily Intake (TDI) of 20 μ g/kg bw/d for DDT (including metabolites).

Epidemiology studies have not established an association between DDT exposure and cancer, even though cancer is an outcome of long-term animal-feeding studies. The International Agency for Research on Cancer (IARC) has classified DDT and DDE as 'possibly' carcinogenic to humans based on evidence from animal studies.

Suppression of reflexes in neonates appears to be associated with levels of DDE in breast milk exceeding 4 μ g/g lipid, however, it has not been substantiated that DDE is the causative factor (Rogan et al. 1986). Elevated levels of DDE in human breast milk (\geq 3 μ g/g lipid) have been correlated with a shortening of breast-feeding duration, and inhibition of lactation was hypothesized as the cause (Rogan et al. 1987).

3.2.1.2. Toxaphene

Toxaphene is commonly found in human tissue, however, its historical quantification has been compromised by difficulties in analysis and in the estimation of amounts of the various chemicals that make up toxaphene. As a result, comparisons using published data on toxaphene are difficult.

The lethal oral dose (LD_{50}) of technical toxaphene in rats is between 80 and 90 mg/kg bw (Gaines 1969), however a range of other effects, including death, can follow chronic exposure to levels of 20 to 30 µg/kg bw/d (ATSDR 1994b). Large doses of toxaphene are likely to affect the nervous system (seizures, tremors, convulsions, paralysis and both behavioral and biochemical effects), the liver (enzyme induction) and the kidney (enzyme release, fatty degenerative changes and focal necrosis). Intermediate exposure duration for toxaphene may affect the adrenals, the immune system and fetal development. This research base is mostly limited to short-term and intermediateterm (i.e., less than a lifetime) animal studies conducted with technical grade toxaphene. Although there is one chronic (eighty week) study, there are no suitable studies to confirm the effects of technical or environmentally available toxaphene on human populations. Because so little is known about the effects of long-term exposure to both technical and environmental toxaphene in animals and humans, there is considerable uncertainty over the applicability of a TDI. Canada uses 0.2 µg/kg bw/d. In the USA, the Agency for Toxic Substances and Disease Registry (ATSDR 1994b) has proposed an 'intermediate-duration oral exposure minimal risk level' of 1 µg/kg bw/d. The WHO has not proposed a TDI.

Animal studies suggest that toxaphene is an animal carcinogen. It has been classified by IARC (1987) as 'possibly' carcinogenic to humans. Toxaphene does not appear to be a very active estrogenic mimic (Soto et al. 1992). Recent data, however, indicate that toxaphene is a potent estrogen receptor antagonist (Jørgensen pers. comm.). Furthermore, it has been shown by Arnold et al. (1996), that the potency of the antagonistic effect of toxaphene was greater by an order of magnitude in synergistic interaction with other chlorinated contaminants such as dieldrin and endosulfan.

Toxaphene is readily absorbed. Intakes in Arctic populations are entirely dependent on the type and amount of food consumed. It is likely that the highest levels in food occur in narwhal and beluga blubber in the eastern Canadian Arctic (Kuhnlein et al. 1995a) and in some fish in a small area of the western Canadian Arctic. Toxaphene is known to be transported northward from past high-use areas in the southern USA. Measurements from other regions of the Arctic are few. Absorbed toxaphene is readily metabolized and excreted (90% in 24-36 hours), however, some constituents remain in fatty tissues for prolonged periods (ATSDR 1994b). Stern et al. (1992) report that the two most common compounds retained are an octachlorocamphene (T_2) and a nonochlorocamphene (T_{12}). There is no toxicological information available for T2 and T_{12} .

3.2.1.3. <u>Hexachlorocyclohexanes (HCHs)</u>

 γ -HCH (lindane) is the most toxic of the HCH isomers. Excessive exposures can affect the liver, the nervous system, the kidney, the reproductive system, and perhaps the immune system. IARC (1987) classifies it as 'possibly' carcinogenic to humans. No effects have been reported in populations exposed to low-level environmental concentrations. Exposure levels for HCH via consumption of store-bought food in several countries were approximately 0.005 µg/kg bw/d (α -HCH), 0.0003 µg/kg bw/d (β -HCH) and 0.03 µg/kg bw/d (γ -HCH) (Gorchev and Jelinek 1985). The TDI for total HCHs is 0.3 µg/kg bw/d (WHO 1991a).

HCHs, especially β -HCH, accumulate readily in fatty tissues and are excreted slowly via feces, breast milk and urine (WHO 1991a). Levels of α -, β - and γ -HCH in breast milk in the general populations of Europe, Canada and the United States are in the ranges 10- 40 ng/g lipid, 10-500 ng/g lipid, and <1-10 ng/g lipid, respectively. The most recently measured 'background levels' of HCHs in blood, serum, plasma, milk and adipose tissues are relatively low compared to values reported during the 1960s and 1970s and much lower than levels reported from countries with extensive current usage. Because of its persistence, β -HCH is found at the highest level of the four isomers reported.

The exposure of babies, resulting from β -HCH concentrations found in breast milk, has been identified as a matter of concern by the WHO (1991a), but not as a reason to stop promoting breast-feeding. The high β -HCH levels that have been found in some breast milk samples in countries using HCH, indicate that some infants may exceed the TDI of 0.3 µg/kg bw/d, temporarily and locally. The β -HCH concentrations in the blood of babies lie within the same range as those in the mothers.

3.2.1.4. <u>Mirex</u>

The acute lethal (LD_{50}) oral toxicity of mirex ranges from 365 to 3000 mg/kg bw in laboratory mammals. The effects of long-term, low-level exposure to mirex have not been extensively studied; the primary organs affected by mirex in laboratory species are the liver (at 50 µg/kg bw/d), kidneys, eyes and thyroid (IPCS 1984). Mirex is considered a 'possible' human carcinogen (IARC 1987) and also has fetotoxic and teratogenic effects on laboratory species in the 1-6 mg/kg bw/d range. There is no WHO TDI, however, the Canadian provisional TDI is 0.07 µg/kg bw/d (Health Canada 1996). Mirex is readily absorbed and stored in fatty tissues. Metabolism to photomirex is slow and elimination is mainly via feces and breast milk.

Because of its persistence and accumulation in the food chain, mirex levels in breast milk are above average for communities consuming high amounts of fish and marine-bird eggs (Dewailly et al. 1991).

3.2.1.5. Chlordane, oxychlordane and cis- and trans-nonachlor

The acute lethal (LD_{50}) oral dose of technical chlordane is between 127 and 430 mg/kg bw in rats (ATSDR 1993a). High dose exposures affect the neurological and immune systems (no-effect levels are 4-6 mg/kg bw/d). Long-term exposure is likely to cause cellular changes in the liver at levels of approximately 0.5 mg/kg bw/d (WHO 1984a). Chlordane has been classified as a probable human carcinogen by the US Environmental Protection Agency (EPA) based on tumor identification in mice (LOAEL of 3.9 mg/kg bw/d for mice). The TDI for chlordane is 0.5 μ g/kg bw/d (WHO 1984a). There are few estimates of intakes for chlordane and its metabolites. US estimates indicate that only very small amounts are consumed, 0.002-0.005 μ g/kg bw/d (Gunderson 1988).

Chlordane and its related compounds accumulate in fat and are found in human tissues. In general, only small amounts of chlordane are found in tissues. However, they often contain relatively larger (one or two orders of magnitude higher) amounts of trans-nonachlor and the metabolite oxychlordane. Excretion of chlordane is primarily through feces and breast milk.

3.2.1.6. Dioxins (PCDDs) and furans (PCDFs)

Polychlorinated dibenzodioxins and polychlorinated dibenzofurans are two structurally similar families of compounds that include 75 congeners (different compounds) and 135 congeners, respectively. These compounds enter the Arctic ecosystem almost exclusively via long-range atmospheric transport. Seventeen members of these two families of chemicals are highly toxic and contribute most to the toxicity of complex mixtures of dioxins and furans. The seventeen more toxic congeners in both families have chlorine substitutions in the 2, 3, 7 and 8 positions. These more toxic congeners cause a wide range of deleterious effects in laboratory animals, these effects varying significantly between species (Environment Canada and Health Canada 1990).

The acute lethal (LD₅₀) oral dose for 2,3,7,8-TCDD in mammals varies almost 10 000-fold (0.6 μ g/kg bw for guinea pigs and 5051 μ g/kg bw for hamsters). Common signs of acute and chronic toxicity in animals include loss of body weight and thymic atrophy. Longer-term exposure to doses below the LD50 can lead to discoloration and thickening of skin, skin eruptions, hair loss, liver damage, hematological changes and immune system dysfunction (IPCS 1989). Exposure of laboratory animals during gestation can lead to developmental deficits and altered sexual differentiation (Lindström et al. 1995).

Laboratory rats have developed cancer when exposed to 2,3,7,8-substituted tetrachloro- and hexachlorodioxins (NOEL for 2,3,7,8-TCDD is 1 ng/kg bw/d) (Kociba et al. 1978). IARC (1987) lists 2,3,7,8-TCDD as 'possibly' carcinogenic to humans. Dioxins and furans can directly affect reproduction, with a NOEL for intake of 2,3,7,8-TCDD for this outcome of 1 ng/kg bw/d (Murray et al. 1979). The majority of TDI values are within an order of magnitude (1-10 pg/kg bw/d). The WHO TDI is 10 pg 2,3,7,8-TCDD/kg bw/d (IPCS 1989). Recent studies, however, suggest that single exposures at close to these 'no-effect' doses leads to impairment of development of the reproductive system in male rats (Mably et al. 1992). The antiestrogenic capabilities of TCDD appear to be related to Ah-receptor mediated events (Zacharewski et al. 1991).

The general population is exposed to small amounts of complex mixtures of PCDDs and PCDFs and other organochlorines. An extensive analysis of adipose tissue samples, from a number of countries, has concluded that almost all humans contain TCDD at concentrations up to, and occasionally greater than, 3 pg/g in lipids (Travis and Hattemer-Frey 1991). These levels in the general population have not been associated with disease. In a few incidents, in the USA, Italy and Japan, where workers and others who have been exposed to very large amounts of a limited number of these compounds, individuals have developed chloracne, a skin disorder. There is also evidence that high level exposure to dioxins and furans can cause variations in serum lipid levels, other dermatological effects related to chloracne, microsomal enzyme induction and gastrointestinal alterations (Schulz et al. 1990). Other studies of high-level occupational exposures have found associations with some types of cancer (Zober et al. 1990, Manz et al. 1991, Fingerhut et al. 1993, Bertazzi et al. 1996). Pluim et al. (1993) concluded that in utero and lactational exposures to PCDDs/PCDFs are capable of affecting the hypothalamic, pituitary, and thyroid regulatory system in human infants.

The best documented poisonings by PCDFs in humans are the Yusho and Yu-cheng incidents when rice oil was accidentally contaminated with polychlorinated biphenyls (PCBs) from electrical transformer fluid. The PCBs were heavily contaminated with PCDFs. Investigators have reported low birth weight, early tooth eruption, sensory losses, skin discoloration, swollen eyelids (Kuratsune et al. 1972) and retarded development (Rogan et al. 1986) in infants exposed transplacentally. It is likely that the PCDFs were the cause of many of the reported effects (Rappe et al. 1983, Rappe and Nygren 1984). The mean total intake of PCDFs by the Yusho and Yu-cheng patients has been estimated to be 0.9 μ g/kg bw/d (Hayabuchi et al. 1979) or 3.3 ng 2,3,7,8-TCDD TEQ/ kg bw/d (Ryan et al. 1990). The smallest amount of total PCDFs causing chloracne has been estimated to be 0.16 μ g/ kg bw/d (Hayabuchi et al. 1979).

Average daily intake of PCDDs and PCDFs over a lifetime is similar in most industrialized regions, between 2 and 10 pg 2,3,7,8-TCDD TEQ/kg bw/d for a 60 kg person (Birmingham et al. 1989). Back calculations from human tissue levels in Canada have confirmed this estimated intake, i.e., deriving likely intakes of 1.9 pg/kg bw/d (Environment Canada and Health Canada 1990). The mean PCDD and PCDF concentrations in the breast milk of Arctic and non-Arctic populations are similar: 10-20 pg 2,3,7,8-TCDD TEQ/g lipid (see Ryan et al. 1993 for Canada, Nygren et al. 1986 for Sweden, Schecter et al. 1987 for the USA). Breast milk levels in the Netherlands can be slightly higher: 30 pg 2,3,7,8-TCDD TEQ/g lipid (Koopman-Esseboom et al. 1994a).

3.2.1.7. PCBs (Polychlorinated biphenyls)

The toxicity of PCBs as mixtures is complicated by the varying amounts of the 209 congeners in the mixture and the traces of other contaminants also present (e.g., PCDFs). Individual congeners and mixtures can affect liver function, reproduction, infant birth weights, neurobehavioral development and the immune system and may cause cancer in laboratory animals (ATSDR 1995). The TDI for PCBs is 1 µg/kg bw/d (Health Canada 1996).

Assessing the human health effects of PCBs is very difficult because PCB mixtures typically used in animal studies or identified in accidental poisonings frequently contain traces of contaminants such as PCDFs and undergo extensive 'environmental filtering' prior to human exposures. Many of the effects of exposure to PCBs observed in humans (e.g., Yusho and Yu-cheng incidents) reflect exposure to high levels of both PCBs and PCDFs. PCDFs are believed to be responsible for many of the observed human health effects. From studies following the Yusho and Yu-cheng incidents, the earliest toxicological signs included chloracne. Additional

generalized adverse health effects included hepatomegaly (enlarged liver), bronchitis and peripheral neuropathy (nervous system damage) (Safe 1987). In Yu-cheng, increased upper respiratory tract infection rates were associated with decreased serum IgA and IgM plus increased IgG levels (WHO 1988). Occupational exposures to PCBs generally do not include the PCDF contaminants found in the rice oils, so the effects seen are often different.

A small group of PCB congeners has dioxin-like activity and has been assigned dioxin toxic equivalency factors (Ahlborg et al. 1994). In a number of human tissue samples, such as breast milk or adipose tissue, it has been found that the dioxin-like PCBs contributed a large proportion of the total 2,3,7,8 TCDD TEQ (Dewailly et al. 1992). Because PCB congeners co-exist with dioxins and furans in the environment, ascribing an effect to one or the other contaminant is almost impossible.

Some studies from Japan have found levels of dioxin-like PCBs, such as the congeners CB 77, 126, and 169, up to several orders of magnitude higher than the levels of 2,3,7,8- TCDD in human adipose tissue samples (Tanabe et al. 1987, Kannan et al. 1988, Kashimoto et al. 1989). Results from analysis of human adipose tissue and serum collected in the USA show that concentrations of coplanar PCBs (cf. chapter 6, section 6.1.1.1.1) can be more than an order of magnitude higher than the concentrations of 2,3,7,8-TCDD (Patterson et al. 1994).

Data obtained from epidemiological studies on cohorts of US infants from Michigan (Jacobson et al. 1990, 1992) and North Carolina (Rogan et al. 1986, 1987, Gladen et al. 1988, 1991) suggest adverse neurobehavioral effects from in utero exposure to PCBs (calculated as Aroclor 1260 equivalents). High cord blood concentrations were associated with low birth weight and small head circumference (Jacobson et al. 1990). Birth size among male infants (Inuit) was inversely related to PCB concentration in breast milk of the mother (Dewailly et al. 1993a). Perinatal exposures to PCBs/ dioxins/furans may impair immune responses to infection, as suggested by a 20-fold higher incidence of infectious diseases (e.g., meningitis, measles) and ear infections (otitis media) among 1-year old Inuit with high PCB exposures than among lesser exposed controls (Dewailly et al. 1993b). The infectious disease data may be confounded by a lower seroconversion rate (successful immunization) among Inuit compared to controls (Dewailly et al. 1993b).

It is not clear whether PCB exposure is the sole factor leading to neurodevelopmental deficits in the Wisconsin or Michigan cohorts, or if other contaminants, such as mercury, or socio-demographic characteristics might also be associated with these results (Ayotte et al. 1996). Caution must be used when examining data from the Lake Michigan cohort in relation to assessing health risks for Inuit newborns in the Arctic. The mixture of contaminants to which Lake Michigan infants were exposed may be very different from that found in Arctic ecosystems (regional industrial sources as opposed to long-range atmospheric transport). In addition, the Lake Michigan population exposure was through fish consumption, while the diet of northern Inuit also includes species at higher trophic levels (e.g., marine mammals). These dietary differences may lead to quite different contaminant exposure profiles. Studies underway in northern Quebec and the Faeroe Islands should help to answer these questions.

3.2.1.8. <u>Hexachlorobenzene</u>

HCB causes a wide range of effects in laboratory animals, including liver pathology, skin lesions (porphyrial cutanea tarda in humans), behavioral changes, reproductive changes in primates, and effects on the immune system. The provisional TDI for HCB is 0.27 µg/kg bw/d (Government of Canada 1993).

HCB is found at higher levels in serum of newborn Inuit from Arctic Canada compared to southern Canadian populations (CACAR 1996).

3.2.1.9. Mercury (Hg)

Methylmercury (MeHg) is readily absorbed through the intestinal wall, and blood concentrations at steady state reflect the daily intake. Sherlock et al. (1984) have suggested the following relationship between blood and exposure:

Hg concentration in blood (μ g/L) = 0.8 daily intake (μ g/adult).

This equation can be used to estimate human exposure levels from data on blood Hg concentration.

The provisional tolerable weekly intake (PTWI) for total mercury has been set by the WHO (1990b) at 5 μ g/kg bw/ week, and for methylmercury at 3.3 μ g/kg bw/week or 231 μ g/kg/week for a 70 kg person. According to the above-mentioned equation, this intake corresponds to a blood Hg concentration of about 26 μ g/L. The lowest

blood Hg concentration at which neurological signs have been observed in exposed adults is often accepted as being 200 μ g/L. For protection of the fetus, maternal blood Hg should not exceed 50 μ g/L.

The biological half-life of methylmercury in humans has been estimated by Åberg et al. (1969). Using a onecompartment model, a half-life of 73 days was determined. This corresponds to the elimination of about 1% of the body burden per day. Recently, Smith et al. (1994) have estimated a half-life of 44 days and an excretion of 1.6% of the body burden per day using a five-compartment model.

Toxic effects: MeHg is neurotoxic and the incidence of signs and symptoms of Hg poisoning are related to its concentrations in the brain. Few data exist on Hg concentrations in human brain; levels between 1 and 2 mg/kg fresh tissue in brain correspond to the lowest blood Hg concentration at which neurological signs have been observed (Berlin 1986).

MeHg readily crosses the placental barrier such that exposure to methylmercury in utero can give rise to severe neurological damage in children (Amin-Zaki et al. 1974, Harada 1977). The fetal central nervous system (CNS) has been found to have higher concentrations than the maternal CNS in both humans and in experimental animals (Marsh et al. 1980, Reynolds and Pitkin 1975). Furthermore, it is likely that the fetal CNS reacts differently and is more sensitive than the maternal CNS. Even if there are similarities in neuropathological findings between adults and infants (Choi et al. 1978), there are specific findings in children, such as ectopic cells and cortical disintegration, which are not seen in adults. This suggests an effect on astroglial cells when the fetal CNS is exposed to methylmercury. Peckham and Choi (1988) have shown in experiments on mice that methylmercury disturbs development of astroglia resulting in abnormal distribution of cortical neurons. This may explain the behavioral abnormalities observed after methyl mercury exposure. In humans, the development of astroglia starts at gestational week seven and continues throughout the fetal life (Reske-Nielsen et al. 1987). Thus, the effects of methyl mercury could be exerted during most of fetal development. Neonatal CNS development can also be affected by exposure to mercury through breast milk.

Methyl mercury may also affect the immune system, however, existing data are not conclusive. Ohi et al. (1976), Koller et al. (1979), and Blakely et al. (1980) did not find significant immunosuppressive effects in mice and rabbits. Petruccioli and Turillazzi (1990) have reported that monkeys (Macca fasciculans), exposed orally to 0.4- $50 \mu g/kg bw/d$, showed a progressive dose-related reduction of IgG and, in the highest exposed groups, reduction in IgM and IgA. The lowest dose used in the monkey study (0.4 $\mu g/kg bw/d$) corresponds to a daily intake by a 70 kg adult human of 28 μg (0.4 70), which is close to the PTWI. Based on these data, immunosuppressive effects of methyl mercury at exposure levels actually reported in several Arctic communities cannot be excluded.

Using the autometallographic technique, it has been demonstrated that methyl mercury exposure leads to heavy accumulation of mercury in the thyroid gland (Hansen et al. 1989a, Hansen and Danscher 1995). As this technique only reveals inorganic mercury, this observation is consistent with a high myeloperoxidase-iodine activity with a supposed high demethylation rate. At present, no studies have been carried out to investigate possible adverse effects.

3.2.1.10. Lead (Pb)

In humans, Pb is initially distributed to various organs and tissues and is gradually redistributed into two compartments: an exchangeable compartment, comprising blood and soft tissues, and a storage compartment, essentially bone. Lead levels in bone continue to increase throughout life while stabilizing, or in some cases decreasing, in soft tissues.

Blood samples are most commonly used for estimating lead exposure. Mean blood Pb concentrations for nonoccupationally exposed persons in industrialized areas in Europe and North America are often reported to be between 100 and 200 μ g/L blood. Studies in remote societies have shown lower concentrations. Poole et al. (1980) found a mean blood concentration of 50 μ g/L in 100 children from an unpolluted area in Papua New Guinea, and Piomelli et al. (1981) found 30 μ g/L in 103 Nepalese children and adults. Concentrations below 10 μ g/L have been reported among Venezuela Indians (Hecker et al. 1974).

Existing data indicate that the northern hemisphere is more polluted with lead than the southern hemisphere, which is in accordance with the fact that most lead-emitting industries and historically the highest concentrations of automobiles burning leaded gasoline are found in the north.

Some studies have indicated a decrease in environmental exposure. Rabinowitz and Needleman (1982) reported a mean annual decline in lead levels of 11% in umbilical cord blood samples taken in Boston between 1979 and

1981. Among children in Chicago, aged 6 months to 5 years, Hayes et al. (1994) found a decline from a median blood Pb concentration of 300 μ g/L in 1968 to 120 μ g/L in 1988. In maternal blood samples collected in Greenland between 1984 and 1989, Hansen et al. (1990b) found a mean annual decrease of 7%.

Toxic effects: Toxic effects from lead constitute a continuum from clinically overt effects to subtle biochemical effects involving the hematologic, neurologic and renal systems. In general, children are more sensitive to lead exposure than adults, due to a higher intestinal absorption and high sensitivity of immature tissues. Prenatal life may be a period of particular vulnerability. The earliest and most sensitive effect is inhibition of δ -aminolevulinic dehydratase (ALA-D), which can be observed at blood concentrations < 100 µg/L. Lead also inhibits ferro chelatase, resulting in anemia.

In 1979, Needleman et al. reported that children exposed to environmental lead who did not exhibit clinical symptoms of lead toxicity, had deficits in psychometric intelligence, speech and language-processing, attention, and classroom performance. In an eleven-year follow-up, the neurobehavioral deficits were found to persist (Needlemann et al. 1990).

Numerous other studies of the effects of lead on children have been published, and the results have been combined by means of quantitative meta analysis to provide a more valid estimate of the true effect level (Needleman and Gatsonis 1990). A WHO/CEC study on 1800 children in eight European countries confirmed that there are small but detectable exposure-related neurobehavioral effects in school-age children. The study also found that it was not possible to identify an effect threshold (Winneke et al. 1990). Epidemiological studies have reported that lowlevel exposure to lead ($\geq 100 \ \mu g/L$ in blood) during early childhood is inversely associated with neuropsychological development at schoolage (Baghurst et al. 1992). Today there is an increasing concern about childhood exposure to lead at levels as low as 100 $\mu g/L$ (Davis et al. 1993).

The risk to the fetus from exposure in utero is uncertain (Rice 1990, Bellinger et al. 1992). The general sensitivity of the prenatal nervous system to chemical insult, the presence of lead in the fetus after maternal exposure, and subtle effects seen in rodents exposed during gestation, all raise concerns.

Recently, Newland et al. (1994) have demonstrated prolonged behavioral effects and learning deficits in squirrel monkeys born to mothers exposed to lead during pregnancy and with blood levels equivalent to those tolerated in humans in occupational settings. These data raise the possibility of human fetal hazards at exposure levels actually present in certain occupational environments and in heavily polluted areas. Studies by Blanzka et al. (1994) suggest that low concentrations of lead are capable of inhibiting nitrite produced by the calcium-dependent constitutive form of nitric oxide synthase (cNOS), while the calcium-independent inducible form of nitric oxide synthase (iNOS) is not affected. These data provide a new hypothesis for the mechanism of lead neurotoxicity, as nitric oxide acts as a neurotransmitter in the brain (Snyder 1992).

3.2.1.11. <u>Tin (Sn)</u>

Although animal experiments indicate that tin is an essential nutrient, naturally occurring tin deficiency is unknown in both animals and humans.

Ingested inorganic tin is poorly absorbed and is mainly excreted in the feces. Apart from rare reports of gastrointestinal symptoms, there is little evidence of human toxicity from inorganic tin in foods. However, the organo-tin compound tributyltin (TBT) may be of importance to the Arctic marine environment. Low concentrations of TBT originating from antifouling paint used on ships have been shown to have reproductive effects in mollusks (Svavarsson and Skarphedinsdottir 1995). TBT has also been shown to be highly toxic in experimental animals, acting through the generation of reactive oxygen species (Clerici 1996). The documented effects of TBT on human health are inflammation of the airway, and eye and skin irritation (WHO 1990a, Snoeij et al. 1987). The effect of TBT on skin is suggested to involve intercellular modulation of interleukin-1 α IL-1 α (Corsini et al. 1996). An evaluation of the human toxicity of TBT, as a marine contaminant, is warranted if it is shown to be transported through the food chain.

3.2.2. National Reports: Levels and Trends of PTSs in Humans

No national report is available from the United States of America.

3.2.2.1. <u>Canada</u>

3.2.2.1.1. DDT and metabolites

For the population in southern Canada, the dietary intake of total DDT (including metabolites) has been declining since the 1960s. This has been reflected by a concomitant ten-fold decline in total DDT in human tissues over the last 20 years (Conacher and Mes 1993, Mes 1994).

Levels of total DDT and its metabolites in human tissue in the Arctic are considerably higher than those in southern Canadians, reflecting the greater consumption of high trophic level species for food. Concentrations of DDE are four- to five-fold higher in human breast milk from Inuit in northern Quebec than populations from southern Canada. People from the north shore of the St. Lawrence River who consume large amounts of fish and gull eggs have intermediate levels of DDE in breast milk. Even higher concentrations of DDE are seen in abdominal fat tissue from Greenland Inuit. The observed differences may be due to increased consumption of contaminated country foods, older age groups in the Greenland Inuit study versus the Quebec Inuit study, or tissue-specific concentration differences (abdominal fat vs. breast milk fat). There is no indication that DDT levels are declining in Arctic populations, however, tissue sampling in the north has only occurred relatively recently and, thus, there are insufficient data for temporal trend analyses.

3.2.2.1.2. Toxaphene

Toxaphene intakes by Arctic populations are entirely dependent on the type and amount of country food consumed (of all tissues tested, toxaphene levels are highest in ring seal blubber and beluga blubber in the eastern Arctic) (Kuhnlein et al. 1995a). There are very few data available on toxaphene in the tissues of Arctic populations. A study of three Inuit women from northern Quebec reported levels of toxaphene in breast milk of 221 ng/g lipid (Stern et al. 1992). This is a very small sample size for any evaluation and is unlikely to be representative of the larger population. A recent analysis of a 1987-88 dietary survey indicates that for one group of Canadian Inuit women the daily intake of toxaphene frequently exceeds the TDI.

3.2.2.1.3. PCBs

In southern Canada, the estimated adult daily intake for total PCB from diet is $0.008 \ \mu g/kg \ bw/day$ (Conacher et al. 1994). Intakes by Arctic residents consuming large amounts of traditional foods from the aquatic and marine environment are considerably higher. In results from two surveys of Canadian Inuit women, 16% and 4% of the women's daily intakes exceeded the TDI.

Several studies suggest that PCB levels in breast milk fat are significantly elevated in some Inuit mothers. In breast milk samples collected from Inuit women residing in the Nunavik region of Arctic Quebec (Hudson Bay, Hudson Strait and Ungava Bay), total PCB levels were elevated 4.7-fold in 1988 and 5.6-fold in 1989/90 when compared to a southern Quebec non-indigenous population in the same sampling years. Women from the north shore of the St. Lawrence River who consume more fish and country food than women from southern Quebec but less than Inuit women had intermediate levels of PCBs in their breast milk.

While levels of total PCBs have declined since the 1980s in breast milk of southern Canadian women (Newsome et al. 1995), there is no similar information yet available for Arctic residents. Historical values are also difficult to compare due to changing analytical techniques and quantitation methods (reported PCB concentrations based on an Aroclor 1260 standard are approximately double those based on a sum of specific PCB congeners). This may partly explain the difference observed between the 1992 southern Canada PCB level in breast milk of 238 ng/g lipid (sum of congeners) and the 1989-90 southern Quebec PCB level of 520 ng/g lipid (Aroclor 1260).

3.2.2.1.4. Dioxins (PCDDs) and furans (PCDFs)

In 40 breast milk samples collected between July 1989 and July 1990 from Inuit women residing in the Nunavik region of Arctic Quebec (Hudson Bay, Hudson Strait and Ungava Bay), 2,3,7,8-TCDD TEQs for PCDDs and PCDFs were moderately elevated when compared to the southern Quebec non-indigenous population (19.0 pg/g lipid vs. 9.6 pg/g lipid) (Dewailly et al. 1992). However, total PCBs were more than five-fold higher in the Quebec Inuit mothers' breast milk than the same control population. When TEQs for non-ortho- substituted PCBs (CBs 77, 126 and 169) were factored in, the 2,3,7,8-TCDD TEQ value increased to 42.3 pg/g for the Inuit milk samples compared to 17.9 pg/g for the southern Quebec samples. For this Inuit population, but not the southern Quebec population, the majority of the 2,3,7,8-TCDD TEQs are due to dioxin-like PCBs.

A large Netherlands study of breast milk and child development found levels of 2,3,7,8-TCDD TEQ (PCDD/PCDF/ coplanar PCBs) in breast milk of 46.2 pg/g lipid. These levels are very similar to the levels in Inuit breast milk. In contrast to the Inuit results, the majority of the 2,3,7,8-TCDD TEQ in this Netherlands study are from the dioxin and furan component. Subtle clinical, immunologic and neurodevelopmental alterations associated with breast-feeding have been reported in the infants/children of the Netherlands cohort (Koopman-Esseboom et al. 1994a, Huisman et al. 1995, Weisglas-Kuperus et al. 1995). A preliminary report on a study of Canadian Inuit also shows an association between increasing PCB/dioxin/furan levels and immune system deficits (Dewailly et al. 1993b).

3.2.2.1.5. Mirex

While mirex has never been registered for use in Canada as a pesticide, it did enter Canada as a fire retardant (dechlorane) from the mid-1960s to mid-1970s, and via water currents from the US side of the Great Lakes basin. Mirex levels in breast milk are above average for communities consuming high amounts of fish and marine bird eggs (Dewailly et al. 1991). Levels in the breast milk of Inuit from Nunavik, northern Quebec, are ten times higher than those in southern Quebec residents. Intermediate levels of mirex can be seen in the lower North Shore St. Lawrence population, which consumes more fish and seabird eggs than the general population (Dewailly et al. 1991). Even higher concentrations of mirex are seen in abdominal fat tissue from Greenland Inuit.

3.2.2.1.6. Chlordane

Chlordane is not currently registered for use in Canada and enters the Arctic ecosystem primarily via long-range atmospheric transport. Chlordane and the compounds related to it accumulate in fat and are found in human tissues although generally only in small amounts. Tissues often contain relatively larger amounts of transnonachlor and the metabolite oxychlordane. The breast milk of Inuit mothers from Nunavik (northern Quebec) had chlordane levels ten times higher than levels seen in southern Canadian residents. Even higher relative levels of transnonachlor (80-fold) and oxychlordane (64-fold) are seen in abdominal fat from Greenland Inuit, but this may be due to several factors as indicated previously. In results from two surveys of Canadian Inuit women, 48% and 75% of the women's daily intakes exceeded the TDI. Only 6% of the Dene women's intakes exceeded the TDI.

3.2.2.1.7. Hexachlorocyclohexanes (HCHs)

Canada still has some registered uses for lindane (α -HCH). Like other POPs, most human exposure to HCHs comes from food consumption. Because of its persistence, β -HCH is found at the highest level of the three isomers reported in the southern Canadian population. There are no data for levels in Arctic populations except for one report from Greenland where β -HCH levels in abdominal fat samples were five times higher than southern Canadian levels in breast milk fat. As previously indicated for DDT, differences in contaminant levels in the samples from Greenland and Canada may be explained by a number of factors.

3.2.2.1.8. POPs in newborn cord blood

The exposure of the developing fetus to maternal contaminants through the placenta is an important route of exposure during many critical stages of development. Data has recently become available on POPs in newborn cord blood from several Canadian Arctic and southern populations (Dewailly et al. 1996b, Walker et al. in prep.). Inuit populations have a greater range of POPs at detectable levels and higher geometric mean concentrations of POPs. This is not unexpected as Inuit consume more marine fats, which have elevated POP residues, than Dene/Métis peoples in the central Arctic (Kuhnlein et al. 1995b). Any comparison of maternal and newborn cord blood concentrations of contaminants needs to consider that the important determinants of contaminants level are age of the mother, number of previous children, and number of children breast fed, as well as the amount and type of country food consumption. While these comparisons are as yet incomplete, it is possible to compare some of the contaminants showing the highest levels and greatest differences between the groups to verify whether previous information on country food consumption supports the contaminant patterns seen.

Based on geometric means, the contaminants present in the highest concentrations in newborn cord blood are hexachlorobenzene (HCB), DDE and PCBs. Levels of PCBs and DDE are twice as high in Nunavik (northern Quebec) Inuit than in Inuit living in the NWT. HCB levels are similar in Inuit in both areas. The difference between Inuit from Nunavik and Inuit from the NWT warrants further study and analysis. Comparing Inuit with other population groups, the concentrations of PCBs and DDE are considerably higher in the Inuit (Nunavik and NWT) than in the other population groups (Dene/ Métis and non-indigenous in the NWT; and residents of

southern Quebec). Levels of HCB in Inuit (both NWT and Nunavik) are two to three times the levels seen in the other population groups.

Analyses were completed for 14 PCB congeners in newborn cord blood from Nunavik and southern Quebec populations. Similar PCB congener patterns can be seen in these two populations, although the levels for each congener in the Nunavik population survey are consistently higher.

3.2.2.1.9. Mercury

From 1970 to 1995, the Medical Services Branch (MSB) of Health Canada, within its Environmental Contaminants Program, has carried out mercury analyses of hair and blood samples obtained from 38 571 residents of 514 native communities across Canada as part of clinical risk assessment (Wheatley 1994). Of 711 Dene and 1511 Inuit from the NWT who were tested, nearly three times as many Inuit (57.2%) exceeded 20 μ g/L as Dene (19.6%) (Wheatley 1995, Wheatley and Paradis 1996a).

The Canadian findings have been assessed in the context of findings in other circumpolar countries and over time. In general, most the mercury data reported by Wheatley and Paradis (1995, 1996a, 1996b) indicate that Inuit from the eastern NWT and Nunavik, and northern Quebec Cree, have higher levels of mercury than other circumpolar peoples. One recent data set from Greenland indicates that high cord and maternal blood mercury levels are also found in this population (Hansen et al. 1990a, 1990b). Some archeological hair sample results from northern Baffin Island (dated to originate from the years 400 and 1150) (Wheatley and Wheatley 1988) and from Greenland (1485) (Hart-Hansen et al. 1991) are also included (converted to blood level equivalents to facilitate comparison). All are low compared with current human methylmercury levels in the same geographic areas. Trend data over 15-20 years for individuals and communities south of 60° N (First Nations) and in the NWT (Inuit) (Wheatley and Paradis 1996a) have been examined. The trend in First Nations communities south of 60° N appears to be downward. Whether this is because of falling mercury levels in fish or because less fish are being eaten by the people has yet to be determined. With the NWT data, there is no obvious trend. There is more recent data from the NWT and Nunavik that does suggest that some Dene and Inuit groups may have decreased blood mercury levels (means 1.74 to 15.97 μ g/L from 1992-95 vs. 10.66 to 48.21 μ g/L before 1992). Dietary, geographic, regional, or seasonal differences may explain the apparent decrease.

In a limited number (n =36) of cord blood samples collected from Inuit and Dene from 1978 to 1986 from various regions of the NWT, 69% had methylmercury levels equal to or greater than 20 μ g/L (mean 36.9 μ g/L, range 4.3-130.4) (Wheatley and Paradis 1996a). This is in contrast to more recent data (1993-95) from the NWT and Nunavik, northern Quebec, which found lower levels in cord blood of Inuit newborns (means 5.72 and 12.09 μ g/L, respectively). The concentration of mercury in maternal blood was also markedly lower in the 1994-95 Walker et al. study (in prep.) than was seen in the 1978-86 study. The lower concentrations seen in 1994-95 cord and maternal blood samples may be due to dietary differences over time (1980 vs. 1994), geographic differences (eastern vs. western NWT) or seasonal differences. In general, levels of methylmercury were higher in the northern and eastern Inuit communities where the consumption of fish and marine mammals is extensive (Wheatley and Paradis 1995). Two recent studies (1987, 1992) indicate that 29% and 37% of Inuit women in these communities had daily dietary intakes of mercury that exceeded the TDI. The source of methylmercury (geological or anthropogenic) in the Canadian Arctic food chain and in traditional foods, such as marine mammals consumed by Inuit, continues to be a subject of debate (Wheatley and Wheatley 1988, Muir et al. 1992). There is also ongoing controversy about possible protective mechanisms involving selenium.

3.2.2.1.10. Lead

In the most recent Canadian market basket survey, the estimated mean dietary intake of lead for a sixty-kilogram adult in 1986-1988 was 24 μ g per day (Dabeka and McKenzie 1995). The estimated daily intakes for adults can range worldwide from 15-316 μ g/d (WHO 1995). In the absence of occupational exposure, the average blood lead concentration in adults is 100-200 μ g/L, depending on the degree of industrialization (air pollution, automobile exhaust, etc.). This value is far in excess of the estimated blood lead level in pre-industrialized humans of 0.16 μ g/L (Flegal and Smith 1992).

Results of the recent Santé Quebec Health Survey of Inuit from Nunavik, showed that the mean blood lead concentrations were approximately 86 μ g/L. Blood lead levels could be positively correlated to smoking and consumption of marine mammals (Dewailly et al. 1994b). In a 1987 survey of northern Greenlandic Inuit hunters and their families, the median blood lead value for males was 96 μ g/L (median age 40 years) and for females, 56 μ g/L (median age 36 years) (Milman et al. 1994). The major source of lead exposure was estimated to be food

items of marine origin (ringed seal, narwhal, walrus and beluga) and there was a positive correlation between increasing blood lead levels and age. In the Faeroe Islands, 52 women (20-50 years old) who consumed fish and pilot whale meat had a median blood lead level of 20 μ g/L (Grandjean et al. 1992). In a 1989 sample of Michigan fishermen (n = 115) who consumed on average 38 fish meals per year, blood lead levels were 55 μ g/L compared to 38 μ g/L in the controls (n = 95, 4.1 average fish meals per year) (Hovinga et al. 1993). Blood lead values for both sample sets were directly influenced by amount of smoking. A study of fisheaters from the Canadian side of the Great Lakes also indicated that women who consumed Great Lakes fish had slightly higher blood lead compared to non-fisheaters (24 vs. 19 μ g/L, Kearney et al. 1995.

Initial results of cord blood screening for lead from Nunavik have found levels approximately threefold higher than comparison samples from Toronto and Quebec City (52 μ g/L vs. 17 and 18 μ g/L, respectively) (Dewailly 1994a).

A similar cord blood study from the NWT found slightly lower lead levels in Inuit cord blood (29 μ g/L), but even lower levels were seen in Dene/Métis and non-indigenous groups (20 and 16 μ g/L, respectively) (Walker et al. in prep).

3.2.2.1.11. Conclusions

Elevated levels of toxaphene, chlordane and mercury, coupled with current intake scenarios, suggest some Inuit groups are exposed to levels of these three contaminants significantly above the Tolerable Daily Intake (TDI). The Dene of the western NWT have smaller exceedances of the TDI for toxaphene and chlordane (CACAR 1997). There is insufficient information to conclude whether the TDI for dioxins and furans and dioxin-like PCBs is being exceeded in Canadian Arctic populations. Also, there is as yet little conclusive scientific information directly linking harmful human effects to these low levels of exposure. The risks associated with a shift in dietary preference need to be considered along with the risks associated with the presence of contaminants in Arctic wildlife consumed as traditional food. Weighing the uncertainty in some of the TDI values (e.g., toxaphene) against the benefits of traditional food gathering and consumption, it is generally recommended that consumption continue. However, consideration needs to be given to reducing intakes of some highly contaminated foods and increasing consumption of other equally nutritious traditional foods that have lower contaminant levels (Dewailly et al. 1996e).

3.2.2.2. Denmark/Greenland and The Faeroe Islands

3.2.2.2.1. Persistent organic pollutants

While there are few studies of the levels of persistent organic pollutants in Greenlanders, results from human studies in the eastern Canadian Arctic and studies on the Greenlandic fauna indicate that concentrations are likely high. A few studies from the late 1970s on fat taken from biopsies from Greenlanders showed higher DDT and DDE levels in Greenlanders than in the population of Denmark, but lower DDE levels than in the United States, eastern Europe and India (Clausen and Berg 1975, Jensen and Clausen 1979). Concentrations of lindane, aldrin-like residue, dieldrin, heptachlor-like residue, heptachlor epoxide, and PCBs were similar in Greenlanders and Danes. Unfortunately, it was not specified where the specimens were taken from in Greenland.

A recent study, in Nuuk and Ilulissat, of POPs in fat taken at autopsy, showed very high levels in Greenland compared with Canada, Finland and the United States.

Under the AMAP human health-monitoring program in Greenland, at the time of writing, 40 maternal blood samples and 29 cord blood samples had been analyzed for POPs, including 24 sample pairs. It should be noted that concentrations on a lipid basis are similar in mothers and babies. Concentrations for Greenland samples are very high compared with samples from southern Canada, but are low compared with another population survey from Greenland (Dewailly et al. 1996d).

3.2.2.2.2. Mercury

The concentration and distribution of mercury in humans in Greenland has been thoroughly studied over the last 15 years. Surveys have been performed in adults, pregnant women and newborn babies in most parts of Greenland, including the central west coast, with the highest population concentration, and the hunting districts in northwestern, northern and eastern Greenland. In all four regions studied, the determining factor for mercury exposure was the daily intake of meat from marine mammals. At a regional level, the median blood mercury concentrations were directly proportional to the registered mean number of seals caught (and consumed) indicating that mercury concentration in meat is probably similar in all regions of Greenland (Hansen 1990).

In adults, median as well as maximum whole blood concentrations of mercury are lowest in the southwest, higher in the northwest and on the east coast, and very high in North Greenland. In North Greenland, 16% of the adult population studied had blood mercury concentrations exceeding 200 μ g/L. WHO regards this level as the minimum toxic blood concentration in adults. More than 80% of the population in North Greenland exceeded 50 μ g/L (Hansen and Pedersen 1986). This concentration is the maximal acceptable blood mercury concentration for pregnant women (WHO 1990b).

In studies of fetal exposure, blood samples were collected from pregnant women prior to delivery and from the umbilical cord at birth. Mercury concentrations in maternal and cord blood were linearly correlated, but concentrations were somewhat higher in cord blood. Samples were collected from 1982 to 1990. No temporal trend was observed. The regional variation was similar to that of the adults. A preliminary analysis of the data showed a negative correlation between blood mercury concentration and birth weight (Foldspang and Hansen 1990), but a reanalysis of data from the whole study showed only a minor and non-significant negative association. Smoking habits and population group were major determinants of birth weight and gestational length.

Under the AMAP human health-monitoring program in Greenland, 20 paired maternal and cord blood samples were analyzed for mercury. For the paired samples, the mean concentrations (arithmetic mean) in mothers and newborns were 24.2 μ g/L and 53.8 μ g/L, respectively, with medians of 16.9 and 56.7 μ g/L. These concentrations were very high compared with samples from Denmark. Compared with earlier results from Greenland, the mercury concentrations in maternal blood were lower than previously found in southwest Greenland, probably a result of decreased consumption of marine food by pregnant women.

3.2.2.2.3. Lead

Lead levels were determined in individuals living in four representative areas of Greenland and were compared with levels in Greenlanders living in Denmark. The differences among the four areas were relatively small and no difference was found between levels in Greenlanders and Danes living in Greenland. The blood lead levels in Greenlanders were also comparable to those of inhabitants of industrialized areas in Western Europe. Umbilical cord blood lead levels were slightly lower than those of the mothers (Hansen and Pedersen 1986, Hansen 1988).

Lead concentration in blood was not related to dietary habits, i.e., consumption of marine mammals, or to smoking habits, except in pregnant women for whom a moderate relationship to smoking was demonstrated. In East and North Greenland, blood lead levels increased with age. This effect was most pronounced in women.

Blood lead concentrations in Greenlanders are decreasing with time, following the same downward trends noted in Europe and North America where declines are directly related to the introduction of lead-free gasoline. Furthermore, it has been demonstrated by analyses of lead in samples of inland ice that concentrations in Greenland reflect the consumption of leaded gasoline in both Europe and North America. Therefore, it seems well substantiated that the surprisingly high lead exposure in Greenland is caused by long-range atmospheric transport of lead from urbanized centers at lower latitudes.

Under the AMAP human health-monitoring program in Greenland, 20 paired maternal and cord blood samples have been analyzed for lead. For the paired samples, the mean concentrations (arithmetic mean) were 45.6 μ g/L in mothers and 34.9 μ g/L in the newborn, with medians of 34.2 and 31.9 μ g/L, respectively. These concentrations are lower than those previously found and are in agreement with the decreasing trends observed in other parts of the world.

3.2.2.2.4. Faeroe Islands

In a recent study of organochlorine concentrations in pilot whales, it was found that the overall average PCB concentration in blubber is very high, i.e., about 30 μ g/g lipid, with total DDT being about 20 μ g/g lipid (Borrell and Aguilar 1993). Expressed on a lipid weight basis, the organochlorine concentrations are about the same in muscle tissue. However, the fresh weight concentration of PCB averages only about 0.6 μ g/g and total DDT is about half as much (Borrell and Aguilar 1993). Based on these findings, the average daily intake of PCBs of the Faeroe Islands is estimated at above 200 g, with considerable inter-individual variation. For comparison, the average daily PCBs intake in Scandinavia is about 15-20 g. Thus, the difference in average PCBs exposure between these two population groups corresponds to one order of magnitude.

Samples of human milk were collected on the third and fourth day after delivery from women who had given birth in the hospital in Tórshavn. Samples were collected over a one-month period. Four pooled samples were made from 22 samples such that each mother contributed the same amount of fat to the pool. The four groups were separated as follows: Group I: low mercury concentration in the milk (< 1 μ g/L); Group II: high mercury concentration in the milk (> 4 μ g/L); Group III: no more than one fish dinner per week during pregnancy; and Group IV: at least five fish dinners per week during pregnancy. The frequency of pilot whale dinners was very low in Group I, very high in Group II, and at the same intermediate level in Groups III and IV. However, the extent of blubber consumption was not quantified.

Umbilical cord tissue from all children born on the Faeroe Islands in 1986/87 was preserved. Preliminary data on 24 cord samples suggest an overall average PCB concentration of about 1.4 μ g/g lipid. This concentration is based on the sum of the concentrations of PCB congeners 153, 138 and 180, multiplied by 1.7 (Grandjean and Weihe 1992).

Umbilical cord blood samples from 1023 consecutive births in the Faeroe Islands showed a median blood mercury concentration of 24.2 μ g/L; 250 of the samples (25.1%) had blood mercury concentrations that exceeded 40 μ g/L (Grandjean and Weihe 1992). Median mercury concentrations in maternal hair were 4.5 μ g/g; 130 samples (12.7%) contained concentrations that exceeded 10 μ g/g. Frequent consumption of whale meat during pregnancy and, to a much lesser degree, frequent consumption of fish, were associated with high mercury concentrations in cord blood and hair. Increased parity or age was also associated with high mercury concentrations in cord blood and hair. Blood mercury levels were slightly lower if the mother had occasionally consumed alcoholic beverages. Mercury in blood correlated moderately with blood selenium (median 110 μ g/L). Increased selenium concentrations were associated with intake of whale meat, alcohol abstention, delivery after term, and high parity. Lead in cord blood was low (median 17 μ g/L), particularly if the mothers abstained from smoking.

A questionnaire completed by 331 Faeroese adults revealed a daily consumption of 72 g fish, 12 g whale muscle and 7 g of blubber. Fish and pilot whale constituted 44% and 9.5% of Faeroese dinner meals, respectively (Vestergaard and Zachariassen 1987). Most of the fish consumed in the Faeroe Islands is cod with an average mercury concentration of about 0.07 μ g/g (Hygiene Institute, Tórshavn, pers. comm.). Almost all of the mercury in fish is methylated. Muscle tissue of Faeroese pilot whales contained an average mercury concentration of 3.3 μ g/g, about half of which was methylmercury. Higher concentrations occurred in the liver, mainly in the form of inorganic mercury (Juhlshamn et al. 1987).

Based on the data from the questionnaires, and the total mercury concentration in whale and fish, an average daily mercury intake from these foods for individuals over 14 years of age can be calculated as being about 36 g. If the steady-state blood level (in μ g/L) is numerically equal to the average daily intake of a 70 kg person (expressed in μ g methylmercury), the expected blood level would be about 36 μ g/L. However, as only two-thirds of the total mercury in food is methylmercury, the expected average blood levels would be about 25 μ g/L. This level is in agreement with the actual findings in the Faeroe Islands. Iceland

There is very little data on levels of contaminants in people. Blood lead levels were measured in 37 individuals living in Reykjavik in 1992 (Thordardottir and Jóhannesson 1993). The levels were about 50 μ g/L (range 25-88 μ g/L) and were three to five-fold lower than had been found in a similar study in 1978 (Pormar and Jóhannesson 1979). Between 1986 and 1990, lead levels in ambient air in Reykjavik decreased five-fold (Gísladóttir 1992). Cadmium was analyzed in the renal cortex of 30 accident victims and was below 40 μ g/g in all samples (Sólbergsdóttir and Jóhannesson 1992). Mercury, arsenic, cadmium, selenium and zinc were analyzed in human hair from 58 individuals in 1981 (Jóhannesson et al. 1981). The levels were all found to be low or normal. Volcanic and geothermal activity did not appear to increase the body burden of mercury in the Icelandic population.

The levels of PCBs and organochlorine pesticides in 22 samples of breast milk collected in Reykjavik in 1993 were similar or higher than reported elsewhere (Jensen 1990). The mean levels in milk fat were: 47 ng/g HCB, 360 ng/g DDT, 830 ng/g PCB, and 206 ng/g CB 153 (Olafsdottir pers. comm.). The levels of PCBs and organochlorine pesticides in adipose tissue and brain from 15 postmortem samples were determined in 1994. The levels were again found to be similar or higher than reported elsewhere (Luotamo et al. 1991, Willams and LeBel 1990). The mean levels in adipose tissue were: 100 ng/g lipid HCB, 1100 ng/g lipid DDT and 550 ng/g lipid CB 153. A study of metals and organochlorines in maternal blood is ongoing in collaboration with the other AMAP nations.

3.2.2.3. Norway, Russia and Finland

The national reports of Norway and Russia, and to a certain extent Finland, are partly integrated because of the proximity of parts of their Arctic regions where related human health studies have been carried out under AMAP. The most severe ecological problems in the area are related to the industrial pollution on the Kola Peninsula of Russia, close to the Norwegian-Russian-Finnish border, and in the Norilsk region of Siberia.

3.2.2.3.1. Persistent organic pollutants

A Norwegian survey of PCBs, PCDDs and PCDFs in human milk during 1985/86 included the town of Tromsø in the Norwegian AMAP area (Clench-Aas et al. 1988). Elverum, an inland community in the south, and the southern industrialized town of Porsgrunn, were also surveyed. No significant differences in the concentrations of PCBs were found between the three locations, or in the levels of PCDDs and PCDFs expressed as TCDD equivalents (TEQs). However, an indication of regional differences was found for some PCDD and PCDF congeners, consistent with suspected sources of these compounds. All the values were close in magnitude to data reported for other western countries.

A more comprehensive study was carried out in the same areas in 1992/93 (Becher et al. 1995), also including locations in Lithuania. There was no significant change in the concentrations of PCBs between the earlier (1985/86) and more recent (1992/93) studies, with means for total PCBs in human milk of 534 μ g/kg lipid and 496 μ g/kg lipid, respectively.

In an investigation of human milk from mothers living in Oslo (n = 28), Skaare et al. (1990) found a mean concentration of total PCB of 488 μ g/kg lipid. Becher et al. (1995) reported total TEQs, including dioxin-like PCBs, between 31 and 42 pg TEQ/g lipid in Norway, compared to 45-49 pg TEQ/g lipid in Lithuania. Dioxin-like PCBs were found to contribute two to three times more to the total TEQs than the PCDDs and PCDFs. Major congeners among the dioxin-like PCBs were CBs 126, 156, 114, 118, and 170. Comparison with the 1985/86 study, indicates a decrease in the mean TEQ levels of about 37% over a 7-year time span for PCDDs/PCDFs, while the levels of total PCBs have remained unchanged or have only slightly decreased. This interesting temporal trend needs to be documented further in future studies.

In 1993, Polder et al. (1996) carried out a study on mothers' milk in Murmansk and Monchegorsk on the Kola Peninsula. Total PCB in mothers' milk from Murmansk was in the range of 250-635 μ g/kg lipid, with a mean value of 484 μ g/ kg lipid (range 7.5-19 μ g/L and mean 12.6 μ g/L). The results from Monchegorsk were in the range of 262-854 μ g/kg lipid, with a mean value of 535 μ g/kg lipid (range 7.8-25.6 μ g/L and mean 16.1 μ g/L). Specific congener concentrations were marginally higher for CBs 74, 99, 118, 138 and 153, while other congeners, such as CB 180 and 187, were lowest. This pattern corresponds well to the known dominance of dietary intake as the major exposure pathway for the general population and the similarity of dietary habits within a country. The conclusion was that the exposures in Monchegorsk and Murmansk are relatively low. No significant geographical differences in levels of PCBs and PCDDs/PCDFs between Murmansk and Monchegorsk were found.

In central Russia, there has been an investigation of breast milk and placentas from mothers employed in a transformer plant in Serpuhov (Moscow County) (Pleskatchevskaya and Bovonikova 1992). The range of the total PCB levels found in milk was 14-105 µg/L (467-3500 µg/kg lipid).

In the Russian AMAP human health monitoring study, PCB levels were analyzed in plasma from women in Salekhard and Norilsk (Klopov pers. comm.). The arithmetic mean concentration of PCBs in plasma of women 18-24 years old was 7.5 μ g/L in Norilsk and 6.8 μ g/L in Salekhard. A mean concentration of 9.9 μ g/L has been reported in a comparable age group of an Inuit population in Canada (Dewailly et al. 1994a). By comparison, the mean plasma PCB concentration in a group of women 25-44 years old was 13.8 μ g/L in Norilsk and 16.1 μ g/L in Salekhard (19.5 μ g/L in the Inuit population in Canada). The mean concentration of PCBs in cord blood samples was 2.1 μ g/L in Norilsk and 1.6 μ g/L in Salekhard (2.8 μ g/L in the Inuit population in Canada). The mean concentration of PCBs in breast milk of the Norilsk women was 799 μ g/kg lipid, while in Salekhard it was 847 μ g/kg lipid. The major food source of PCBs is freshwater fish, which constitutes a significant portion of the diet.

Very high levels of DDT have been reported during 1970-1980 in human blood samples from the southern regions of the former Soviet Union (Klopov pers. comm.). The reason for this might be the huge quantities of pesticides used in agriculture practices. In certain regions of Moldavia, this has been suggested to be a reason for a high incidence of miscarriages and congenital malformations (Klopov pers. comm.). The mean concentrations of p, p'-

DDE in maternal plasma of delivering women in Norilsk and Salekhard were found to be rather low: 0.67 and 0.38 μ g/L, respectively, compared to 11.3 μ g/L in the Inuit population of Quebec, Canada (Dewailly et al. 1994a). In cord blood, the concentrations showed the same pattern: Norilsk 0.23 μ g/L and Salekhard 0.28 μ g/L, compared to 1.63 μ g/L in northern Quebec. The mean p, p'-DDE in breast milk showed levels of 1291 μ g/kg lipid (Norilsk) and 978.3 μ g/ kg lipid (Salekhard), compared to 1212 μ g/kg lipid (Nunavik in Canada).

Under the Norwegian-Russian AMAP human health monitoring study, ten samples of breast milk were collected from first-delivery mothers in Arkhangelsk and Severodvinsk, both located in Arkhangelsk County. These samples were analyzed for chlorinated organic compounds. The observed concentrations match those found in Murmansk, Monchegorsk and Lithuania. Other than for total chlordanes, and possibly also total PCBs and total diortho PCBs, concentrations of chlorinated compounds in breast milk are considerably higher in samples from Russia and Lithuania than in samples from Norway. The relative enhancement of total mono-ortho-PCBs in the Russian samples is especially interesting, since these have significant associated TEQ values because of their resemblance to dioxins.

A final observation concerning the DDE to DDT ratio values is appropriate. This ratio provides information about current use of p, p'-DDT, or other possible current exposure, such as through imported foods. The relatively low DDE/DDT values found for the Russian breast milk samples suggest a higher current exposure to DDT here than in Norway or Lithuania.

3.2.2.3.2. Mercury

Under the joint Norwegian-Russian AMAP human health monitoring study of Norway, Kola and Arkhangelsk, the mean maternal blood mercury levels were marginally higher in the Norwegian population (p < 0.01); and for both the Russian and Norwegian groups as a whole, they corresponded to reference values for moderate to no fish consumption (Odland et al. 1996, 1997). When broken down by community, the following mean values (in $\mu g/L$) are found: 1.6 (Arkhangelsk, n = 50); 2.3 (Nikel, n = 50); 2.5 (Hammerfest, n = 57); and 3.4 (Kirkenes, n = 40), with the difference between Hammerfest and Arkhangelsk being statistically significant (p < 0.0001).

Consistent with the previously observed correlation between fish intake and blood mercury levels (Brune et al. 1991), the mean for the fishing town of Kirkenes was the highest. The very low levels for Arkhangelsk suggest that methylmercury does not enter the food chain. It is clear that the levels observed in Kirkenes, Hammerfest, Nikel and Arkhangelsk do not pose any health risk.

3.2.2.3.3. Lead

Under the joint Norwegian-Russian AMAP human health monitoring study in Norway, Kola Peninsula and Arkhangelsk, the mean lead levels in mothers and neonates in Russia as a whole are significantly higher than in Norway (p < 0.0001); both sets of data conform to the expected reference values (Odland et al. 1997). The recently obtained results from Siberia and Tromsø concur. A linear relationship between maternal blood and cord blood lead is clearly observed (e.g., for the town of Nikel, r = 0.96, p < 0.02, n = 24).

Even though the Russian values are significantly higher as a whole, both population groups have mean blood lead concentrations within the reference interval of 0-40 μ g/L. Russian cars are still using leaded gasoline, but the traffic density is not as high as in western cities, even in a town like Monchegorsk with a population of about 60 000 inhabitants. If the currently observed blood lead concentrations of the Russian women are compared with western measurements from 20 years ago (Environmental Health Perspectives 1990, 1991), much lower concentrations are found in the Russian women. Overall, the Norwegian blood lead results constitute some of the lowest concentrations reported in the literature. Improvement in preventing contamination during collection, sampling, handling and storage of the specimens might be one reason for this. Another, of course, is that following introduction of lead-free gasoline, the effect of reduced emissions is being observed in humans.

Whole blood lead concentrations were determined in the children aged 2 to 13 from three Russian towns (Lovozero, Krasnochelie and Apatity). Lovozero is the Russian center for the Saami population with approximately 2000 inhabitants. Krasnochelie is a very isolated community on the Ponoi River, far east on the Kola Peninsula, with a mixed population of Saami, Komi, Nenets and Russians, totaling 850 people. Apatity is one of the bigger cities of Murmansk County, with approximately 89 000 inhabitants. The blood lead concentrations of the children in Krasnochelie are significantly higher than the concentrations in both Lovozero and Apatity (p < 0.001). Blood lead concentrations greater than 100 µg/L are of medical concern in children, and these occur mostly in Krasnochelie, but also in Lovozero. Other investigations have also revealed a trend of higher

blood lead levels in small, remote communities than in bigger cities (Nieboer pers. comm.). One possible explanation might be the lead content of wild meats due to the use of leaded ammunition, especially lead shot. A more thorough dietary evaluation is necessary to further elucidate this important finding.

The concentrations reported here for chlorinated organic compounds in breast milk from Finland are intermediate in magnitude to those found in samples from Norway (lower levels) and Russia (higher levels), except for total PCBs and total chlordanes, which appear to exceed the Russian values.

For lead and mercury, the levels found are mostly within baseline reference intervals, in all the populations examined, including groups from the Russian town of Nikel, which is regarded as seriously polluted.

In the available studies reporting levels of toxic metals in biological material from Finland, no concentrations have been found near any 'lowest-observed-adverse-effect- levels'.

3.2.2.4. <u>Finland</u>

3.2.2.4.1. Persistent organic pollutants

Organochlorine pesticide and PCB residues were analyzed in 183 human milk samples obtained in 1984/85 from 165 women living in different parts of Finland (Mussalo-Rauhaama et al. 1988). The p, p'-DDE concentrations were above the detection limit in 99.5% of the samples, p, p'-DDD and p, p'-DDT in 57.9%, isomers of HCH in 30%, cis-chlordane in 4.9%, oxychlordane in 3.3%, trans-nonachlor in 6%, heptachlor in 12%, and heptachlor epoxide in 6.6% of samples. Mirex was not found in any of the milk samples, whereas the signals of toxaphenes were detected, but could not be quantified. The mean fat-adjusted residue levels above the detection limit in Finnish human milk samples of primipara mothers were 660 μ g/kg lipid for total DDT compounds, 80 μ g/kg lipid for HCB, 930 μ g/kg lipid for heptachlor epoxide. The corresponding geometric means were 460, 60, 570, 20, 20, and 10 μ g/kg lipid, respectively. The age of the mothers correlated positively with the DDE concentrations in human milk. The residues of organochlorine compounds in human milk did not differ between mothers living in industrial regions and other mothers. Small, but insignificant differences between regions were found. No relation was found between the organochlorine contents and fish consumption, smoking habits, weight loss or social group of the donors. Additional AMAP data are currently being gathered.

3.2.2.4.2. Mercury

Mussalo-Rauhamaa et al. (1996) have described trends in the concentrations of mercury, copper, zinc and selenium in hair and serum of inhabitants of northeastern Finnish Lapland in 1982-1991. The mean concentration of mercury in the hair of the 19 individuals in the study from northeastern Lapland in 1991 was 1.5 mg/kg (range 0.2-6.2 mg/kg). The men consumed more reindeer meat and fish than the women did, but no significant difference in mercury content between the sexes was observed. The mean mercury concentrations in hair from people living in the Russian-Finnish border region were slightly higher, but not significantly different. The comparison of people in Lapland from 1982 to 1991 showed slightly lower values in 1991, but the differences were not significant.

3.2.2.5. <u>Sweden</u>

3.2.2.5.1. Persistent organic pollutants

It is now clearly established that environmental levels of several persistent organochlorines are declining in Sweden. In the northern areas, close to or above the Arctic Circle, levels in Arctic char, pike, burbot, white-tailed eagle, falcon and osprey are lower compared with southern locations, and are decreasing. Levels in humans are also following the downward trends (Noren 1993). In breast milk sampled in Stockholm between 1967 and 1989, a decrease in the levels of certain pesticides and polychlorinated biphenyls (PCBs) was found. The changes were related to the prohibitions and restrictions applied to the use of these compounds. Downward time-trends were also seen for polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and specific congeners of PCBs, including non-ortho and mono-ortho coplanar PCBs. Between 1972 and 1989, average levels of CB 153 in breast milk decreased from 220 to <150 ng/g lipid, CB 138 decreased from 190 to 120 ng/g lipid, CB 180 decreased from 90 to 70 ng/g lipid, and CB 118 decreased from 60 to <30 ng/g lipid. Levels of oxychlordane and trans-nonachlor also dropped from 0.02 to 0.012 μ g/g lipid. Calculations using toxic equivalent factors relative to 2,3,7,8-TCDD revealed that PCB constituted the major part of the toxic equivalents in human milk. Levels of PCB, total or isomer specific, have also been reported in human milk from Umeå (Lindström 1988). In 1986, an average level of 580 μ g/kg lipid was re-ported (n = 10).

PCDDs were first reported in human tissue in Sweden in the early 1980s. In a 1986-88 study, levels in human milk from Umeå, Sundsvall, Uppsala, Borlänge and Göteborg showed similar mean values, of about 20 pg TEQ/g milk fat (Lindström 1988).

3.2.2.5.2. *Mercury*

Total mercury (Hg) concentration in hair, sampled at delivery, was determined in 122 women living in the smelter area, and in 75 women from the control area (Oskarsson et al. 1994). The average Hg concentration in hair was 0.27 mg/kg (range 0.07-0.96 mg/kg), which is somewhat lower than previously reported for pregnant women in Sweden and very low compared to levels in fish-eating populations in other parts of the world.

3.2.2.5.3. Lead

The blood lead levels were low in both the smelter and the control areas, although women in the smelter area and their newborn children had significantly higher lead levels than the controls. During pregnancy, there was a 20% and 15% increase of blood lead concentrations in the smelter area and reference area, respectively. Umbilical cord blood concentrations were significantly correlated with maternal blood lead levels and were 80-87% of the maternal levels recorded at delivery. Blood lead levels in pregnant women were influenced by place of residence, employment at the smelter, smoking, and wine consumption. Because blood lead concentrations increased during pregnancy, despite increased blood volume and unchanged or decreasing environmental lead levels, mobilization of lead from bone during pregnancy was considered a possibility (Lagerkvist et al. 1996).

In 1977, maternal and cord blood lead levels were measured in Kiruna and Västerbotten (Zetterlund et al. 1977). The mean blood lead concentrations in mother-infant pairs were 61 and 44 μ g/L in mothers and infants, respectively, from Kiruna, and 92 and 80 μ g/L in mothers and infants, respectively, from Västerbotten. In 1991, the mean blood concentrations in the smelter area in Västerbotten were 32 μ g/L in mothers and 28 μ g/L in infants. Current levels in Kiruna are also expected to be lower than 1977 levels.

3.2.3. International Study

The foregoing discussion presented results from individual countries. Comparisons between countries and different studies are difficult. In order to overcome some of these difficulties, under the AMAP Monitoring Program, all circumpolar countries agreed to monitor, in a single study, certain contaminants in specific human tissues. Canada agreed to coordinate and lead a special project for the monitoring of contaminants in maternal blood covering the entire circumpolar region, using a standard sample collection procedure. The objective of this project was to obtain sufficient samples during 1995/96 to assess geographical variations of contaminants (organochlorines and heavy metals) throughout the Arctic. Some results are available.

Persistent organic pollutants: The levels of HCB, mirex and three chlordane metabolites were markedly higher in Greenlandic maternal blood samples than those from the other participating countries. Levels of DDE in Greenlandic and Russian samples were similar and three to five times higher than levels in the other four countries; however, the DDE/DDT ratio for Russian samples was markedly lower than that for samples from all other countries, suggesting current continuing use of DDT.

Interestingly, β -HCH levels in Russian blood were 8-28 times higher than those in the other countries. In addition, the levels of DDT were 3-20 times higher. These findings appear to suggest either that there are significant uses of both HCHs and DDT in the Nikel area, or that there are significant amounts of these pesticides in the food products consumed in this area. These findings are similar to those reported by Polder et al. (1996) in human breast milk from the Kola Peninsula.

Pesticide levels in Canadian samples tended to be lower than those from Greenland, but similar in terms of their relative abundance, however, concentrations of cis- and trans-nonachlor, DDT/DDE, oxychlordane, mirex and hexachlorobenzene were greater than the levels reported in the Swedish, Norwegian and Icelandic samples.

The highest concentrations of PCBs are in the maternal blood samples from Greenland. This is true both for PCBs measured against an Aroclor 1260 standard and based on a sum of 14 congeners. Maternal samples from other countries contain fairly similar levels of PCBs. A review of the relative amounts of the top seven congeners (153 > 138 > 180 > 187 > 170 = 118 > 99) indicates that the congener patterns are similar in all countries except Russia where there are relatively lesser amounts of congeners 153, 170, 180 and 187 and relatively more of congeners 99 and 118 calculated as a percent of the total amount of PCB (sum of 14 congeners).

With the exception of the β -HCH and DDT/DDE levels found in the Russian samples, the patterns of POPs found in these maternal blood samples are consistent with the relative amounts of traditional food consumed, especially where marine mammals make up a larger amount of the diet. The greater reliance of indigenous people on marine species and the highest concentrations of contaminants in the species consumed are found in Greenland, followed by Canada. Mothers sampled in Sweden, Iceland and Norway consumed marine fish species and terrestrial mammals such as reindeer, sheep and cattle, but very few marine mammals. Hence, levels in these countries are very similar and virtually indistinguishable from values found at lower latitudes.

Metals: Mercury levels in maternal blood from the six circumpolar areas sampled are relatively similar, except in Greenland where they are 5-12 times higher than the levels in the other countries. Even though blood mercury levels have declined, there is still a significant percentage of samples with values above the 20-50 μ g/L risk range used by the WHO for adverse effects on fetal development.

Lead levels are again highest in Greenland, with intermediate levels in Canada, but all are well below the 100 μ g/L no-observed-effect-level for protection of the fetus.

3.3. ECOTOXICOLOGY OF PTSs OF REGIONAL CONCERN

3.3.1. Overview of Harmful Effects

3.3.1.1. <u>POPs</u>

The majority of the POPs 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 (MeSO2) group is added during metabolism and a number of MeSO2-DDE and MeSO2-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 characteristic of the Arctic is that most marine mammals have very high fat content in breast milk in order to facilitate fast growth in the young during the short growing season. 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. 1997), 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 HCHs do not. This implies that DDT and HCHs 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 PCBs only biomagnify PCBs 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 before 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.

3.3.1.2. <u>Metals</u>

The ability of organisms to accumulate metals to concentrations of one or more orders of magnitude greater than concentrations in their food usually represents the major pathway leading to chronic toxicity. However, accumulated metal may be present in tissues in a relatively non-toxic or inert form even if it was originally toxic, because the toxicity of the metal can be modified through interactions between metals or through biotransformation by the organism. In contrast to POPs, which are highly lipophilic and therefore accumulate primarily in body lipids, heavy metals are preferentially accumulated in proteinaceous tissues. The degree to which metals are accumulated varies greatly depending both on the metals involved and on the organ or tissue.

Once absorbed, heavy metals are distributed in the body by the circulatory system, irrespective of their chemical form (Foulkes 1995). The fraction of transported metal absorbed by various organs and the fraction subsequently

excreted vary greatly for each metal. The mechanisms involved in selective metal uptake in organs are not well understood (Foulkes 1995). Metals are initially distributed to a variety of organs and tissues, and subsequently redistributed to other tissues for storage and inactivation.

The formation or breakdown of metal-carbon bonds or a change in the oxidation state of a metal within an organism (biotransformation) will affect the chemical activity of heavy metal compounds, and therefore their toxicity. Changes in the oxidation state influence the ability of a metal to interact with various tissue ligands. Hg, for example, exists in three oxidation states: elemental (Hg(0)), the mercurous ion (Hg2(II)), and the mercuric ion (Hg(II)). Hg(0) easily penetrates biological membranes because of its high lipid solubility. The mobility of Hg2(II) and Hg(II) are much more restricted due to their tendency to form salts and their high affinity for sulfhydryl groups on proteins (Clarkson 1986).

Two biotransformation processes are important to the toxicity of metals:

- 1. Methylation/demethylation of certain heavy metals and metalloids (e.g., As, Hg, and Se). In some cases, cleavage may serve as a detoxification pathway, whereas in others the metabolite is the more toxic species. For example, methylation (the formation of metal-carbon bonds) of inorganic As and Se has been seen to lead to reduced toxicity in a number of animals and to form the basis for excretable metabolites (e.g., methylated selenides), while the reverse is true for Hg. In the case of methylmercury exposure, processes of demethylation are important for detoxification. In the case of Se, biotransformation in the liver seems to be the major mechanism by which homeostasis is maintained.
- 2. Formation of inert complexes also plays an important role in heavy metal detoxification. The biochemical relationship between metallothionein and such metals as Cd, Zn, Cu, and Hg, for example, is fundamental to their toxicity. Metals binding with metallothionein form inert complexes, which can be retained in body tissues (Clarkson 1986). Similarly, Se can reduce the toxicity of certain metals such as As, Cd, and Hg by forming inert compounds (Högberg and Alexander 1986), which usually accumulate within organisms.

The fact that many Arctic animals at high trophic levels (e.g., seals, whales, seabirds) are consumed by Arctic people has provided the impetus for analyzing tissue concentrations of various pollutants. Until now, measurements have been primarily carried out on those organs and tissues that are consumed. As a result, the literature on metals and other contaminants in the tissues of Arctic animals may or may not represent targets of toxicological action in the animals themselves. Arctic fish and marine mammals accumulate relatively high levels of methylmercury in kidney, liver, and muscle. Although knowledge of these concentrations is important for assessing the potential exposure of human consumers to methylmercury, in terms of evaluating the toxic effect to mammals the most important concentration is that in the brain, which is generally not measured.

The most important excretory pathways for metal compounds in animals are gastrointestinal and renal. Gastrointestinal excretion includes excretion of metals into bile (and pancreatic fluid) and excretion by the intestinal mucosa. Excretion of most organic metal compounds occurs primarily by the bile, while inorganic compounds are excreted in the gastrointestinal tract. Considerable quantitative differences have been reported for different animal species. Metals excreted in the bile may be reabsorbed farther down the intestinal tract, and subsequently re-excreted into bile. Intestinal re-absorption can be prevented if excreted metals form bonds with non-absorbable compounds. In this way a net gastrointestinal excretion can occur, with the heavy metals being eliminated from the body with feces. The other mechanism of gastrointestinal excretion involves the removal of metal compounds in association with the rapid turnover of cells of the intestinal mucosa. Small quantities of certain heavy metals (e.g., inorganic Cd and Hg) are eliminated from the body when the cells are shed.

Urinary excretion is probably the second most important excretory route for animals. The glomerula membrane acts as a filter, allowing only those molecules with relatively low molecular weights to pass through into the renal tubules. Thus, metals bound to low-molecular weight proteins such as insulin or metallothionein may be cleared from the blood plasma in this way, although a proportion of this is subject to re-absorption. A brief overview of the toxicokinetic characteristics of Pb, and Hg, is followed by details relating to each metal.

In general, invertebrates and fish take up only a small proportion (0.1% for Cd) of the metals in the water through the gills. Intestinal intake by invertebrates and fish is larger; the most efficient intake is for methylmercury (70-80%) and a much smaller uptake for Cd (1%). Excretion by invertebrates and fish compared with higher trophic levels is rather fast, with biological half-times of 3-40, 2-63, and 53-323 days for Pb, Cd, and Hg, respectively. This means that the major route of metals is through food, where the levels also are higher than in the surrounding water.

For higher organisms, metals are efficiently (7-94%) absorbed through the lungs, with the degree of absorption varying by animal species, metal, and chemical form of the metal. Mercury vapor and H₂SeO₄ are taken up most efficiently (80-94%), but Pb and Cd readily cross the lung epithelium (7-50%). Air levels of these metals, however, are rather low in most Arctic areas, so this pathway is of only minor importance. The uptake through the intestine depends on the organism, metal type, and chemical form. Only 1-16% of Pb and Cd are taken up by various species, whereas 57-95% of Hg and Se are absorbed through the intestine. Methylmercury is taken up more than six times as efficiently as inorganic Hg. Lead and Se are readily excreted, which results in moderate levels of these metals in internal organs. Lead, however, is deposited in bone, where its half-time can be up to 20 years. Cadmium is excreted slowly once it is taken up by organisms, with a half-time of 10-50% of the organism's life span (up to 30 years), whereas Hg has intermediate half-time (12-1000 days). Differences in half-time between the different species partly explain the generally higher observed metal levels in longer-lived species. The different half-times also explain differences between the observed levels of the different metals in the various tissue compartments.

3.3.1.2.1. Mercury

Much of the Hg in the environment is unavailable to organisms, as it is strongly bound to sediment or organic material. Inorganic forms can be methylated by microorganisms and transformed to methylmercury, which is much more readily taken up and accumulated in both aquatic and terrestrial organisms.

Uptake in aquatic plants increases with increasing concentrations of Hg salts, with greater levels occurring in roots than in shoots. A similar pattern of absorption and distribution is found in studies with terrestrial plants. Soil type strongly influences the extent of uptake. As with aquatic plants, the highest Hg concentrations occur in the roots, though there is some evidence of translocation to other parts of the plant including the leaves.

Most of the studies of Hg uptake in invertebrates do not differentiate between external absorption and actual uptake. In the case of methylmercury compounds, uptake usually correlates with surface absorption capacity. Assimilation of Hg in arthropods depends on both form (organic or inorganic) and source of exposure, and is highly variable between taxa (Zauke *et al.* 1996).

Environmental variables such as temperature, pH, and redox potential are particularly important for Hg uptake in fish. Data on uptake suggest that absorption increases with higher temperatures and lower pH. Tissue concentrations of Hg increase with age for both marine and freshwater fish. Mercury accumulated in fish is usually in the form of methylmercury, whereas the source is usually inorganic. Mosquito fish (Gambusai affinis) have been shown to absorb metallic Hg five times faster than inorganic Hg. This is presumably related to the high lipid solubility of the metallic form, which allows it to pass through gill membranes while salts become tightly bound to mucoproteins.

As with fish, birds can assimilate organic forms of Hg more readily than inorganic compounds. Methylmercury is distributed evenly in tissues while inorganic compounds accumulated primarily in the liver and kidneys of adult birds. Excretion appeared to be enhanced by egg laying, with concentrations of methylmercury occurring in the egg white and other Hg compounds typically in the yolk. Overall, inorganic forms are more rapidly excreted than methylmercury.

The primary uptake route of Hg in marine and terrestrial mammals is though diet. This is related to the relatively high concentrations of methylmercury in food items (e.g., fish), which is more effectively taken up than inorganic forms.

3.3.1.2.2. Lead

It has been shown that in terrestrial plants, Pb is taken up through the roots and to a lesser extent through the shoots. Some of the Pb entering plant root cells becomes incorporated in the cell wall.

For many animals, it is not known for certain whether Pb is absorbed through the skin or actually taken up via inhalation or contaminated food. Accumulation in mussels (Mytilus edulis) has been demonstrated to occur in all tissues, but highest concentrations are seen in the kidney. In fish, Pb accumulates primarily in the gill, liver, and kidney, though it is not known whether accumulation in the gills represents uptake into the tissue or absorption onto exterior surfaces. Birds dosed with lead shot show signs of tissue accumulation in liver, muscle, and bone, and appear to be influenced by the amount of fiber in their diet. Lead also accumulates in eggs and embryos.

3.3.2. Observed Effects

Biological effects can be measured at different levels of biological organization, from the molecular to the ecosystem level. Biomarkers measurable at a molecular level respond early, but are not readily interpreted ecologically, while measures with established ecological relevance, such as population declines or reduced reproductive rates, respond too late to have diagnostic or preventative value. 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.

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.

3.3.2.1. <u>POPs</u>

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.

3.3.2.1.1. Reproduction

Studies in Canadian, Swedish, and Russian peregrine falcon still indicate that eggshell thinning occurs due to high DDT 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, PCB, and HCB, but the degree of thinning was below that affecting reproduction. No correlations could be found between PCB 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.

3.3.2.1.2. Cytochrome P450 activity

Liver enzyme induction (EROD) seems to be correlated with concentrations of PCBs 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 PCB and dieldrin concentrations in ringed seals from Arviat and between EROD activity and PCB 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.

3.3.2.1.3. Thyroid and retinol effects

A significant negative correlation was found between retinol concentrations and PCB concentrations in polar bear plasma. A similar trend was found for thyroid hormones, but was not statistically significant.

3.3.2.2. <u>Metals</u>

Numerous laboratory experiments have been carried out to assess the toxicological effects of acute and chronic doses of heavy metals. In contrast to this, the data currently available for heavy metal levels in tissues of Arctic organisms are generally obtained from environmental monitoring of natural populations. In this case, the emphasis is on exposure rather than dosage, and the focus is on the circumstances and exposure levels that may elicit effects (Elinder 1984). In the Arctic, metal concentrations, and hence dosages, generally do not change quickly (except in the event of a spill or an accidental release), and therefore information in relation to acute toxicity is less applicable than information relating to chronic toxicity.

A generalized overview of reported effects-threshold levels for Hg, Pb, Cd and Se in tissues of main animal groups has been developed. These thresholds, together with in-formation presented earlier on concentrations of metals observed in Arctic biota, have been compared to produce an assessment of the potential for effects in different Arctic ecosystems. The resulting potential effects constitute a basis for future studies but do not contribute important information to this assessment of actual effects of metals in the Arctic. The overview is not

included here. Instead, the following simple statements constitute our current state of knowledge concerning the impacts of heavy metals on arctic biota.

No biological effects attributable specifically and unambiguously to heavy metal pollution in the Arctic terrestrial ecosystem have been reported.

The only biological effect thought to be attributable specifically and unambiguously to heavy metal pollution in the Arctic freshwater ecosystem is the decline in the ringed seal population of the Lake Saimaa, Finland. There it is thought that Hg contamination (and lack of sufficient Se to detoxify it) has rendered the seals more prone to premature and stillbirths.

3.4. DATA GAPS

Availability of information on contaminant levels in the tissues of northern residents is very recent. It is therefore important to recognize that the paucity of health data or the absence of overt illness or malfunction does not imply that the exposure of peoples in the Arctic to contaminants is without effects.

There is very little information on temporal trends of POPs in Arctic populations.

There is insufficient information to conclude whether the TDI for dioxins and furans and dioxin-like PCBs is being exceeded in Arctic populations. Also, there is as yet little conclusive scientific information directly linking harmful human effects to low levels of exposure to these contaminants.

Existing epidemiological evidence on the adverse effects of POPs in humans is inconclusive and needs to be replicated because of the specific context in the Arctic in which there are differences in genetics, climate, food consumption patterns, and lifestyle among population groups.

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.

Though toxaphene is the major OC contaminant in all freshwater fish and invertebrates, relatively little data on toxaphene levels in freshwater environments are available in some regions and no data are available for surface waters, sediments, or lower food web organisms in the European Arctic. 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.

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 PCB (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).

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 HCHs because most of the OCs were in the dissolved (or dissolved organic carbon associated) phase.

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

Higher Σ PCB 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. In general, documentation of the transfer of OCs via ice is lacking.

The information available on contaminants in Arctic terrestrial mammals is not adequate 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.

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.

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.

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.

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.

On a regional basis, there is scattered coverage of OC levels in waterfowl and game birds, and mink and otter populations, but no single species can be compared on a circumpolar basis.

No information is available on OC levels in polar bear from the Russian Arctic, except for Wrangel Island.

Though PCBs appear to be the most prominent contaminants in the animal species analyzed, the detailed comparison of the results from one region to another of animal or plant levels is problematic because too few samples have been analyzed from each sub-region; there is considerable between-year variation in the results; and percent lipid results are not available.

A large suite of biological effects indicators is 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.

Data on nPCBs and PCDD/Fs levels in fish, marine mammals, and polar bears are very limited. No PCDD/Fs or nPCB data are available for seabirds in the Arctic.

There are no analyses of sediment cores from the Eurasian High Arctic (e.g., Spitsbergen and Bear Island), which could confirm whether observations trends of PCBs in cores from some regions are part of a generalized circumpolar phenomenon.

Health effects associated with metals have so far not been investigated in Arctic biota.

Overall, the information on temporal trends in the Arctic is very limited. The results available reinforce the importance of judicious sampling and archiving programs, which would allow continuous long-term monitoring of key populations and retrospective analysis for new contaminants.

3.5. CONCLUSIONS

3.5.1. POPs

3.5.1.1. Levels

Despite the number of controls on several POPs imposed during the 1970s and 1980s, there is no evidence that levels in Arctic peoples have decreased. The persistence of POPs and the use of some POPs for disease vector control, contribute to the constant influx of POPs into the arctic environment and to sustained levels of human exposure.

Elevated levels of toxaphene and chlordane, coupled with current intake scenarios, suggest some indigenous groups are exposed to levels of these contaminants significantly above the Tolerable Daily Intake (TDI).

Existing data from the literature do not allow a valid estimate of spatial and temporal trends of exposure of Arctic peoples to mercury while for lead, a declining trend is observed. There is some evidence that the general decline in lead exposure parallels the decline in lead levels in industrialized areas.

Lead levels in Arctic indigenous peoples have declined since the implementation of controls on lead emissions. Concentrations of lead in blood currently reported are below a level of concern, however, continued monitoring is warranted because of the potent effects of lead on neurological development in the fetus and children.

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 PCBs and DDT were up to ten times higher in snow from the Taimyr Peninsula and Laptev Sea than in the Canadian Arctic.

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 ice cover 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.

Concentrations of PCBs in lake waters in Canada and Russia exceed levels associated with negative biological effects. Higher levels of HCHs, DDT, and PCBs are found in the Russian lakes.

Exceptionally high HCH levels are found in Russian river water, especially the Ob. Ratios of γ -HCH to α -HCH indicate use of lindane.

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. TEQ levels of OCs in most freshwater, marine, and estuarine sediments exceed some guidelines for protection of aquatic life.

PCBs and Σ DDT 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 PCBs 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.

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 over-wintering habitats.

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 PCBs, in fish at higher trophic levels.

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 Σ DDT and Σ PCB levels, reflecting accumulation of OCs at wintering grounds farther south as well as from preying on migratory birds in the Arctic.

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.

High PCB levels were found in polar bear with highest levels in some individuals from the Svalbard population.

3.5.1.2. Effects

The influence of contaminants on fetal and neonatal development is of special concern. Preliminary results indicate that POP and methylmercury concentrations are two- to ten-fold higher in breast milk and cord blood in some Arctic areas than in breast milk and cord blood from regions south of the Arctic. The fetus and the neonate are very vulnerable to the effects of many of these contaminants during this critical period of development.

There is both scientific and public concern about the possible adverse effects of POPs on pregnancy outcome, fetal development, child development, reproduction, male and female fertility, and the immune system. Several of these effects may be mediated through endocrine disrupting properties of some POPs. DDT and its metabolites and some dioxin and PCB congeners have been implicated.

The high exposure of indigenous peoples to methylmercury in some Arctic areas is a matter of concern because of its neurotoxic effects on the fetus. Further investigation of both the levels and the influence of mercury on fetal development is warranted.

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.

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/Fs and planar PCB levels expressed as TCDD TEQs are in the same range as those associated with immunosuppression.

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, Σ PCB 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.

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.

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 (PCBs, dieldrin, non- and mono-ortho PCBs, 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 Σ PCB levels that exceed some reproductive effects thresholds.

Harbour porpoise from the southern Barents Sea have Σ PCB levels that exceed those associated with reproductive effects and immunosuppression. Some walrus populations, most notably those where some individuals feed on ringed seal, have Σ PCB 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. PCB 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.

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 that exceed some threshold levels associated with potential risk for reproductive and immunosuppressive effects, as well as subtle neurobehavioral effects in offspring.

 Σ PCB 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.

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.

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, over-hunting, 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.

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.

3.5.1.3. <u>Trends</u>

3.5.1.3.1. Spatial

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 PCBs 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.

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.

3.5.1.3.2. Temporal

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 HCHs 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.

Studies of freshwater sediment cores show recent declines in PCDD/Fs deposition following major increases after the 1940s. Sediment cores from lakes in the North American High Arctic show later onsets of Σ PCB inputs (in the 1950s) compared with cores from sub-arctic 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) that could confirm whether observations in the North American High Arctic reflect 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 PCBs were found to be similar for freshwater sediments and the ice core.

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 sub arctic (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 1970's and the 1980's, after many POPs were restricted or banned. Based on the more precise studies in Sweden, PCB, DDT, PCDD/F, HCH, and HCB levels have declined in biota in recent years. The decline seems to be slower for PCBs. This may indicate continued low-level leakage of PCBs to the environment from unknown or poorly studied sources. From the Canadian seabird data, it is not clear whether this decline continued from the 1980's to the 1990's. OC levels did not decline in peregrine falcon between the 1980's and 1990's.

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 1970's and 1980's, 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 1970's and early 1980's 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.

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.

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 that would allow regular monitoring of key populations and retrospective analysis for contaminants, including newly detected contaminants.

3.5.2. Metals

The distribution of metals among the various environmental compartments of the Arctic is dynamic and driven by natural sources, processes, and environmental factors. Significant anthropogenic inputs of metals are detectable against the highly variable natural background on local scales, commonly in the order of tens of kilometers or less.

Metals are taken up by Arctic biota and their levels often reflect local geology or local anthropogenic activities.

Of the metals considered, the most important in the Arctic biosphere is Hg because it occurs in some biota at concentrations that may have health implications for individual animals or may have implications for human consumers.

3.5.2.1. Levels

Existing data from the literature do not allow a valid estimate of spatial and temporal trends of exposure of Arctic peoples to mercury, while for lead, a declining trend is observed.

The high exposure of indigenous peoples to methylmercury in some Arctic areas is a matter of concern because of its neurotoxic effects on the fetus. Further investigation of both the levels and the influence of mercury on fetal development is warranted.

Lead levels in Arctic indigenous peoples have declined since the implementation of controls on lead emissions. Concentrations of lead in blood currently reported are below a level of concern, however, continued monitoring is warranted because of the potent effects of lead on neurological development in the fetus and children.

The highest concentrations of atmospheric heavy metals in Arctic air occur near smelter complexes on the Kola Peninsula and at Norilsk and result from emissions from these smelters.

Near point sources such as mine sites and some Russian estuaries, heavy metals exceed background levels up to 30 km from the source.

Heavy metal concentrations in air in the High Arctic are one order of magnitude lower than concentrations in other remote locations and about two orders of magnitude lower than the concentrations around major point sources in the Kola Peninsula. Air concentrations measured on the Kola Peninsula are comparable with the concentrations in the most polluted regions of Europe and North America.

Background levels in soil, lakes, rivers, and oceans generally fall within the global ranges.

Mercury levels in marine mammals are high, but do not exceed the highest global levels. Lead levels in large parts of the Arctic are at the lower end of global background.

In almost all cases, Pb levels in marine organisms from the Arctic are well below food standard limits; however, this is not the case for hot spot areas such as mining areas and some Russian estuaries.

3.5.2.2. <u>Effects</u>

It is uncertain whether Hg poses a health threat to the most highly exposed groups of marine mammals, those in the western Canadian Arctic as well as pilot whales from the Faeroe Islands.

3.5.2.3. <u>Trends</u>

Few spatial or temporal trends are apparent in the existing data, largely due to poor temporal or spatial coverage or to irresolvable artifacts in the data related to differences in sampling, analytical, and reporting protocols.

3.5.2.3.1. Spatial

The concentrations in surface deposition around the sources, e.g., on the Kola Peninsula, decrease by between one and two orders of magnitude within 10-100 km from the emission source. The concentrations within the large area of the High Arctic are uniformly distributed, varying by a factor of 2-3.

The concentrations of trace elements in marine sediments are dependent on local geology, particle size, the amount of organic matter, and anthropogenic influence. The background geographical distribution of Pb and Hg in marine sediments is related to the geological provinces of the Arctic.

Regional geographical differences in metal concentrations of benthic flora and fauna as well as in those of fish are not very apparent. For some metals, seasonal and local trends in some stationary marine species are larger than regional differences in baseline data.

3.5.2.3.2. Temporal

The concentration of most heavy metals measured in sub-arctic air has decreased during the last two decades. All the heavy metals show strong seasonal variation in the High Arctic.

Mercury in Arctic sediments shows an increase over time, indicating a widespread regional process. As the anthropogenic fluxes do not show the same pattern, further investigations are needed before firm conclusions can be drawn.

Temporal trend data are scarce in Arctic biota. There is some evidence of Hg increasing by a factor 2-3 in some marine mammals within the last two decades. Only liver, and in certain cases kidney, shows such increase. It remains uncertain, however, whether this is a real increase or reflects year-to-year variation. Mercury concentrations in human and seal hair from the 15th century are 2-3 times lower than present-day samples.

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4. MAJOR PATHWAYS OF CONTAMINANTS TRANSPORT

The objective of this chapter is to provide a basic understanding of the physical processes and pathways, both within and between the main environmental compartments, which determine the fate of contaminants in the Arctic environment. The three compartments considered are, the atmosphere, oceans and freshwater. For each compartment, local (within the region) and long-range (into and out of the region) transport mechanisms are identified. In other regions of the world, the groundwater compartment and biological transport are included in the discussion. Since the Arctic is defined in part by the permafrost zone, there is essentially no groundwater. Concerning the long-range biological transport of PTSs, this mechanism has not been studied extensively in the Arctic and is not reported here. It is not expected to be of the same order of magnitude as the major transport mechanisms.

The following section introduces the general and regionally specific features of PTSs pathways in and into the Arctic. Subsequent sections give a detailed discussion of pathways and transport in the three environmental compartments.

4.1. INTRODUCTION

4.1.1. General Features

The contaminants found in the Arctic as a result of transport over long distances are those that have been produced in large quantities and are environmentally stable. Properties which promote the long-range transport of pollutants into the Arctic via the atmosphere are, large-scale and geographically widespread emissions, low water solubility, chemical stability and the presence of the chemical in the gas phase or on small particles with atmospheric residence times of several days or longer. These properties are also important with respect to long-range riverine and marine transport, with the additional requirement that these compounds must be biologically stable.

The sources of most contaminants of interest to AMAP generally lie outside of the Arctic region; however, there are some internal regional sources, especially from mineral extraction and related activities. This situation applies in the case of some industrial compounds as well.

The pathway from the environment to humans is not clear, and seems to involve several sources of exposure, not just those related to contamination of food. As a consequence, human exposure estimates based only on results of environmental monitoring of consumed foodstuffs are not generally adequate. Though food is the major exposure route for contaminants in the Arctic, human exposure to environmental contaminants also varies according to the amount and type of food consumed and the method of food preparation, not just the level of contaminants in the food. For these reasons, wildlife-monitoring data provide a very uncertain basis for precise human exposure estimates. However, they are of great value for risk characterization since they identify both the contaminants present in wildlife used as traditional food and the most contaminated species, and, therefore, can contribute to the basis for dietary recommendations.

4.1.2. Regionally Specific Features

4.1.2.1. <u>POPs</u>

Persistent OCs of concern in the Arctic originate mainly 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 \rightarrow atmospheric transport \rightarrow deposition to the terrestrial environment on snowpack \rightarrow snowmelt \rightarrow river transport to lakes or estuaries \rightarrow transport to the ocean \rightarrow 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.

4.1.2.1.1. DDT/DDE/DDD

The use of DDT has been declining in the temperate regions of the Northern Hemisphere since the 1960s and especially since the mid-1970's when it was banned by many western nations. Its persistence in the environment,

and its continued entry into the Arctic region via long-range atmospheric transport and in some river systems, has meant that it is detectable in almost all compartments of the ecosystem and in human tissues.

4.1.2.1.2. Toxaphene

Toxaphene, also known as camphechlor, is an insecticidal mixture of over 670 chemicals. It enters the Arctic region via long-range atmospheric transport. Significant amounts have been reported in the Yukon Territory and coastal regions of Arctic Canada.

4.1.2.1.3. Hexachlorocyclohexanes (HCHs)

HCHs are a group of organochlorine pesticides: alpha- (α -), beta- (β -), gamma- (γ -) (more commonly known as lindane), delta- (δ -) and epsilon- (ϵ -) are the most stable isomers. Many countries in the world use large amounts of lindane. HCH isomers are characterized 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.

4.1.2.1.4. Mirex

Mirex has never been used in any of the circumpolar jurisdictions. It has been manufactured and applied extensively in the continental USA and has become widely distributed via long-range atmospheric transport.

4.1.2.1.5. Chlordane, oxychlordane and cis- and trans-nonachlor

Technical chlordane is a mixture of chlordane, nonachlor, heptachlor and other chlordane isomers. It is not registered for use in circumpolar jurisdictions and enters the Arctic ecosystem primarily via long-range transport through the atmosphere.

4.1.2.1.6. Dioxins (PCDDs) and furans (PCDFs)

Polychlorinated dibenzodioxins and polychlorinated dibenzofurans are two structurally similar families of compounds that include 75 congeners (different compounds) and 135 congeners, respectively. These compounds enter the Arctic ecosystem almost exclusively via long-range atmospheric transport. Local sources are poorly documented. They include waste incineration, wood burning and metallurgical industries.

4.1.2.1.7. PCBs

Polychlorinated biphenyls (PCBs) are a group of 209 structurally similar compounds (congeners) commercially produced as mixtures. Although PCBs have been released due to improper use, storage and disposal, or accidents at military sites in the Arctic, they have primarily contaminated the Arctic ecosystem through atmospheric transport from regions farther south. Their manufacture and new usage is banned in most circumpolar jurisdictions, but they are still present in older electrical transformers and at a number of contaminated industrial sites and waste sites throughout the Arctic.

4.1.2.1.8. Hexachlorobenzene

Hexachlorobenzene (HCB) was widely used as an anti-fungal agent for various seed crops and is also an important industrial feedstock for production of chlorinated solvents and pesticides (Government of Canada 1993). HCB is still widely used in the world and atmospheric transport is a major pathway to the Arctic.

4.1.2.2. <u>Metals</u>

In the case of POPs, the atmosphere is the predominant pathway. In the case of HMs, the situation is somewhat different. The four major pathways are briefly introduced in the following sub-sections.

4.1.2.2.1. The Atmosphere

Following release into the atmosphere, heavy metals can be either deposited in the vicinity of the emission source or subject to long-range transport via air masses. In most cases (except Hg and to some extent Se), emission of metals occurs on particles. The size of emitted particles, as well as the temperature and speed of exhaust gases and the height of the emission source, are the major factors influencing the relative proportion of metals transported locally or over long distances.

During winter, about two-thirds of the metalss in air in the High Arctic are transported from Eurasia, particularly from the Kola Peninsula, the Norilsk region, the Urals, and the Pechora Basin. Five to ten percent of these emissions are deposited in the High Arctic. The remaining one third of the metals in High Arctic air in winter is

transported from industrial regions in Europe and North America. In summer, local sources dominate the contamination of the High Arctic.

In the case of lead, the evidence for long-range transport is uncertain. Industrial and vehicle-exhaust emissions are regarded as being the most important sources of environmental lead. As such, most of the Arctic is expected to be an area with low human lead exposure. However, in Greenland, blood lead levels have been reported that are comparable to those found in West European cities (Hansen 1981, Hansen et al. 1983, 1984). The relatively high blood levels in Greenland have still not been explained, but could possibly be a result of long-range transport of lead by atmospheric particles. The studies of Murozomi et al. (1969) and Rahn and McCaffrey (1980) both indicate that a combination of mid-latitude pollution and meteorological conditions could account for the lead exposure in the Arctic.

4.1.2.2.2. Rivers

Rivers are one of the major pathways of contaminants 1) to the Arctic region from the lower latitudes and 2) to the Arctic marine ecosystem from the Arctic terrestrial ecosystem. The speciation of metals in rivers is controlled by the physico-chemical properties of individual metals and by river chemical conditions, particularly the presence of complexing ligands such as natural organic compounds. Concentrations of natural organic matter in the Mackenzie River and in Siberian rivers are similar but much lower than those of the Russian Euro-arctic rivers (North Dvina and Pechora). Though water discharges by the Siberian rivers are much higher than those of the North American rivers, sediment transport by Siberian rivers is significantly lower (except the Yana, Indigirka, and Kolyma rivers in Eastern Siberia). For example, the Mackenzie River sediment load is seven times higher than that of the Yenisey River.

Riverine transport of heavy metals toward the Arctic Basin is approximately half the atmospheric contribution for metals like Pb and Cd, while for others such as Zn, the rivers are more important, carrying five times the atmospheric load. Such mass balance calculations will change considerably with the distance from the sources and the time of year, since the source contributions are strongly seasonal.

4.1.2.2.3. Oceans

Contaminants, including heavy metals in water or ice in the Arctic marine environment, are transported directly by ocean currents. Ocean circulation is driven by a combination of forces. A particular force can dominate in a particular geographical area; for example, tidal forces are dominant in many channels of the Arctic Archipelago whereas wind stress is most important for surface currents in the Canadian Basin (Barrie et al. 1992).

4.1.2.2.4. Ice

Various pollutants, including heavy metals, deposited on the sea ice from the atmosphere can be transported long distances in the ice or in blowing snow and then released to the atmosphere or the ocean during the melt processes. In addition, there is also a vertical transport, because growing sea ice rejects salt from the ice matrix in the form of dense brine, which drains into the surface waters beneath. This vertical convection thus enhances exchange between the surface and deep ocean compartments (Gade et al. 1974).

4.2. OVERVIEW OF EXISTING MODELLING PROGRAMMES AND PROJECTS

4.2.1. Introduction

A complete review of modeling as it relates to the physical and chemical fate of contaminants in the various compartments of the Arctic environment is beyond the scope of this report. Consequently, only a summary of some of the more relevant models that have been adapted for northern conditions, or specifically applied to contaminant transport in the Arctic are reviewed here. Within the terrestrial/freshwater compartment, only models related to the fluvial transport and fate of contaminants in freshwater systems will be considered. However, some of the atmospheric models do consider the soil surface in the context of sources and exchanges. It appears that data gaps and data inconsistencies among different compartments remain a problem for these kinds of linkages.

To date, there has been no attempt to fully integrate all of the compartments in a single model, due to the complexity that would be required. However, the recent advances in atmospheric models directed at understanding contaminant transport from temperate source regions to the Arctic and the exchanges between the atmosphere and the land and ocean in the northern hemisphere have made significant progress toward this end.

4.2.2. Atmospheric modeling

For most contaminants, the atmosphere provides the fastest transport medium. Therefore, in order to understand how contaminants are brought to the Arctic, it is important to quantitatively calculate the atmospheric transport in an explicit manner. Furthermore, having established a measurement-validated modeling tool, the model can be used to estimate the relative importance of different emission sources, to evaluate possible effects of new emissions, and to increase our understanding of the relative importance of various processes involved in the transport. There are two types of models used for contaminant pathways studies: 1) three-dimensional atmospheric models suitable for studying the movement of one-hop compounds and, to a lesser extent, multi-hop compounds; and, 2) two-dimensional multi-compartmental models suitable for multi-hop compounds. Both types are usually run on a domain that is global, although single-hop compounds can often be studied effectively with a hemispheric scale model.

There have been two attempts at simulating the dispersion of OCs on a zonally-averaged, global scale. Strand and Hov (1996) modeled the distribution of HCHs using a 2-D, atmospheric model (Bergen Model), while Wania and Mackay developed a multimedia compartment model for OCs based on the fugacity approach (Toronto Model) (Wania and Mackay 1993, Wania 1994, Mackay and Wania 1995, Wania and Mackay 1995).

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

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

4.2.3. Freshwater systems

The river system as a whole, with all its processes, from watershed runoff and riverine processes to the discharge via estuaries into the ocean, is not yet adequately represented by a single model or even coupled models. As well, unlike atmospheric and oceanic models, which are larger in scale and not confined by national boundaries, freshwater models tend to be country-specific and are presented here.

Norway

A new computer model system has recently been developed in Norway. This River System Simulator (RSS) combines 14 different models to describe river processes, with particular emphasis on the environmental effects of river regulations.

The 14 models included in the River System Simulator can be grouped into four main categories. These are:

- input from the watershed into the river system;
- hydropower system simulation models;
- physical, chemical, and biological processes in rivers and lakes; and,
- consequences for humans and ecosystems.

Although the RSS has proven very useful in its applications, there has been no consideration, apparently, of using this model to predict the effect of these changes on contaminant transport within the system, though this project 108

seems quite feasible. More effort should be put into applying the RSS to help understand contaminant transport and fate in Arctic river systems.

Canada

Much of the modeling work in Canada has focused on the Mackenzie River, especially hydrology and ice.

Other modeling efforts have looked at evapo-transpiration from Arctic wetlands, snowfall, and fresh snow and snowpack processes, but there is as yet no attempt at integrating these on a larger scale.

Data from an intensive study of the processes controlling contaminant fate at Amituk Lake, NWT (Diamond et al. 1996) has been used to attempt to fully integrate physical process data with contaminant fate data. These modelers built a general, whole lake, mass balance model based on the QWASI model (Quantitative Water Air Sediment Interaction), developed originally by Mackay and coworkers (Mackay 1991). The model uses equivalence as the equilibrium criterion, which is suitable for most chemicals, rather than fugacity, which is suitable for VOCs only. A multi-species time-dependent model was developed to represent the large proportion of meltwater and chemical loadings that flow through the lake without mixing with the water column, because of the rapid and dominant spring freshet. The model also accounts for ice cover for up to 46 weeks of the year, during which no air-water exchange of chemicals occurs, and contaminant concentrations in the water column may increase as they are excluded from the ice layer.

United States

Model development in the United States parallels that in Canada. Some examples of these models include Kane et al. (1993) for energy-related modeling of snowmelt and Cassell and Pangburn (1991) who modified the Streamflow Synthesis and Reservoir Regulation (SSARR) model to account for cold region effects.

Russia

A deterministic, mathematical model has been developed for runoff, called the HYDROGRAPH SHI-96. It covers all types of runoff and may be applied to any physiographic region and basins of any size. The design interval of the model is daily or shorter and the model input includes precipitation amount and duration, air temperature, and air humidity deficit. The model generates a continuous hydrograph for the design interval. Other information required by the model includes:

- formation and melting of snow cover;
- evapotranspiration;
- infiltration and surface runoff;
- dynamics of soil moisture and drainage water;
- formation of underground runoff;
- runoff transformation as part of overland flow and within the channel; and,
- runoff at the outlet.

This model is part of the 'Runoff – Erosion – Contamination' System model (V. Vuglinsky, pers. comm., State Hydrological Institute, St. Petersburg, Russia, 1996).

4.2.4. Marine system modeling

4.2.4.1. <u>Types of models</u>

4.2.4.1.1. Ice models

Häkkinen and Mellor (1992) developed a coupled dynamic model that will predict ice motion, which is primarily driven by winds. The model is built on governing ice equations. The dynamic prognostic variables are ice concentration, mass, and velocity.

4.2.4.1.2. Thermodynamics

In the model, the conductive heat flux at the sea-ice surface is balanced by the atmospheric heat flux, including radiative, sensible, and latent components, if surface temperature is below freezing. If there is a net heat gain at the

surface and the surface is at or above freezing, the heat gain is used to melt the snow and ice. At the ice/water interface, any imbalance between the conductive heat flux from the ice slab and the sensible heat flux from the water column results in either freezing or melting.

A common conclusion from the ice modeling studies is that the snow-ice system has its largest sensitivity to the surface albedo. In effect, all other variability in surface forcing components, such as cloudiness and sensible and latent heat flux, are secondary compared to the albedo effect. However, once one defines the albedo model, in which the Arctic sea ice will not vanish during summer in the present climate, the other forcing components, i.e., oceanic heat flux, ice divergence, snowfall, cloudiness, and air temperatures (and winds), also show strong sensitivity in determining the Arctic ice mass. Considering all the uncertainties in all of the forcing components listed above, the ice models appear to be relatively stable in giving an average Arctic sea-ice thickness of about 2.5-3.3 m when climatological radiative and turbulent heat fluxes, snowfall, and oceanic heat fluxes representative of the central Arctic are used.

4.2.4.2. Ocean Models

The discussion here is restricted to three-dimensional, primitive equation numerical ocean models which include description of temperature and salinity variations and which have been used in large-scale Arctic studies in a coupled ice-ocean model. These models have the potential to describe contaminant transport. For mesoscale ice-ocean models and ice-ocean process studies, the reader is referred to a review by Häkkinen and Mellor (1990).

4.2.4.2.1. Coupled ice-ocean, Arctic modeling studies

In the Arctic Ocean and the peripheral seas, the Greenland, Iceland, Norwegian, and Barents Seas, the overall stratification structure is determined by both river runoff and Atlantic and Pacific inflow, and by dynamic and thermodynamic interactions with the ice cover. As a result of heat exchange with the atmosphere, deep waters are formed in this region, a process that is augmented by brine rejection due to ice formation. Alternatively, since density is strongly salinity-dependent, freshwater fluxes at the ocean surface are especially important because of their stabilizing effect. In the Arctic Ocean, the freshwater layer prevents heat exchange between upper and deeper parts of the water column, and in the Greenland Sea, an excess freshwater cap in the form of ice can prohibit the renewal of deep waters.

4.2.4.2.2. Partially diagnostic models

Hibler and Bryan (1987) were the first to present results from a coupled ice-ocean model for the Arctic. The ocean model was partially diagnostic for the deeper ocean, in that a Newtonian damping factor forced the temperatures and salinities toward climatological values, while the upper ocean could adjust prognostically to the surface forcing determined by ice freezing and melt. The main result from the simulations was to show the importance of the northward heat transport by the Norwegian Atlantic Current, which is responsible for determining the ice extent in the Greenland and Barents Seas. This same heat source is responsible for year round ice melt in the Greenland Sea. The coupling also intensified the oceanic Beaufort Gyre and the East Greenland Current, which the authors described as a readjustment to the forcing because the initial salinity and temperature fields were smoothed estimates of the observations.

The climatology of the Arctic ice thickness field supports high ice thicknesses north of Greenland due to the mechanical pileup of ice transported by the Transpolar Drift to the vicinity. However, the model, using surface forcing from 1979, produced a highly anomalous ice thickness field with a large ice buildup along the East Siberian coast and a weak build up north of Greenland.

Another coupled ice-ocean model by Piacsek et al. (1991) is also partially diagnostic. The Hibler ice model is coupled to a high resolution mixed layer, where turbulence is calculated according to the level 2.5, Mellor and Yamada turbulence closure model (Mellor and Yamada 1982). However, the deeper ocean is diagnostic with a geostrophic velocity field determined from Levitus' (1982) climatology. Corresponding to a perpetual year, 1986 forcing, the model produces a very realistic seasonal variability, with the exception of the wintertime ice extent in the Barents Sea. They consider the inclusion of the mixed layer dynamics to give a superior ice thickness field compared to coupled models without mixed layer dynamics. The mean oceanic heat flux in the model varies in the ice-covered area from 5 to 15 W/ m^2 , attaining even larger values northeast of Spitsbergen in the area of the submerged Atlantic waters. These values are on the high side compared to the traditional view of about 2 W/ m^2 , and to even lower values, less than 0.5 W/ m^2 , as suggested by other one-dimensional modeling studies (Mellor and Kantha 1989). Comparison of observed buoy tracks and simulated drift tracks are in reasonably good agreement considering the coarse resolution of the model.

4.2.4.2.3. Prognostic models

The first comprehensive prognostic ocean model for the Arctic Ocean was described by Semtner (1976a) using the Bryan-Cox- Semtner (BCS) ocean model. The model was later expanded by Semtner (1987) to include a dynamic-thermodynamic ice cover comprised of a three-level snow-ice system (Semtner 1976b) and an ice rheology simplified from the model by Hibler (1979). This ice-ocean system was driven by monthly surface forcing and specified inflow-outflow fluxes at the boundaries as in the Semtner (1976a) model.

Overall, the results from the ice model component showed agreement with the observed ice extent, but ice thicknesses were much lower compared to the generally accepted average values of 2.5-3 m. Surface salinities were reasonably well reproduced in the model, except that their gradients were not strong enough in the central Arctic.

This model has been applied to the study of inter-annual ice variability by Fleming and Semtner (1991) for the period of 1971-1980. Their main conclusion is that using inter-annually varying forcing produces much improved sea-ice cover variability for the annual cycle, compared to the model forced by mean monthly climatology.

4.2.4.2.4. Evaluation and verification of model results

There now exist fairly sophisticated, numerical models for ice cover and oceans. They appear to give fairly realistic results even though they are mostly coarse resolution models. The basic criterion of evaluating an equilibrium ocean model is the question of how well it can reproduce the seasonal hydrography. The comparison is inadvertently limited to scalar variables instead of velocity fields of which we know very little except for coarse features, such as the cyclonic flow of the Atlantic origin waters around the Arctic Basin. Remote sensing gives an ideal way to validate ice model results, at least for ice concentration fields. Also the Arctic Buoy Program can provide information on ice drift for validation purposes. The least known quantity observationally is the ice thickness. To date, the best means of measuring ice thickness is through submarine sonar observations, but this type of data is classified, and thus is largely unavailable for monitoring and validation.

4.2.4.3. Modeling of oceanic contaminant transport in Arctic seas

The starting point for modeling the transport of contaminants in Arctic seas is a coupled ice-ocean model as described above. In addition, a transport model is needed, either as an extension integrated into the ocean model or as a separate model driven by the results from the ocean model.

Depending on the contaminant in question, the transport model can be more or less complicated. If the contaminant is dissolved in seawater, it is treated as a passive tracer. This can be done in two ways. In the Eulerian approach, an advection- diffusion equation is solved for the development of the tracer concentration in a similar way to the equations for salinity and heat in the ocean model itself. An alternative is the Lagrangian, or particle-tracking approach. Here, trajectories of passive particles representing the contaminant are calculated from the current field of the ocean model. Both methods are well developed, and have well-known good and bad properties.

If the contaminant is bound to particles, the situation is a lot more complex. The particle may go into the sediments, be resuspended, captured in ice, etc. The physical, chemical and geological processes are not known well enough. Realistic models are not available at present. Further process studies and modeling are needed to gain knowledge before these processes can be incorporated or parameterized into regional or large-scale pollution transport models.

There has been relatively little activity on modeling of dispersion of oceanic contaminants in Arctic areas. Recently, the activity has increased somewhat, but most of the results are so fresh that they have not yet reached the reviewed literature. This increased activity is mostly concerned with the potential threats of nuclear contamination from former Soviet sources, in particular in the Kara Sea.

The largest modeling effort has been in the USA under the Arctic Nuclear Waste Assessment Program (ANWAP). The model used is the coupled ice-ocean model of the US Naval Research Laboratory. This model consists of a Bryan-Cox ocean model coupled with a Hibler ice model. The modeling work is reported in Preller and Edson (1995).

More regional work for the Kara and Barents Seas has been done by Harms (1997). He uses the Hamburg Shelf Ocean Model (HamSOM) documented by Backhaus (1985), together with an ice model based on Hibler's model. The Eulerian transport model is used to study the dispersion from waste dump-sites east of Novaya Zemlya.

Canada's Institute of Ocean Science (IOS) uses a prognostic ocean model coupled with a sea-ice model to investigate how a tracer is transported within the Arctic Ocean. A novel feature of the IOS model is the representation of the subgridscale eddies as a driving force in the mean circulation rather than as traditional eddy viscosity. This eddy force ('neptune') was described by Holloway (1992) and implemented in models by Alvarez et al. (1994). Other features of the IOS model include the use of flux corrected transport (FCT) after Gerdes et al. (1991).

4.3. ATMOSPHERE

4.3.1. General features

The atmosphere is the most important pathway relative to ocean and terrestrial/ freshwater pathways for one-hop compounds (acids, metals except mercury, involatile organics). Given the current configurations of anthropogenic sources at mid-latitudes, the most favored pathway into the Arctic is from the Eurasian continent from November to May. The relative importance of atmospheric transport compared to marine and terrestrial/ freshwater is very contaminant-specific for the multi-hop compounds, such as OC pesticides, Hg, and PCBs. While all of the compartments play a role in transporting these contaminants, the speed of transfer through the atmosphere suggests that this compartment is particularly important in the global cycling of these types of compounds. The air-shed for the Arctic is the northern hemisphere for one-hop contaminants, but is global for multi-hop contaminants.

The occurrence of contaminants in the Arctic is influenced by the nature and rate of emissions from sources, the processes by which these compounds are transported to the Arctic, removal processes, and the exchange of compounds between the different compartments. Generally, the atmosphere contains a relatively small amount of a contaminant compared to the total amount in other environmental compartments (e.g., for HCH, see Strand and Hov 1996). The importance of the atmosphere is that it provides a significant mode of rapid transport of contaminants from source areas to the Arctic region. Transport times of contaminants via air currents are rapid compared to those in the oceans. The time for an air parcel to completely mix in the troposphere of the northern hemisphere is of the order of six months. Mixing in the region north of 30° N is of the order of three months (Plumb and Mahlman 1987). In contrast, transport times of water parcels in northern marine systems are measured in years and decades.

4.3.2. Single and multi-hop pathways

Atmospheric transport pathways can be subdivided into two types: one-hop pathways and multi-hop pathways. One-hop pathways describe the movement of compounds that are emitted to the atmosphere, transported, and then deposited to the surface, never to return to the atmosphere. In such cases, the source region of an Arctic contaminant is simply defined by its source distribution, its lifetime in the atmosphere (governed by removal processes), and atmospheric circulation. This applies to acids, heavy metals with the exception of mercury, and nonvolatile, particle-bound organics, such as benzo(a)pyrene [B(a)P], black carbon particles, and radionuclides. The pathways of these constituents follow that of Arctic haze from mid-latitudinal sources into the Arctic (Barrie 1986, Barrie 1995, Barrie 1996). Single-hop compounds are marked by longer atmospheric residence times in winter (~20-30 days) compared to summer (~3-7 days), as well as stronger south to north transport into the Arctic from Eurasia in winter than in summer.

With multi-hop pathways, a compound re-enters the atmosphere after initial deposition to the Earth's surface, and continues over time to move through the environment in multiple hops. Processes by which this can occur include volatilization from the Earth's surface under temperatures warmer than during initial deposition; sudden exposure to the atmosphere of ocean water saturated with a volatile contaminant after being covered by ice; and, resuspension by wind, dust or snow. For multi-hop compounds, the source region affecting the Arctic is not only defined by atmospheric transport, removal and circulation, but also by surface processes that control its re-entry into the atmosphere. Mercury, most organochlorines (OCs), and many PAHs fall into the multi-hop group.

4.3.3. Atmospheric transport

Barrie (1992) and Iversen (1989a, 1989b, 1996) have discussed the role of atmospheric circulation in the transport of contaminants from mid-latitudes to the Arctic region.

In winter, the lower tropospheric circulation of the northern polar region is dominated by high pressures over the continents and low pressures over the northern Pacific and Atlantic Oceans. In particular, the intense Siberian high-pressure cell tends to force air on its western side northward into the Arctic. The high-pressure ridge over

North America generally drives air out of the Arctic southward. The mean flow in winter is out of Eurasia into the Arctic, and out of the Arctic into North America. Some air is also exchanged with the south when low-pressure vortices along the Arctic front mix warm southern air with cold northern air in a large-scale turbulent eddy.

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

Contaminants having ground-level sources at mid- and low latitudes will, on average, experience rising air motions during their atmospheric transport to Arctic areas. This is a consequence of the temperature distribution in the atmosphere and the energy conversions driving the air motions. With few exceptions, atmospheric flow systems convert potential energy to kinetic energy through rising flows of relatively warm, light air, and sinking flows of relatively cold and heavy air. The reservoir of potential energy remains unchanged as the tropics are warmed by solar radiation, while the polar areas are cooled by terrestrial outgoing radiation. Consequently, contamination at higher altitudes (2-8 km) in the Arctic generally originates from lower latitudes than contamination in the lower Arctic atmosphere (0-2 km), the latter originating in colder and more northerly regions (Carlson 1981, Iversen 1984). Since many contaminants are water-soluble, and rising motions frequently are connected with precipitation release, upper level atmospheric Arctic contamination is generally less concentrated than in the lower atmosphere. Nevertheless, it is characteristic of the Arctic that the vertical extent of anthropogenic aerosol particles is greater than at mid-latitudes (Hansen and Rosen 1984, Radke et al. 1984, Schnell and Raatz 1984, Barrie 1986, Pacyna and Ottar 1988). In summer and autumn, when temperature contrasts between major pollution source areas and the Arctic are smaller, dispersion of contaminants in the vertical is less pronounced than at other times of the year.

In contrast to the winter period, in the summer, south-to-north transport from Eurasia is much weaker as the Siberian high dissipates. Marked variation in cloud cover and precipitation accompanies this seasonal variation. In summer, temperatures in the High Arctic are near 0°C, allowing much more drizzling marine stratus than in winter, when temperatures between -25 and -45°C prevent buildup of moisture in the atmosphere. Photochemical activity during summer months is also higher, and provides an additional loss mechanism for some contaminants. Thus, during summer, transport from mid-latitudes to the Arctic is least for soluble, cloud-reactive or particle-bound compounds that are more easily removed by precipitation scavenging than insoluble or cloud-unreactive gases.

4.3.4. Atmosphere–surface exchange

The major contaminant pathways between the atmosphere and the surface include, snow, particle deposition, particle re-suspension from the surface, gas exchange in both directions, and rain. Transfers from the atmosphere to the Earth's surface in the polar regions are complicated by the presence of a seasonally varying intermediate media, namely, snowpack and ice. In the Arctic Ocean, the marginal seas undergo the greatest variations in percentage of ice cover. The snowpack plays an important role in atmosphere–surface exchange. During the winter, as snow accumulates on the surface as snowpack, it stores contaminants gained during its formation in addition to those delivered by particle dry deposition and gaseous exchange. The latter component may be negative (surface loss), or positive (surface gain). At present, there is no way of measuring it directly. However, there are indirect means that can be used to infer gas exchange. The net exchange between the atmosphere and the Earth is the result of all these processes.

4.4. OCEANS

4.4.1. Introduction

The ocean has been for a long time the final resting place for many anthropogenic waste materials. Because of its huge volume, the ocean has been thought a safe place for disposal, which would not pose any danger to humans. This view has to be reconsidered. Today, the ocean water column carries too many signatures of our society: radioactive isotopes, such as strontium and cesium, produced during nuclear-bomb detonations and in nuclear fuel reprocessing plants; pesticides, including DDT and its degradation products which have been found in all ocean organisms analyzed; chlorofluoromethanes (freons) which have been measured in the deep waters of the ocean; petroleum products which soil the surface of the ocean; and so on (e.g., Goldberg 1975, Dahlgaard et al. 1986, Krysell and Wallace 1988).

Once a contaminant has been introduced into the marine environment it becomes a matter of interest to determine where this contaminant goes and how its concentration varies with time and location. The processes by which contaminants are dispersed in the ocean fall into two basic groups: advection and dilution. Advection transports contaminants from one place to another, while dilution reduces its concentration by mixing (Williams 1979). To be able to understand environmental changes in the Arctic Ocean, it is important to have a good assessment of the present situation, as well as data on which chemicals are being added, subtracted, and transported from one place to another.

Our schematic knowledge of the general circulation in the Arctic Ocean has improved over the past decade due especially to tracer studies carried out during basin transects.

Tracer fields often give a direct measure of the rate of propagation of a property and, in the most widely cited example, radionuclide tracers from the European reprocessing plants have shown dramatically that transport times are much faster than earlier believed. However, tracer fields often do not provide good estimates of volume flows. Poorly constrained volumetric flows, together with a very poor knowledge of the contaminant concentrations (particulate and dissolved), mean that, at present, only approximate guesses of quantitative transport rates are possible within the Arctic Ocean. Although volumetric flows are better constrained at the entrances and exits from the Arctic Ocean, there is still considerable uncertainty and variance, and the contaminant concentrations are, as yet, poorly determined or not determined at all. Therefore, we need better documentation of volume transports of ice and water, and their contaminant burdens into, out of, and within the Arctic.

The Arctic Ocean is more dynamically coupled to its surrounding seas than believed earlier. However, little is known about the variability in the different seas and how they affect the main system. For example, river runoff is transported with relatively little dilution over remarkably large distances in the Arctic – more so than in any other ocean basin. Our understanding of the role of this redistributed river water in long-range contaminant transport is limited, as is the influence of variable riverine discharge on ocean circulation.

The Arctic is not in a steady state, but how it oscillates is still uncertain. Because most data are from the summer and have been obtained relatively recently, there is less known about winter conditions and inter-annual variability. The Beaufort Gyre seems to shift between a small gyre and a larger one, but the causes of these changes and their effects are unknown. The Atlantic layer seems to have warmed by about 0.5-1°C during recent years. The invasion of warm water, following boundary currents around the Eurasian and Canadian Basins, is accompanied by a shift in the front separating the Atlantic and Pacific water masses in the upper part of the Arctic Ocean. The fate of this displaced Pacific water, including its pathway out of the Arctic Ocean, is not known.

Transport pathways and mechanisms are not the same for all contaminants. Some contaminants tend to become bound to particles while others remain in the dissolved phase. Since particles tend to sediment and, therefore, take different pathways than dissolved constituents, it is crucial to understand both the phase geochemistry of the contaminant and the ocean processes of particle production and vertical flux. Currently, there are few data with which to estimate the relative importance of particles in the transport of contaminants and the movement of contaminants from surface water and coastal zones to deeper parts of the ocean.

4.4.2. Rivers Discharge

Being nearly landlocked, the Arctic Ocean receives a variety of materials from the surrounding continents, including both anthropogenic and naturally-occurring constituents. Contaminants discharged by rivers will most noticeably influence the coastline and nearby seas.

Ten percent of all riverine discharge to the world oceans occurs in the Arctic. The total discharge is about 3300 km³/y (Aagaard and Carmack 1989). The Arctic rivers Yenisey, Ob, and Lena are ranked as 5, 6, and 7 in the world, respectively, in order of annual discharge, and the Canadian rivers Mackenzie and Yukon are ranked as 11 and 18, respectively (Goldberg 1976).

Siberian rivers discharging into the Kara, Laptev, and East Siberian Seas have a huge combined drainage area of 9 000 000 km² extending far to the south (Shiklomanov and Skakalsky 1994), encompassing many industrial and agricultural regions. The Ob River and its tributaries originate as far south as 45°N (Futsaeter et al. 1991). While smaller rivers are frozen in the winter, some of the major rivers discharge year round. A unique characteristic of the Arctic Ocean is that the riverine waters can be traced throughout the Arctic Basin due to the extensive ice cover, which minimizes mixing.

4.4.3. Atmospheric deposition

Emissions to the atmosphere are returned to the land or the sea as wet and dry fallout as discussed earlier. Atmospheric transport is recognized as a major route for the transfer of contaminants to the ocean (GESAMP 1985). The time it takes for a contaminant released to the atmosphere on any continent to reach northern waters ranges from days to a few weeks, as compared to years by means of oceanic circulation (Gaul 1989).

Atmospheric contributions of OC compounds, such as PCBs and DDT, remain significant today. They are released into the atmosphere in the vapor phase and are transported as gases, aerosols, and adsorbed to particles (Duce et al. 1983). Eventually, the more persistent contaminants are permanently deposited in the sea floor sediments, but only after cycling through the water, biota, sediments and ice, perhaps many times. Once introduced into aquatic environments, synthetic organics partition to all components of the ecosystem. The insecticide DDT, found in the fat of polar bears, is a good example (Ballschmiter and Zell 1980).

In winter and spring, the Arctic atmosphere contains high levels of contaminants from Eurasia, and to a lesser extent from North America, known as Arctic haze. As contaminated particles settle out of the atmosphere, sea ice acts as a lid on the surface of the Arctic Ocean. Each year that ice floes drift, contaminants and other materials are deposited on its surface from the atmosphere in the form of snow, rain, fog, and dry deposition (Pfirman et al. 1995a, Chernyak et al. 1996). Heavy metals accumulating in the snow cover of central Arctic sea ice can reach values that are characteristic of snow deposits on sea ice near Siberian industrial areas (Melnikov 1991). Contaminants deposited on sea ice by atmospheric transport could percolate into the ice surface when melt-water refreezes, and could also be added to the ice underside when melt-water runs off and refreezes. During drift, contaminants concentrated in the oceanic surface micro-layer may also be incorporated in the ice (Gaul 1989).

The presence of sea ice in the Arctic Ocean generally inhibits ocean-atmosphere exchange. This is important because many contaminants, such as OCs, are semi-volatile. In winter, when surface water is being convected, the ice acts like a lid, keeping volatile contaminants introduced below it from entering the atmosphere. In summer, stratification of the surface ocean from sea-ice melt or river runoff limits atmospheric exchange to the top 5-10 m. Contaminants discharged in river water and frozen into the sea ice may not be released until the ice breaks up and melts. Polynyas and leads – places where there are breaks in the ice cover – may represent regions for exchange of volatile compounds including OCs (Barrie et al. 1992).

Because volatilization increases with temperature, some chemicals deposited from the atmosphere on the ice surface in winter are released back to the atmosphere in the summer when the surface warms and the snow melts (Barrie et al. 1992). If deposition occurs on drifting sea ice, release may occur far from the original incorporation location.

4.5. FRESHWATER

4.5.1. Introduction

The Arctic land mass, which includes freshwater lakes and river systems as well as the islands of the Arctic Ocean and adjacent seas, measures a full 13.4 10^{6} km² or about 40% of the total AMAP area. This compartment, referred to as the 'terrestrial/freshwater' compartment, is important in its own right in that it supports terrestrial and freshwater ecosystems, but also because its large surface area serves as a receptor for atmospherically transported contaminants.

Rather than considering each process and its role with respect to contaminant transport, this discussion will emphasize the processes that are unique or specific to the Arctic. In addition, the mass transport of water, sediment, and organic matter, which are important determinants for the transfer of contaminants in fluvial systems are quantified where possible.

Contaminants are delivered to the terrestrial/freshwater environment by the atmosphere and from direct discharges of wastes to land and water. Large river systems that drain to the Arctic, but that have a major portion of their drainage basin outside of the Arctic, including many of the rivers of Russia (e.g., Yenisey, Lena, Ob) as well as several Canadian Rivers (e.g., Mackenzie, Churchill and Nelson), are important long-range conduits to the northern marine environment.

4.5.2. Atmospheric deposition

The most important, large-scale contaminant delivery process to the terrestrial/freshwater compartment is deposition from the atmosphere through wet and dry particle scavenging as discussed earlier. The influence of

atmospheric contaminants on Arctic freshwater systems starts with their deposition onto surface waters (lakes, rivers, wetlands) or land surfaces in their catchment area. While the total surface area of some lakes, reservoirs, and rivers is large, it remains small relative to the total land surface area (< 1%).

4.5.3. Local wastewater discharges

Local development has a direct influence on water quality. In addition to the effects on the environment from land clearing, construction, and decreased permeability, the quality of water returned to the basin is usually diminished even after treatment. The disposal of industrial (oil and gas, mining and smelting) and municipal wastewaters is always a concern. Storm and melt-water runoff, which is usually routed directly to receiving water bodies without treatment, may be highly contaminated due to spills and localized atmospheric fallout. These factors can exacerbate the problems of providing clean water to basin residents.

4.5.4. Regional wastewater sources

Rivers draining to the Arctic Ocean receive contaminants from different sources within and outside their catchment areas. The Arctic rivers with vast basins may receive contaminants discharged in heavily industrialized zones and large cities located far to the south of the Arctic region. For example, in the territory of Russia, southern reaches of the large Arctic rivers include large industrial complexes and cities with populations often exceeding a million inhabitants. Two examples follow.

The Barents region of the Russian Arctic is the most highly populated and industrially developed part of the circumpolar Arctic. The province of Murmansk has over one million inhabitants with many in major industrial cities, including Murmansk, Apatity, Kirovsk, Kandalaksha, and Monchegorsk. In 1994, only 4.7% of the wastewater in the province was treated to specified standards. Approximately 6.1% of the wastewater (103.5 10⁶ m⁻³) was discharged into water bodies without any treatment and 13.6% (229.1 10⁻⁶ m⁻³) was considered insufficiently treated. The remaining 75.6% (1269.8 10⁻⁶ m⁻³) was classified as 'conditionally clean' and allowed to be discharged without treatment. Most of this latter group is generated by the cooling waters of the Kola nuclear power plant (NEFCO 1995). To the east of Murmansk is the province of Archangel, which, including the Nenets autonomous area, in 1993 had a total population of 1561 000 inhabitants with over 70% in major cities (including the cities of Archangel and Severodvinsk).

There are only five major cities in Canada that discharge effluents into rivers that drain to the AMAP region. These cities are Edmonton, Calgary, Regina, Saskatoon and Winnipeg with a total population of about three million. All of these are within the Nelson River system, with the closest, Winnipeg, being more than 1000 km from the outlet to Hudson Bay. It should be noted that all industries in these cities are required to discharge through the sewage treatment plants, except for uncontaminated wastes, such as cooling waters. Within the Canadian Arctic, the cities of Whitehorse, Yukon and Yellowknife, NWT are relatively small in population and have little industry.

4.5.5. Snowpack and snowmelt

Snow merits separate discussion for a number of reasons even though snow is just one form of precipitation in the Arctic and concentrations of contaminants in the atmosphere are, on average, higher in summer than in winter (Barrie 1995 and 1996, Pacyna 1995, Fellin et al. 1996, Oehme 1996). First, snow is an important medium for the wet deposition of contaminants in the Arctic, not only because winters are long and snow is the main form of precipitation, but also because snowflakes are effective scavengers (Gregor 1996). Second, the snowpack is a major storage variable in the overall water budget of Arctic systems, and contaminants accumulated in the snowpack during the winter are released to the underlying land and water during a short period of snowmelt in spring. Third, snowfall is to a large extent decoupled from the entire terrestrial compartment throughout the winter season because of frozen soil.

4.5.6. Hydrology

The hydrologic regimes of large river systems reflect the climate and physiography of the total watershed, as well as the nature of the river (i.e., reservoirs and lakes). As a result, large river systems tend to have a less well-defined hydrograph pattern. A unique feature of some of these large rivers is that because they flow north, their headwaters generally melt first. Thus, the peak flow is transferred downstream often resulting in a hydrograph significantly dominated by snowmelt, which will have taken several weeks to progress from the upstream areas, during which time downstream snowmelt will have supplemented the flood crest. This is well illustrated by the Mackenzie River in Canada and the North Dvina, Mezen, Pechora, Ob, Yenisey, and Lena Rivers in Russia.

4.5.7. Ice

Ice plays a unique and important role in the Arctic. River and lake ice restrict the exchange between the atmosphere and the water. River ice incorporates particles when it freezes to the bottom, as well as during anchor and frazil ice formation. River and lake ice also receive contaminants from atmospheric deposition throughout the ice-on season. During the spring melt, ice transport can gouge sediments and create ice jams, which can result in accelerated stream flows and associated sediment erosion.

River ice stores and transports contaminants, and may therefore be considered an indirect source of contaminants. Factual information about the rates of uptake of contaminants to the ice or the extent of their subsequent accumulation and transport is scarce.

The contaminants in the water, suspended matter, and bottom sediments are subjected to processes of ice trapping, transfer, and release. These processes are controlled by the varying patterns of freezing, ice growth, and ice cover decay, as well as by the water regime in the autumn-winter period. During freeze-up or ice jams, flow is reduced downstream of the ice accumulation. Ice-induced floodwaters upstream can replenish water in the flood plain (Prowse 1994). The reduced water velocity on the flood plain can result in sedimentation of fine-grained material and associated contaminants.

4.6. DATA GAPS

Much emphasis has been placed on the pathways for long-range transport of contaminants, nevertheless, the contribution of local Arctic sources of contaminants to the total contaminant transport budget requires further investigation.

Physical/chemical processes in the polar atmosphere are unique, and not as well understood or documented as those for temperate regions. Some of the processes that need to be understood better to improve the modeling capability and the general understanding of contaminant transport include:

- cloud, fog, and precipitation scavenging of contaminants
- chemical transformation of contaminants in air;
- air-surface exchange of contaminants;
- gas-particle phase partitioning within the atmosphere.

The role of ice in controlling air-water exchanges and the magnitude of contaminant loadings by ice should be evaluated.

The role of ice in remobilizing contaminants in rivers and in delivering contaminants to the ocean has received minimal attention.

Larger-scale studies and monitoring are required to more precisely measure the total mass of contaminants delivered to oceans and the temporal distribution of this delivery, as well as to characterize the particulate and dissolved phase contributions.

Tracers have been used in ocean pathway studies (e.g., radionuclides released from Sellafield), but greater use needs to be made of natural and anthropogenic tracers in all compartments. Emphasis should be placed on tracers that mimic the major contaminants, or are representative of groups of contaminants, thereby enhancing pathway studies. Selected PAH, PCB congeners, stable isotopes, and a range of other contaminants could prove to be very useful in this regard and need to be examined more fully.

It is essential that transport processes and their relative importance or magnitude within and between compartments (air, terrestrial, water, ice, sediments, biota) be determined. Prioritization would ensure that the most important processes are investigated.

In general, there is a qualitative understanding of the transformation and fate of different contaminants or groups of contaminants under varying Arctic conditions. However, detailed information and the ability to quantify these processes are lacking. In order to be able to fully assess the magnitude and direction of pathways, a better understanding of transformation and fate of pollutants in the Arctic is required.

Using multi-media models, a great deal could be achieved efficiently by fully integrating processes, observed levels, and trends. We would obtain assessment and feedback on; the design and implementation of future

monitoring and process research activities; management and/or mitigation measures; and the sensitivity of individual or linked processes. In particular, models have not been used to their fullest in the area of terrestrial/freshwater.

4.7. CONCLUSIONS

Atmospheric pathways are important in the overall context of the delivery of contaminants to the Arctic. The atmosphere is the most important pathway relative to ocean and terrestrial/ freshwater pathways for one-hop compounds (metals except mercury, involatile organics).

The relative importance of atmospheric transport compared to marine and terrestrial/ freshwater is very contaminant specific for the multi-hop compounds, such as OC pesticides, Hg, and PCBs. While all of the compartments play a role in transporting these contaminants, the speed of transfer through the atmosphere suggests that this compartment is particularly important in the global cycling of these types of compounds.

Given the current configurations of anthropogenic sources at mid-latitudes, the most favored pathway into the Arctic is from the Eurasian continent from November to May.

The air-shed for the Arctic is northern hemisphere for one-hop contaminants, but global for multi-hop contaminants. Models exist that quantitatively simulate atmospheric pathways.

The most accurate models for studying atmospheric transport pathways are those using globally or hemispherically gridded meteorological fields based on climatic observations. The density of meteorological observations in the Arctic is lower than elsewhere and, consequently, the accuracy of the predictions is reduced.

Field studies provide evidence that up to 80% of the total solute load in the snowpack is released with the first 20-30% of melt-water. Laboratory studies indicate that, whereas dissolved pollutants (organic compounds with high water solubility) are rapidly dispersed into the water at the onset of melting, those that are adsorbed (to particles) are more recalcitrant, and only leave the snow with the final melt-water.

From scattered ocean observations and deductions based on interpretation of Arctic processes, the following can be postulated (Roots 1982):

A considerable proportion of the dissolved and colloidally suspended material, organic or inorganic, which is delivered to the North Atlantic Drift, eventually reaches the Arctic Ocean.

Material reaching the Arctic Ocean in solution or suspension normally spends several years (and probably much longer if entrained in the Beaufort Gyre) circulating under very stable oceanographic conditions, mainly under an ice cover where photosynthetic reactions and gas exchange are much reduced. The extreme stability of the oceanic stratification probably retards the mixing of contaminants with deeper layers of water, but ensures that those, which do reach deeper levels, stay there.

Airborne contaminants which are deposited on the surface of the Arctic Ocean (typically originating from northern mid-latitude industrialized areas) become trapped on the surface of the sea ice, get flushed into the layer of comparatively fresh water that floods the ocean surface, undergo repeated freezing and melting, and, in general, have less interaction with other oceanic constituents than would similar contaminants in most other oceans.

Contaminants and introduced material which remain in or on the surface layers eventually get delivered to the North Atlantic Ocean, less reduced or altered by biological action or chemical interaction than would be the case after the same length of time in oceans at lower latitudes.

Material that reaches abyssal depth in the Arctic Ocean appears likely to stay there for very long periods of time due to the semi-closed nature of the basins.

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5. PRELIMINARY ASSESSMENT OF THE REGIONAL CAPACITY AND NEEDS

5.1. INTRODUCTION

The sources of most contaminants of interest to this assessment lie outside of the Arctic region.. PTSs released to the environment outside the Arctic are transported to the Arctic via air currents, rivers, and ocean currents. It follows therefore, that the management of anthropogenic PTSs lies outside of the region to a great extent.

The situation is somewhat complex from the perspective of both sources and management. Three types of sources can be identified. First, there are the local sources in the region. Second, there are the sources in the industrialized, non-Arctic portion of the circum-polar countries. These sources can be referred to as national sources. Finally, there are the international sources.

At the same time, the principal management and regulatory authorities for the Arctic region are located outside of the Artic region, in the national capitals to the south. This is reasonable, based on population alone, since the circumpolar countries have a total population of about 550 million of which only about four million live in the Arctic region. The net result is that, except for the management and regulation of local sources, the region must rely on the effectiveness of national and international programs and agreements to reduce current and future PTSs levels in the Arctic.

Iceland is the one exception to this description. Iceland lies entirely in the Arctic region. It is impacted by local and international sources only, and its principle management and regulatory authority lies within the region.

5.2. MONITORING CAPACITY

5.2.1. POPs

5.2.1.1. Regional

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 and chlordane-related compounds] and industrial products (PCBs and chlorobenzenes). DDT-related compounds are 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 yearround 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. Since, 1995, the Canadian site at Tagish has been closed, the Russian site has been relocated to a newly established monitoring site at Amderma in western Russia, and several sites have been added to the network, namely, at Pallas in northern Finland, on Bear Island (halfway between mainland Norway and Svalbard) and in northern Norway.

Information on levels of OCs in soils, plants, and terrestrial species in Arctic and sub-arctic 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, have been too limited to derive any conclusions about spatial or temporal trends of contaminants.

Information on contaminant loadings to the Arctic Ocean from northward-flowing rivers is limited. Knowledge of levels of persistent OCs in lake waters in the circumpolar Arctic is limited. However, recent studies of contaminants in water, sediments, and biota from Arctic lakes and rivers have greatly expanded the information available on OC levels.

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 decade has seen a large increase in the amount of data on persistent OCs available, especially from the European and Russian Arctic.

5.2.1.2. Local

In general, sources of POPs in the Arctic are not well documented, 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. 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. Many of these sources are known but have not been quantified through monitoring studies. The following presents a number of examples.

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.

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.

Several circumpolar countries produce hydroelectricity from rivers in the Arctic, including Norway, Sweden, Finland, Iceland, the USA (Alaska), and Canada. No studies of PCB levels near hydroelectric plants have been done, although transformers at the plants most probably contained PCBs at one time.

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 metals in the Arctic, the possible contamination by POPs from these sources has not been studied.

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.

5.2.2. Metals

Few spatial or temporal trends are apparent in the existing data, largely due to poor temporal or spatial coverage or to irresolvable artifacts in the data related to differences in sampling, analytical, and reporting protocols.

Progress has been made recently in addressing the poor coverage. Since the publication of the AAR in 1998, additional data on metals in air/aerosols and precipitation have been reported for the following monitoring stations: Alert (Canada), Station Nord (Greenland), Irafoss and Storhofdi (Iceland), Pesosjarvi, Vuoskojarvi and Pallas (Finland), Jergul, Øverbygd, Karpdalen and Svanvik (Norway) and Ny-Ålesund (Svalbard). The stations at Alert, Station Nord and Ny-Ålesund, together with Barrow (Alaska, operating from 1999) and Amderma (Russian European Arctic, operating from 2000) form the basis of a circumpolar background mercury-monitoring network. Plans are underway to add further stations with one proposed in the Russian Far East.

5.2.3. Human Health

There is very little information on temporal trends of POPs in Arctic populations. Monitoring of POPs in blood over the next decade is essential to establish whether risk management strategies for POPs are effective.

Concerning metals, existing data from the literature do not allow a valid estimate of spatial and temporal trends of exposure of Arctic peoples to mercury and cadmium, while for lead, a declining trend is observed. There is some evidence that the general decline in lead exposure parallels the decline in lead levels in industrialized areas.

Food is the major exposure route for contaminants in the Arctic. The type and amount of human exposure to environmental contaminants varies throughout the circumpolar region according to the level of contaminants in the food, the amount and type of food consumed, and the method of food preparation. For these reasons, wildlife-monitoring data provide a very uncertain basis for precise human exposure estimates. However, they are of great value for risk characterization as they identify the contaminants present in wildlife used as traditional food, and 124

the most contaminated species, and can, therefore, contribute to the basis for dietary recommendations. A uniform methodology for dietary intake studies applied across the circumpolar region would greatly assist risk assessment.

5.3. EXISTING REGULATION AND MANAGEMENT STRUCTURES

The AAR is the source document for the UNEP assessment of PTSs in the Arctic Region. The AAR did not examine the existing regulation and management structures. Hence, little information on regulations and infrastructure is included here. However, aside, from Iceland, the main discussions of these topics will be found in the reports for the bordering regions to the south, regions where the regulatory and management mandate resides, for the most part. The following is a belief summary of the limited information contained in the AAR.

5.3.1. National

There are thirteen POPs included in the AMAP study. Table 2 provides information on the legal status of each compound in each of the eight AMAP countries. No similar overview of the metals was available.

Table 2: Current legal status within the eight circumpolar countries of	of
persistent organic pollutants (POPs) covered in this report	

	U.S. (Alaska)	Canada	Russia	Iceland	Denmark	Norway	S
Aldrin	Banned 1987.	Voluntarily withdrawn 1990.	NI ¹	Never registered as a pesticide. Banned 1996.	Prohibited for plant protection use.	Not marketed since 1970.	B
Dieldrin	Banned 1971.	Restricted 1987.	NI	Never registered as a pesticide. Banned 1996.	Prohibited for plant protection use.	Not marketed since 1970.	B
Chlordanes	Restricted use 1988.	Registration discontinued 1990.	Use prohibited.	Banned 1996.	Prohibited for plant protection use.	Not marketed since 1967.	B
DDT	Banned 1972. Products with more than 0.1% banned 1986 (Dicofol).	Voluntarily withdrawn 1985.	,	Never registered as a pesticide. Banned 1996.		Restricted 1969. Banned 1988.	B D 19
НСВ	Registration cancelled 1984.	Registration not renewed 1976.	NI	Never registered as a pesticide. Banned 1996.	Banned.	Withdrawn 1987.	W 19
α-НСН	Mixed isomers cancelled 1977.	Mixed isomers discontinued 1976.	Technical product probably still in use.	Never registered as a pesticide.	Mixed isomers prohibited for plant protection use.		S
β-НСН	See α-HCH.	See α-HCH.	NI	See α-HCH.	See α-HCH.	NI	S
γ-HCH (lindane)	Cancelled for most uses 1983.	Restricted use.	Still in use.	Restricted use.	NI	Banned 1991.	В
Mirex	Use cancelled 1988.	Never registered as a pesticide.	NI	Never registered as a pesticide.	NI	Never used.	N
Toxaphene	Banned 1982, use ceased 1986.		Severely restricted 1992.	Never registered as a pesticide. Banned 1996.	Banned 1987.	Never used.	S. 19 us
Endosulfan	Still in use.	Restricted use for commercial pest control.		Restricted use in greenhouses.	NI	Still in use.	W 19

	U.S. (Alaska)	Canada	Russia	Iceland	Denmark	Norway	S
TBT	Restricted use 1988.	Restricted use - aluminum hulled boats in salt water.	NI	Banned on vessels <25 m, docks and fishing gear.	NI	Restricted use - aluminum hulled boats >25 m. All other use banned.	fr B or
РСВ	cancelled 1970;	Severely restricted 1985; open and new uses banned, closed use allowed.		>0.2% banned 1988. >0.005% banned 1996.		New use banned 1980. All use banned 1995.	b

 1 NI = no information.

5.3.2. Regional

The Nordic Council of Ministers has proposed guidelines for PTSs concentrations in food, particularly cadmium concentrations. For example, in 1992 the council proposed maximum Cd concentrations for the kidney, liver, and muscle tissue of meat (pigs and cattle) for human consumption of 1 μ g/g, 0.5 μ g/g, and 0.05 μ g/g (ww), respectively.

5.3.3. International

Pollution issues are covered by several international agreements or arrangements that form an important focus for political efforts aimed at reducing impacts on the Arctic environment and its ecosystems. The following have particular relevance to the UNEP assessment of the Arctic Region.

5.3.3.1. Long-range Transboundary Air Pollution (LRTAP)

The purpose of the UN Economic Commission for Europe's LRTAP Convention is to prevent, reduce and control trans-boundary air pollution both from existing and new sources. By covering not only the entire Arctic region, but also mid-latitude regions which are the origin of a major part of the pollution reaching the Arctic, this regional, binding agreement, and its related protocols, represents the most appropriate instrument for addressing significant components of the Arctic pollution problem.

5.3.3.2. <u>Convention for the Protection of the Marine Environment of the North East Atlantic, 1992</u> (OSPAR)

Although covering only a restricted segment of the circumpolar Arctic (between longitudes 44° W and 51° E), the 1992 OSPAR Convention is currently one of the most applicable international agreements addressing Arctic marine pollution from various sources. On both monitoring and source-related assessment issues, therefore, OSPAR 1992 represents a relevant agreement to be taken into account in the work of AMAP and UNEP.

5.3.3.3. International Convention for the Prevention of Pollution from Ships, 1973, as modified by the Protocol of 1978, (MARPOL 73/78)

The MARPOL Convention is a combination of two treaties adopted in 1973 and 1978. It covers all technical aspects of pollution from ships, except the disposal of waste into the sea by dumping, and applies to ships of all types. The Convention has five annexes covering oil, chemicals, sewage, garbage, and harmful substances carried in packages, portable tanks, freight containers, etc.

5.3.3.4. International Arctic Science Committee (IASC)

The non-governmental International Arctic Science Committee, founded in 1990, was established to encourage and facilitate cooperation in all aspects of Arctic research. IASC is a coordinating body for research in a number of fields of relevance to the work of AMAP. Examples of IASC programs which are followed with particular interest by AMAP include those concerned with 'Effects of Increased UV-B Radiation in the Arctic', and 'Mass Balance of Arctic Glaciers and Ice Sheets'.

5.3.3.5. Barents Euro-Arctic Region (BEAR)

Established in 1993 following the Kirkenes Declaration, the Barents Euro-Arctic Region aims to identify areas of cooperation between the Nordic countries, Russia and the European Union, and promote these through the Barents Council.

5.3.3.6. WMO Environmental Health Criteria

Over the past twenty years, the WMO has published an extensive list of environmental criteria for many of the PTSs discussed in this assessment. These criteria provide quantitative guidance for human concentrations including PTDI, TDI and TWI values.

5.3.3.7. Stockholm Convention On Persistent Organic Pollutants

This convention was adopted at the December 2000 meeting of the intergovernmental negotiating committee for an international legally binding instrument for implementing international action on certain persistent organic pollutants in Johannesburg. The objective of this Convention is to protect human health and the environment from persistent organic pollutants. The selected list of POPs is of direct relevance to the UNEP assessment of PTSs. The Convention was opened for ratification signatures on 23 May 2001.

5.4. STATUS OF ENFORCEMENT

The status of enforcement in non-Arctic countries that impact on the Arctic is discussed in other regional reports. This section considers Arctic countries only.

The Arctic countries should take all necessary steps to ensure that their domestic responsibilities and arrangements to reduce contaminant inputs to the Arctic region are fully implemented. If these responsibilities and arrangements are not addressed in an appropriate manner, the justification for recommending actions aimed at reducing transboundary contaminants with sources outside of the Arctic will be accordingly diminished. There is a need for actions to clean up contaminants from industrial and military sites. In addition, the treatment of municipal wastewater and the incineration of wastes require urgent attention.

Levels of many contaminants in the Arctic are likely to remain at or close to existing levels for decades because of their resistance to degradation, the slow rate of degradative processes, and the recycling of existing accumulations. Thus, ameliorative actions to reduce exposure to humans and to protect wildlife are an essential adjunct to emission controls.

5.5. ALTERNATIVES OR MEASURES FOR REDUCTION

Most reduction technologies are well known and widely practiced though their application to the Arctic may not always be straightforward or possible. Technologies and methodologies, which are practiced outside of the Arctic region, may require special adaptations to be effective in the distinctive Arctic circumstances.

One significant success has been the application of alternative processes in the pulp and paper production industry. Emissions of 2,3,7,8-TCDD/TCDF, particularly in Sweden and Canada, have been significantly reduced in recent years because of the substitution of molecular chlorine by other bleaching agents.

In the case of human health, the substitution of non-traditional foods is not considered an effective alternative for reducing the levels of some PTSs in humans. The reasons are wide ranging and compelling.

5.6. TECHNOLOGY TRANSFER

The Arctic region is located in a highly developed, technological part of the world and is politically part of industrialized countries that are responsible for the well being of the region. Therefore, technology transfer does not pose any intrinsic administrative difficulties. However, difficulties can arise when technologies designed for temperate zones are applied in the harsh Arctic environment.

5.7. IDENTIFICATION OF NEEDS

Throughout this report, reference is made to the need for more information on sources, pathways and trends. The conclusions and recommendations will not be repeated here. The following needs relate to information required to improve our understanding of the levels and effects of PTSs on the inhabitants of the Arctic.

Despite the number of controls on several POPs imposed during the 1970s and 1980s, there is no evidence that levels in Arctic peoples have decreased. The persistence of POPs, their presence throughout the ecosystem, and the continued use of some POPs for disease vector control, all contribute to the constant influx of POPs into the arctic environment and sustained levels of human exposure. Monitoring of POPs in blood over the next decade is essential to establish whether risk management strategies for POPs are effective.

Elevated levels of toxaphene and chlordane, coupled with current intake scenarios, suggest some indigenous groups are exposed to levels of these contaminants significantly above the Tolerable Daily Intake (TDI). Information on the levels of toxaphene in human tissues is limited.

There is insufficient information to conclude whether the TDI for dioxins and furans and dioxin-like PCBs is being exceeded in Arctic populations. In addition, there is as yet little conclusive scientific information directly linking harmful human effects to low levels of exposure to these contaminants.

Existing epidemiological evidence on the adverse effects of POPs in humans is inconclusive and needs to be replicated because of the specific context in the Arctic in which there are differences in genetics, climate, food consumption patterns, and lifestyle among population groups.

The high exposure of indigenous peoples to methylmercury in some Arctic areas is a matter of concern because of its neurotoxic effects on the fetus. Further investigation of both the levels and the influence of mercury on fetal development, is warranted.

Lead levels in Arctic indigenous peoples have declined since the implementation of controls on lead emissions. Concentrations of lead in blood currently reported are below a level of concern, however, continued monitoring is warranted because of the potent effects of lead on neurological development in the fetus and children.

Determining adverse health effects in human populations due to the presence of contaminants in traditional foods and human milk is extremely difficult for methodological and ethical reasons. Results are also difficult to interpret because of a wide range of confounding factors (socio-economic, lifestyle and gender/age related).

Monitoring contaminant concentrations in human tissues and using these data to estimate exposures will continue to be necessary as will a general reliance on animal studies of the effects of mixtures of contaminants and nutrients found in Arctic foods. Regional tissue banks would greatly assist the requirements for retrospective comparative studies of contaminant levels and effects.

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6. FINAL RESULTS AND RECOMMENDATIONS

At the end of Chapters 2, 3, 4 and 5 there are subsections listing conclusions, recommendations and data gaps. The following two sections, 'Main Results' and 'Recommendations for Future Activities', present the key findings.

The 'Main Results' have been selected from the previous chapters. The selection is subjective; hence, the interested reader is encouraged to refer to the full record in each chapter. The 'Recommendations for Future Activities' are general statements, which are not found in the text above. In order to present a quantitative prioritisation of the chemicals selected, a scoring exercise was undertaken. The results of this scoring exercise are shown in **Annex I**. It is worthy to note that the results are presented in terms of Sources, Environmental Levels, Ecotoxicological and Human Effects and by Data Gaps. No effort is made to prioritise on a complete chemical basis.

6.1. MAIN RESULTS

6.1.1. Sources

Over much of the Arctic, the levels of POPs cannot be related to known use and/or releases from potential sources within the Arctic and can only be explained by long-range transport from lower latitudes. Of the heavy metal contamination in the Arctic, industrial sources in Europe and North America account for up to one-third of the deposition, with maximum input in winter.

The Barents region of the Russian Arctic is the most highly populated and industrially developed part of the circum-polar Arctic. In such urbanized areas, surface runoff waters, including organized industrial and communal wastewater discharges, can contribute up to 40-50% of the total pollution entering the water bodies (WHO/UNEP).

Military and civilian sites and dumpsites that contain significant amounts of electrical equipment may be sources of PCB-contamination. Local PCDD/F contamination in the vicinity of a smelter has also been demonstrated. Surveys of all such sites within the Arctic should be made to determine the circumpolar scope of the problem.

Numerous local sources of other POPs also exist, but have not been studied. Surveys of local sources of contamination by POPs within the Arctic are needed to quantify the emissions and leakage. For example, in some locations in Russia, there are high HCH levels in lake water and high PCB and DDT levels are now seen in snow, rivers, seawater, coastal sediments, and the few samples of invertebrates, fish, reindeer, lemming, seabirds, seal, and beluga. These findings indicate possible fresh releases or improper disposal. These high levels must be verified.

6.1.2. Levels and Effects

Despite the controls on several POPs imposed during the 1970s and 1980s, there is no evidence that levels in Arctic peoples have decreased. The persistence of POPs and the use of some POPs for disease vector control, contribute to the constant influx of POPs into the arctic environment and sustained levels of human exposure.

Elevated ambient levels of toxaphene and chlordane, coupled with current intake scenarios, suggest some indigenous groups are exposed to levels of these contaminants significantly above the Tolerable Daily Intake (TDI). Information on the levels of toxaphene in human tissues is limited.

Existing data from the literature do not allow a valid estimate of spatial and temporal trends of exposure of Arctic peoples to mercury.

Lead levels in Arctic indigenous peoples have declined since the implementation of controls on lead emissions. Concentrations of lead in blood currently reported are below a level of concern, however, continued monitoring is warranted because of the potent effects of lead on neurological development in the fetus and children.

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 ambient 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.

Concentrations of PCBs in lake waters in Canada and Russia exceed levels associated with negative biological effects. Higher levels of HCHs, DDT, and PCBs are found in the Russian lakes.

Exceptionally high HCH levels are found in Russian river water, especially the Ob. Ratios of γ -HCH to α -HCH indicate use of lindane.

Availability of information on contaminant levels in the tissues of northern residents is very recent. It is therefore important to recognize that the paucity of health data or the absence of overt illness or malfunction does not imply that the exposure of peoples in the Arctic to contaminants is without effects. Studies to date have shown:

The influence of contaminants on fetal and neonatal development is of special concern. Preliminary results indicate that POP and methylmercury concentrations are two to ten-fold higher in breast milk and cord blood in some Arctic areas than in breast milk and cord blood from regions south of the Arctic.

There is both scientific and public concern about the possible adverse effects of POPs on pregnancy outcome, fetal development, child development, reproduction, male and female fertility, and the immune system. Several of these effects may be mediated through endocrine disrupting properties of some POPs. DDT and its metabolites and some dioxin and PCB congeners have been implicated.

The high exposure of indigenous peoples to methylmercury in some Arctic areas is a matter of concern because of its neurotoxic effects on the fetus.

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.

Complete circumpolar coverage of contaminants in biota is lacking, particularly for Alaska and the Russian Arctic. This lack of circumpolar data limits our ability to identify trends and to understand sources, transport pathways, and mechanisms for focusing contaminants.

Overall, the information on temporal trends in the Arctic is very limited. The results available reinforce the importance of judicious sampling and archiving programs, which would allow continuous long-term monitoring of key populations and retrospective analysis for new contaminants. Nevertheless, studies to date have shown:

A nine-fold decline in concentrations of HCH 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 be due to regional differences in inputs of HCH isomers.

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. However, the interpretation of profiles of POPs in snow and sediment cores can be problematic where melting or mixing has occurred.

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.

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.

The concentration of most heavy metals measured in sub-arctic air has decreased during the last two decades. All the heavy metals show strong seasonal variation in the High Arctic.

The highest concentrations of atmospheric heavy metals in Arctic air occur near smelter complexes on the Kola Peninsula and at Norilsk and result from emissions from these smelters.

Health effects associated with metals have so far not been investigated in Arctic biota.

6.1.3. Pathways

The Arctic is a focus for major atmospheric, riverine, and marine pathways, which result in the long-range transport of contaminants into and within the Arctic. The Arctic is, therefore, a potential contaminant storage reservoir and/or sink.

The relative importance of atmospheric transport compared to marine and terrestrial/ freshwater is very contaminant specific for the multi-hop compounds, such as OC pesticides, Hg and PCBs. While all of the compartments play a role in transporting these contaminants, the speed of transfer through the atmosphere suggests that this compartment is particularly important in the global cycling of these types of compounds.

Given the current configurations of anthropogenic sources at mid-latitudes, the most favored pathway into the Arctic is from the Eurasian continent from November to May.

6.1.4. Regional Capacity

The Arctic countries should take all necessary steps to ensure that their domestic responsibilities and arrangements to reduce contaminant inputs to the Arctic region are fully implemented. If these responsibilities and arrangements are not addressed in an appropriate manner, the justification for recommending actions aimed at reducing transboundary contaminants with sources outside of the Arctic will be accordingly diminished.

There is a need for actions to clean up contaminants from local industrial and military sites. In addition, the treatment of municipal wastewater and the incineration of wastes require urgent attention.

6.2. Recommendations For Future Activities

The following recommendations are intended to provide broad guidance for future studies concerned with the PTSs 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.

Monitoring of POPs in blood over the next decade is essential to establish whether or not risk management strategies for POPs are effective.

Determining adverse health effects in human populations due to the presence of contaminants in traditional foods and human milk is extremely difficult for methodological and ethical reasons. Results are also difficult to interpret because of a wide range of confounding factors (socio-economic, lifestyle and gender/age related). Monitoring contaminant concentrations in human tissues and using these data to estimate exposures will continue to be necessary as will a general reliance on animal studies of the effects of mixtures of contaminants and nutrients found in Arctic foods. Tissue banks would greatly assist the requirements for retrospective comparative studies of contaminant levels and effects.

Contamination of the Arctic is part of a global process. While human exposures in the Arctic can be moderately reduced with some dietary modifications (provided these are culturally, socially and nutritionally suitable for the specific communities involved), it must be recognized that long-term exposure reductions can only be accomplished through international conventions resulting in bans and restrictions on production and use of the most toxic chemicals.

There is ample evidence of the limited effectiveness of sporadic and poorly integrated process, pathway, and flux studies. Using multi-media models, a great deal could be achieved efficiently by fully integrating processes, observed levels, and trends. We would obtain assessment and feedback on the design and implementation of future monitoring and process research activities, as well as management and/or mitigation measures.

Models are essential in assessing the sensitivity of individual or linked processes, which in turn can be instructive in assigning priorities to often-complex research questions. In particular, models have not been used to their fullest in the area of terrestrial/freshwater pathways and other complex questions, including evaluation of the relative importance of processes, estimation of transport fluxes, and assessment of remedial measures.

The AAR includes a chapter on climate change. The discussion does not address the profound linkage between a warming climate and the occurrence and consequences of PTSs in the Arctic region. There is growing evidence that the climate in the Arctic is changing, in some places more rapidly than in any other place on Earth. A thorough assessment of this cross-issue linkage is urgently required. The implications of warmer temperatures on PTS transport into and in the region, on cold condensation, on revolatilization and on the melting of snow and ice with their associated burdens of PTSs, are just a few of the issues that need attention.

Instructions:

- 1) Chemicals to be grouped by matrix (sources, environmental conc., etc.) and by score. <u>There is no total</u> <u>score for any chemical.</u>
- 2) An associated column for data gaps is attached to each matrix. For example, Sources has an accompanying column to score the degree of data gaps experienced for Sources.
- 3) A short summary with representative, specific data must be used to justify the score given. Use <50 words.
- 4) All chemicals selected for the study must have a score for each category
- 5) The guidelines attached provide a qualitative measure for scores. Scores are measured as follows:

Scores: Score = 0 – chemical is of no concern or knowledge base limited/ supportive data is collected

Score = 1 – chemical has local concern/ supportive data is limited

Score = 2 – chemical has regional concern/ supportive data is lacking

NB. The score given for a matrix on a chemical does not have to be the same score for data gaps on that matrix

6) Chemicals should be tabled according to their placement by score. For example, a table should be presented for chemicals under Sources of Regional concern (score 2) with the accompanying data gap score for Sources. An example of a chemical placed in the Score 2 table for sources for a particular region is given below:

7. SOURCES: REGIONAL CONCERN

7.1.	CHEMICAL	7.2. DATA GAPS	7.3. COMMENTS
DDT		1	DDT used historically for > 30 years. Approximately 800,00kg now produced annually. 240,000kg used annually in the region (6 countries) for malaria control (75%) and 80,000kg used in agriculture (8 countries).

Guidelines for Scoring Issues Associated with Each Chemical

	Score 0 =	Score 1 =	Score 2 =
MATRIX	No concern	Local concern	Regional concern
Sources of the Chemical	 No evidence of production or product contamination No evidence of air emissions No evidence of emissions from solid residues No evidence of chemical stockpiled No evidence of chemical being contaminant in production of other chemicals No evidence of use of the chemical No evidence of release from liquid effluent 	 Evidence of limited production Presence of small sources with possible emissions (e.g.,small incineration plants or bleached kraft/pulp mills using chlorine); Some limited evidence of releases but on a small scale invoking local concerns Some use of the chemical locally Over time, levels remain below threshold or are decreasing Use of chemical in agriculture or industry sub- regionally Evidence of limited stockpiles of the chemical Increasing spatial and/or temporal trends from levels below threshold and localised 	 Major production of chemical for local and export use. Chemical evident as contaminant in large scale production of other chemicals Known emission of chemical from large scale incinerators or chlorine bleaching of pulp or other related combustion facilities Evidence of leakage from major stockpiles of the chemical poorly packaged Large-scale use of the chemical throughout the region Spatial and/or temporal trends increasing regionally from levels above threshold
Environmental Levels of the chemical	 No known or historical levels of chemical contaminant in the environment except background levels of naturally occurring substances No available data to quantify evidence of the chemical found in fish, wildlife animal or human tissue 	 Chemical contaminants are detectable in the environment but below threshold limits defined for the country or sub-region Chemical contaminants are detectable from fish, wildlife, foodstuff or human samples but below threshold limits established for the country or sub-region Over time, levels remain below threshold or are decreasing Increasing spatial and/or temporal trends from levels below threshold 	 Chemical contaminant is analysed repeatedly well above threshold limits in the environment defined for a country or region Known contamination of fish, wildlife, foodstuff or humans at levels far exceeding the threshold established for the country or region Spatial and/or temporal trends increasing from levels above threshold

Ecotoxicological Effects from exposure to the chemical	 No fisheries closures or advisories due to chemical pollution No incidence of food/fisheries product tainting No unusual fish or wildlife mortality events 	 Inconclusive evidence of limited fish or wildlife mortality events on a local or sub-regional scale Temporal trend shows constant or decreasing effect of chemical 	 Public health and public awareness of food/fisheries contamination problems with associated reduction in the marketability of such products either through the imposition of limited advisories or by area closures Large-scale mortalities of aquatic or wildlife species Temporal trend showing increase in effects of chemical regionally
Human Effects from exposure to the chemical	 No indication of any ill effects from exposure to the chemical No correlation between human diseases and chemical exposure 	 Odd incidence of ill effects that may be related to exposure to the chemical Evidence of localised effects from spot exposure to the chemical Temporal trend shows constant or decreasing effect of chemical locally 	 Indications of health effects resulting from use of pesticide/industry chemical Wide spread health effects from involuntary exposure to the chemical Temporal trend showing increase in effects of chemical regionally
Data Gaps	 Full data sets established Evidence complete Ongoing monitoring data available 	 Limited data available Minimum data required to confirm findings Data available conflicting Further monitoring data required on a wider scale Limited anecdotal evidence of local human and environmental effects 	 Sparse data available Data unreliable Data unreliable No data available Only historical data (>20 years) available Limited data available shows major concern Widespread anecdotal evidence of human and environmental effects

RESULTS OF SCORING OF PTS FOR THE ARCTIC REGION

SOURCES					
Chemical	Score	Data Gaps	Comments		
PCBs	2	1	Legal status in 8 Arctic countries as of mid-1990s: see table 1. Cumulative global PCB production is estimated at 1.3 million tonnes. Like most POPs, PCBs have primarily contaminated the Arctic ecosystem through long-range transport from more southern regions. Open use is currently banned in all circumpolar countries, but there are still large amounts present in the Arctic in permitted use (in older electrical equipment - transformers, capacitors, etc.), for example 460 tonnes at Norilsk. At present, the major source of PCB exposure seems to be environmental recycling of PCBs from former usage. Current sources are therefore: leakage from historical open use (e.g. sealants and other building materials); leakage from a number of contaminated industrial and waste sites throughout the Arctic (e.g. landfills); PCB contamination from military sites in the Arctic (e.g. DEW line), solid waste disposal sites at mines, older oil installations, and possibly ships/submarines (incl. dumped vessels). PCB-contaminated waste oil was applied to soils to control dust in Arctic settlements (e.g. Fairbanks). An inventory PCB production in the fSU and amounts of PCB-contaminated wastes, etc., in the Russian Federation (including Arctic areas) has been completed.		
Chlordane	1	1	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. Total global usage estimated at 78 Ktonnes (Barrie et al., 1992). It is not registered for use in circumpolar jurisdictions and enters the Arctic ecosystem primarily via long-range transport. Patterns of chlordane and heptachlor epoxide in Arctic air sampled during the mid-1990s suggest releases from soils (not necessarily within the Arctic) to which pesticides were previously applied.		
DDT	1	1	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. DDT was commonly used as a pesticide on a variety of agricultural crops and is still in use for the control of mosquitoes that spread malaria (vector control). Reported global usage (1948-93) 1500 Ktonnes; interpolated global usage (1950-93) 2600 Ktonnes. Anecdotal information that household and institutional insecticides containing DDT are still in use in Russia supported by DDE:DDT ratios in monitoring data. Historical use as insecticide (mosquitos and black fly) in Canada, US, and Russia and probably all Arctic countries (possibly still continuing in Russia).		
Dioxins	1	1	Pulp and paper industry emissions of 2,3,7,8-TCDD in circumpolar countries reduced during 1990s. Uncontrolled local incineration of waste in Arctic may be a source of dioxins. PCDD contamination from smelters and metallurgical plants (Sweden, Norway, Russia - Kola and Norilsk). Low-temperature combustion sources of PAHs exist throughout the circumpolar Arctic.		
Furans	1	1	Pulp and paper industry emissions of 2,3,7,8-TCDF in circumpolar countries reduced during 1990s. Uncontrolled local incineration of waste in Arctic may be a source of furans. PCDF contamination from smelters and metallurgical plants (Sweden, Norway, Russia - Kola and Norilsk)		
НСВ	1	1	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. The estimated current global annual emission is 23 tonnes. Uncontrolled local incineration of waste in Arctic may be a source of HCB.		
Heptachlor	1	1	See chlordanes. Heptachlor is now banned in several countries and severely restricted in others. Heptachlor is metabolized in animals to heptachlorepoxide.		
Org Lead Compds.	1	2	Organolead compounds are used in leaded gasoline which is available in parts of the Arctic and still widely used in Russia.		
Org. Mercury Compds.	1	2	No information available on anthropogenic sources of organic mercury compounds in the Arctic - metallic mercury is however methylated in sediments by microorganisms.		

SOURCES cont'd.					
Chemical	Score	Data Gaps	Comments		
НСН	1	1	Legal status in 8 Arctic countries as of mid-1990s: see table 1 (alpha-, beta-, gamma- HCH). Many countries still use large amounts of lindane and there are some registered uses of lindane in some circumpolar countries. Total world production is estimated at 0.7 million tonnes of lindane and 10 million tonnes of technical HCH. Reported global usage (1948-93) 550 Ktonnes technical HCH, 720 Ktonnes technical lindane. Household and institutional insecticides containing HCH still in use. An HCH mass balance budget for the Arctic Ocean has been developed, and there is some evidence that the Arctic Ocean (Beaufort Sea) is now be a 'secondary source' of alpha-HCH as air concentrations from primary emissions decrease.		
Org. Tin Compds.	1	1	Legal status in 8 Arctic countries as of mid-1990s: see table 1 (TBT). Main contamination in Arctic from use of TBT as marine antifoulant on ships and offshore structures; use on small vessels controlled, other main uses subject to global ban (in 2003).		
PAHs	1	1	Anthropogenic sources of PAHs in the Arctic include uncontrolled local incineration of waste, and burning of fossil fuels, in particular wood-burning for residential heating is common in some parts of the Arctic. PAH emissions from oil/gas development in the Arctic. Low-temperature combustion sources of PAHs exist throughout the circumpolar Arctic.		
PBDE	1	1	Releases from PBDE-containing equipment and PBDE-containing wastes with minor use in the Arctic. Increasing trends in emissions that ultimately may contaminate the Arctic. Penta-BDE under consideration for withdrawal in Europe.		
Toxaphene	1	1	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. Toxaphene is one of the most heavily used pesticides, with a total world production of 1.3 million tonnes up until 1993. Reported global usage (1948-93) 450 Ktonnes; interpolated global usage (1950-93) 1330 Ktonnes. Toxaphene and comparable mixtures are now banned in most countries. Similar products have been, and may continue to be used in fSU.		
Aldrin	0	2	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. Total global production estimated at 500 Ktonnes. Aldrin is readily metabolized in plants and animals to dieldrin.		
Atrazine	0	2	Mid-1990s status: banned in Norway and Sweden , heavily used in USA (29 Ktonnes in 1988) and Canada		
Chlordecone	0	2			
Chlorinated Paraffins	0	2	Information on historical production volumes of polychlorinated-n-alkanes is difficult to find but annual global production during the 1990s was estimated at 300 Ktonnes, of which 50 Ktonnes were SCCPs. Due to concerns about their possible environmental impact the use of SCCPs is declining.		
Dieldrin	0	2	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. Worldwide total production estimated at 34 Ktonnes. Today, primary uses include controlling termites, woodborers, and textile pests. Action has been taken to ban or restrict the use of dieldrin, one of the most common pesticides.		
Endosulphan	0	1	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. Estimated global use of 57 Ktonnes since mid-1950s		
Endrin	0	2	Endrin has been used as an insecticide on cotton and grain, and as a rodenticide to control mice and voles. Like aldrin, endrin have been banned or severely restricted in most countries.		

	SOURCES cont'd.					
Chemical	Score	Data Gaps	Comments			
Mirex	0	2	Legal status of use and production in the 8 Arctic countries (in mid-1990s): see table 1. The compound has never been used in the Arctic and is now banned in many countries. Mirex is used as an insecticide in Canada and US. It has been manufactured and applied extensively in the United States and has reached the Arctic via long-range atmospheric transport.			
Nonylphenols	0	2				
Octylphenols	0	2				
PCNs	0	2	In the 1920s, worldwide annual production was approximately 9000 tonnes. One of the largest PCN producers voluntarily ceased production in the late 1970s. However, general information about world production volumes and history is limited. Although the use of PCNs has declined in the past few decades, they are not prohibited in most countries and still occur in many PCB-like applications.			
РСР	0	2	Mid-1990s status: banned in Canada and Scandinavia, registered only for restricted use in USA and western Europe. Worldwide production estimated at less than 30 Ktonnes per year. PCP and its salts have been used as algicides, fungicides, insecticides, molluscicides, and herbicides. Current use is limited to the treatment of lumber, in particular utility poles. Because of its volatility and water solubility, PCP evaporates or is leached from wood structures to a large extent.			
PFOS	0	2	PFOS has been manufactured for over 50 years and its many useful properties have steadily increased its use, making PFOS a high volume product with an estimated U.S. production of 3000 tonnes in 2000. The 3M Company plans to phase out production in 2003 thus U.S. production is anticipated to decline. Elsewhere production is not well documented.			
Phthalates	0	2	Still extensively used in plastics			

	ENVIRONMENTAL LEVELS					
Chemical	Score	Data Gaps	Comments			
DDT	2	0	DDT is detectable in almost all compartments of the Arctic ecosystem. Organochlorine pesticides in the Arctic air reflect current and past use in mid- latitudes of the northern hemisphere and, for some OCs, global transport from use areas at low latitudes. Levels in fish and wildlife have decreased during 1980s and 90s following bans. Air and precipitation data suggest fresh releases (use or leakage from stored stockpiles/wastes) in the Barents region. High DDT levels found in suspended sediments of the Ob and Yenisey rivers. Elevated DDT concentrations in predators, in particular marine mammals such as polar bear and toothed whales. High DDT in burbot from some Arctic lakes is associated with historical use.			
Org. Tin Compds.	2	1	Because of its low water solubility and lipophilic character, TBT is readily adsorbed onto particles and may persist in sediments for several years. It bioaccumulates in organisms, with the highest concentrations found in liver and kidney. Uptake from food is more important than uptake directly from water. However, TBT is rapidly metabolized and excreted from aquatic and terrestrial mammals and has not been shown to biomagnify in higher organisms. TBT has been detected in marine sediments in harbors throughout the world, including the Arctic region			
Aldrin	1	2	Detected in Arctic environments.			
Atrazine	1	1	Detected in Arctic environments in low levels			
Dioxins	1	1	Local contamination around smelters. Elevated dioxin levels in reindeer and freshwater fish on the Kola peninsula may be related to local contamination from smelters.			

	ENVIRONMENTAL LEVELS cont'd.					
Chemical	Score	Data Gaps	Comments			
Chlordane	1	1	Chlordanes are among the most abundant environmental pollutants. CHBs, the major constituent of toxaphene, are semi-volatile and enter the Arctic region via long-range atmospheric transport. Organochlorine pesticides in the Arctic air reflect current and past use in mid-latitudes of the northern hemisphere and, for some OCs, global transport from use areas at low latitudes. The compounds are persistent (half-life in soil up to 12 years). Chlordane concentrations more pronounced in predators.			
Chlorinated Paraffins	1	1	Detected in Arctic environments. Chlorinated paraffins detected in biota on Svalbard. SCCPs are relatively mobile, subject to long-range transport, and are found in the Arctic. Studies indicate that SCCPs are less persistent and bioaccumulative than PCBs hence levels in the environment are low compared to PCBs. Medium- and long chained chlorinated paraffins are not yet reported in the Arctic.			
Dieldrin	1	1	Detected in Arctic environments.			
Endosulphan	1	1	Detected in Arctic environments. Endosulfan is moderately persistent in the environment. In soil, the half-life ranges from 30 to 200 days, and is affected by pH, the presence of microorganisms, and humidity. In water it is hydrolyzed slowly to form sulfur dioxide and endosulfandiol. In mammals, endosulfan is absorbed rapidly though incompletely by the digestive system. Endosulfan is lipophilic but is rapidly converted to a number of hydrophilic metabolites, including endosulfan sulfate, endosulfandiol, endosulfan ether, and hydroxy endosulfan carboxylic acid, which are more easily excreted. Thus it does not accumulate in the fat, and hence shows little potential for bioaccumulation or biomagnification			
Endrin	1	2	Endrin is rapidly metabolized by animals and does not accumulate in fatty tissues to the same extent as aldrin and dieldrin.			
Furans	1	1	Local contamination around smelters. Elevated furan levels in reindeer and freshwater fish on the Kola peninsula may be related to local contamination from smelters.			
НСВ	1	1	One of the most prevalent POPs in Arctic soils, plants and herbivores.			
НСН	1	0	HCHs partition into all environmental media in the Arctic. Lindane in Arctic air is correlated with long-range transport. Highest levels of alpha-HCH in the world's oceans occur in the Arctic Ocean (Canada Basin, Arctic archipelago), explained by proximity to source regions in Asia. Organochlorine pesticides in the Arctic air reflect current and past use in mid-latitudes of the northern hemisphere and, for some OCs, global transport from use areas at low latitudes. High levels of HCH found in water from some Russian rivers with gamma-/alpha-HCH ratios indicative of use of lindane in the catchments. HCHs are prevalent POPs in Arctic soils, plants and herbivores. Biodegradation seems to be the dominant decomposition pathway in soil and water.			
Heptachlor	1	2	Heptachlorepoxide found in Arctic abiotic and biotic environments.			
Mirex	1	1	Mirex is one of the most stable and persistent pesticides with an environmental half- life of up to 10 years. Organochlorine pesticides in the Arctic air reflect current and past use in mid-latitudes of the northern hemisphere and, for some OCs, global transport from use areas at low latitudes. Mirex is readily absorbed and stored in fatty tissues.			
Org. Mercury Compds.	1	1	Methylmercury is readily taken up by aquatic organisms and biomagnified through the food chain. Methylmercury exposure has been extensively documented in Arctic and subarctic populations since the 1970s.			
PAHs	1	0	Levels in air indicate long-range transport from Eurasian sources, with a signature typical of coal combustion. Local contamination associated with oil/gas development.			

	ENVIRONMENTAL LEVELS cont'd.					
Chemical	Score	Data Gaps	Comments			
PBDE	1	1	Detected in Arctic environments. The physical properties of brominated flame retardants (in particular penta-BDEs) encourage long-range transport and accumulation. The fact that they are found in air and biological samples from remote areas of the Arctic supports the indications that these substances are globally distributed in the environment. Although decabromodiphenyl ether (BDE 209) accounts for most PBDE use, the most commonly found congeners are the tetra and penta derivatives. Increasing trends in beluga and seals reflect trends in global emissions. PBDEs detected in biota on Svalbard.			
PCNs	1	1	Detected in Arctic air. PCNs detected in biota on Svalbard.			
РСР	1	2	Detected in Arctic environments as anisole (PCA). Environmental contamination is widespread, PeCP being found in ambient air, surface and ground waters, sediments, soil, and aquatic and terrestrial organisms. PeCP is degraded by light and microorganisms to form numerous transformation products, of which pentachloroanisole and tetrachlorocatechol are the most abundant in the environment. PeCP bioaccumulates in fish but in mammalian systems it is rapidly excreted in the urine, either free or conjugated with glucuronide.			
PFOS	1	1	PFOS has been detected at elevated concentrations in some Arctic animals (esp. marine mammals)			
Phthalates	1	2				
Toxaphene	1	1	Organochlorine pesticides in the Arctic air reflect current and past use in mid-latitudes of the northern hemisphere and, for some OCs, global transport from use areas at low latitudes. Evidence of fresh inputs of toxaphene in the White Sea region. Elevated toxaphene levels found in Arctic freshwater fish feeding at higher trophic levels.			
Chlordecone	0	2				
Nonylphenols	0	2				
Octylphenols	0	2				
Org Lead Compds.	0	2				

	ECOTOXICOLOGICAL EFFECTS					
Chemicals	Score	Data Gaps	Comments			
DDT	2	1	Effects: see table 2. Implicated in reduced reproduction or egg-shell thinning in some populations of terrestrial predatory birds and sea-eagles. Possibly implicated in population declines in Stellar sea lions. DDT levels in some Arctic freshwater and marine fish, seabird eggs and ringed seal exceed guidelines for protecting fish-eating/aquatic wildlife. DDT levels raise concern for population level effects in some Arctic wildlife populations.			
Org. Tin Compds.	2	1	Effects: see table 2. TBT is an endocrine disruptor for certain species of snail and other types of mollusk, but has not been shown to similarly affect higher organisms. Evidence of reproductive effects of TBT on mollusks (imposex) in or close to harbours in investigated Arctic areas (W. Greenland, Iceland, Faeroe Islands, north Norway, Svalbard). TBT levels raise concern for population level effects in some Arctic wildlife populations.			
Chlordane	1	2	Effects: see table 2. Levels associated with reduced reproduction or egg-shell thinning in some populations of terrestrial predatory birds. Chlordanes in Canadian Peregrine falcon higher in 1990s than 1980s.			

ECOTOXICOLOGICAL EFFECTS cont'd.					
Chemical	Score	Data Gaps	Comments		
Dieldrin	1	2	Effects: see table 2. Levels associated with reduced reproduction in some populations of terrestrial predatory birds.		
Dioxins	1	1	Effects: see table 2. PCDD TEQs in some Arctic sea-eagles exceed thresholds for reproductive effects. Levels of dioxin-like substances raise concern for population level effects in some Arctic wildlife populations.		
Furans	1	1	Effects: see table 2. PCDF TEQs in some Arctic sea-eagles exceed thresholds for reproductive effects		
НСВ	1	2	Effects: see table 2. Levels associated with egg-shell thinning in populations of Fennoscandian white-tailed sea-eagles		
Org. Mercury Compds.	1	1	Methyl mercury is highly toxic.		
Toxaphene	1	2	Effects: see table 2. Some Arctic fish populations have toxaphene levels high enough to effect bone development in fry and increased mortality at spawning.		
Aldrin	0	2	Effects: see table 2		
Atrazine	0	2			
Chlordecone	0	2			
Chlorinated Paraffins	0	2	Effects: see table 2. SCCPs appear to elicit fewer acute and chronic toxic effects when compared to PCBs, but information is very limited. Risk assessments of SCCPs are inconsistent with respect to their conclusions regarding ecological risk, including carcinogenicity, clearly indicating the need for more data		
Endosulphan	0	2	Effects: see table 2		
Endrin	0	2			
НСН	0	2	Effects: see table 2		
Heptachlor	0	2	Carcinogenic		
Mirex	0	2	Effects: see table 2		
Nonylphenols	0	2			
Octylphenols	0	2			
Org Lead Compds.	0	2			
PAHs	0	1	Usually PAHs with two to six fused aromatic rings are those of toxicological interest.		
PBDE	0	2	Effects: see table 2		
PCNs	0	2	Effects: see table 2. Some of the PCN congeners appear to exhibit dioxin-like toxicity of a similar magnitude to some of the co-planar PCBs		
РСР	0	2			
PFOS	0	2	Effects: see table 2. PFOS has been shown to affect cell-cell communication, membrane transport, energy generation, and proxisome proliferation, but concentrations in wildlife are believed to be lower than those required to cause adverse effects.		
Phthalates	0	2			

	HUMAN EFFECTS					
Chemical	Score	Data Gaps	Comments			
Org. Mercury Compds.	2	1	Methylmercury levels are correlated with the consumption of fish or sea mammals. In non-consumers, blood values are generally $< 2 \ \mu g/L$. In people who consume large amounts of fish, however, levels as high as 800 $\mu g/L$ have been observed, with means of 20–40 $\mu g/L$. These levels can be compared to the 'no risk' level of 20 $\mu g/L$. Blood plasma levels in some Arctic population groups exceed Canadian and US established public health guidelines. Epidemiological studies in the Faeroe Islands have implicated (methyl)mercury exposure in neurodevelopmental effects in children, and possibly combined effects with PCBs. Similar or higher methylmercury exposures to those seen in the Faeroe Islands are found in other Arctic populations and WHO established tolerable weekly intake values for methylmercury are exceeded by some Inuit grops in Greenland and Canada. Methylmercury affects the nervous system and can cause paresthesia, ataxia, and tunnel vision.			
PCBs	2	1	Main source of human exposure is through diet, especially to population groups exploiting traditional foods and that have a relatively high consumption of marine mammals (e.g. Inuit in Greenland and Canada). Tolerable daily intakes exceeded by some Arctic population groups. Blood plasma levels in some Arctic population groups exceed national public health guidelines for 'levels of concern' (>5 μ g/L and < 100 μ g/L) and 'action levels' (>100 μ g/L). Epidemiological studies in the Faeroe Islands have implicated PCB exposure in neurodevelopmental effects in children, and possibly combined effects with mercury. Similar or higher PCB exposures to those seen in the Faeroe Islands are found in other Arctic populations. Typical sum-PCB values in plasma are in the range 1–60 μ g/L. The most abundant hydroxy-PCBs in human blood samples equals the presence of some abundant PCB congeners with sum values in the range 0.1–10 μ g/L. Several possible adverse human health effects to PCB exposure have been suggested, including effects on the immune system, cancer, effects on the reproductive system, and cognitive development . Some of their toxicity may be linked to the biotransformation products. Due to structural similarities (with hydroxy group in para- or meta-position), the hydroxy metabolites are assumed to be able to mimic hormonal activities. The non-ortho and mono-ortho substituted PCBs are planar with toxic properties similar to the dioxins.			
Chlordane	1	1	In general, only small amounts of chlordane are found in human tissues. However, relatively larger amounts of trans-nonachlor and the metabolites oxychlordane and heptachlor epoxide are found. Typical values (sum chlordanes) in human plasma are in the range $0.2-2 \mu g/L$. The compound class is a possible carcinogen and is believed to affect the immune system. Tolerable daily intakes exceeded by some Arctic population groups.			
Dieldrin	1	1	Main source of human exposure is through food. Adverse effects include toxicity, immune system depression, and carcinogenic action. Tolerable daily intakes exceeded by some Arctic population groups.			
НСВ	1	1	Dietary intake is the major route of human exposure, with highest concentrations found in oils and fats, meat, poultry, and fish. Tolerable daily intakes exceeded by some Arctic population groups. Exposure via inhalation or through drinking water is considered to be low. Typical plasma levels in Arctic populations are in the range $0.3-2 \mu g/L$. HCB is transformed into both pentachlorophenol and conjugates; however, elimination appears to be small compared to adipose deposits.			

HUMAN EFFECTS cont'd.				
Chemical	Score	Data Gaps	Comments	
Toxaphene	1	1	CHBs, the major constituent of toxaphene are commonly found in human tissue. Tolerable daily intakes exceeded by some Arctic population groups. Absorbed CHBs may be transformed and excreted; however, some congeners remain for prolonged periods. The two most common congeners are an octachlorobornane (Parlar no 26) and a nonachlorobornane (Parlar no 50). Values reported in human plasma are in the range $0.1-1 \ \mu g/L$ for both congeners. Several adverse effects of toxaphene, including being a possible human carcinogen, are reported. However, structure–activity models have shown that the most toxic CHB congeners are not accumulated.	
Atrazine	0	2		
Chlordecone	0	2		
Aldrin	0	2	Main source of human exposure is through food. Adverse effects include toxicity, immune system depression, and carcinogenic action	
Chlorinated Paraffins	0	2		
DDT	0	1	Levels of total DDT in human tissue in the Arctic are considerably higher than those in most southern populations, reflecting biomagnification in Arctic food-chains and the higher consumption of high trophic level species for food in the Arctic. Tolerable daily intakes exceeded by some Arctic population groups. Typical plasma levels are in the range 1–9 μ g/L. DDT and its metabolites are stored in fatty tissue and are excreted very slowly. Due to its lipophilicity, DDT and its metabolites are found in breast milk and can readily cross the placental barrier. Evidence suggests that the DDT group may suppress the immune system, mimic hormones and be a possible human carcinogen	
Dioxins	0	1	Chloracne is a well documented effect following dioxin/furan exposure. Other possible effects include effects on the immune system, cancer, reproductive disturbances, and acute toxic reactions. Typical sum PCDD/F values reported in human milk from the Arctic are in the range 10–40 pg/g lipid weight	
Endosulphan	0	2		
Endrin	0	2	Main source of human exposure is through food. Adverse effects include toxicity, immune system depression, and carcinogenic action	
Furans	0	1	Chloracne is a well documented effect following dioxin/furan exposure. Other possible effects include effects on the immune system, cancer, reproductive disturbances, and acute toxic reactions. Typical sum PCDD/F values reported in human milk from the Arctic are in the range 10–40 pg/g lipid weight	
Heptachlor	0	2	Typical plasma levels are in the range 0.05–0.15 μ g/L. The compound and its metabolite are classified as carcinogens	
Mirex	0	1	Mirex is found at low levels in human tissue. The primary source of exposure is food, especially meat, fish, and game. Transformation to photomirex is slow and elimination is mainly via feces and breast milk. Plasma levels in the range $0.1-0.6 \mu g/L$ are reported. Mirex has toxic and possible carcinogenic effects	
Nonylphenols	0	2		
OCS	0	2	OCS has not been regarded as a major contaminant in the Arctic. However, the metabolite 4-hydroxy heptachlorostyrene has recently been identified as one of the major metabolites in human plasma in the Arctic. The potential human health concern with OCS is because it has a binding affinity for both the androgen and estrogen receptor. The metabolite is believed to have an influence on thyroid hormone and retinol homeostasis.	
Octylphenols	0	2		
Org Lead Compds.	0	2		

HUMAN EFFECTS cont'd				
Chemical	Score	Data Gaps	Comments	
Org. Tin Compds.	0	2	In humans, acute exposure to organotin compounds (industrial exposure or laboratory accidents) has led to severe central nervous system damage, as well as to skin irritation. Although no data were found concerning low-level effects of TBT in humans in the Arctic, <i>in vitro</i> studies using human cells indicate potential immunological effects. Butyltin compounds were recently identified and measured in human blood from Michigan with total concentrations ranging up to $100 \mu g/L$.	
PAHs	0	2	The main sources of non-occupational exposure are cigarette smoke, smoke from wood combustion, and smoked or broiled (over charcoal or fire) food. Levels in humans are affected by smoking status, occupational exposure and consumption of PAH-containing foods. In non-smokers, the levels of the metabolite (2-naphthol) are below 2 μ mol/mol creatinine, whereas smokers exhibit average levels of 4 μ mol/mol creatinine. PAHs are not acutely toxic to humans. However, many PAHs have been shown to be potentially carcinogenic as well as genotoxic. Metabolites (e.g., hydroxides, epoxides, and nitro-PAH) appear to be responsible for these effects	
PBDE	0	2	Typical sum values of PBDEs (mainly BDE 47 and BDE 99) in plasma are in the range 5–40 ng/L (about 0.5% of levels of sum PCBs). Although little information is available, data on possible effects suggest that PBDEs are likely to be carcinogens, endocrine disrupters, and neurodevelopmental toxicants	
PCNs	0	2	Data on human liver and adipose tissue indicate PCN levels 200 to 500 times lower than the total PCB burden. Hydroxylated metabolites of PCN have also been reported.	
РСР	0	2	PCP concentrations in Arctic samples of 0.6–8 μ g/L plasma. A suspected carcinogen, however contaminants in technical grade PCP may be the actual culprits	
PFOS	0	2	No data exist on levels in humans from the Arctic	
Phthalates	0	2		

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