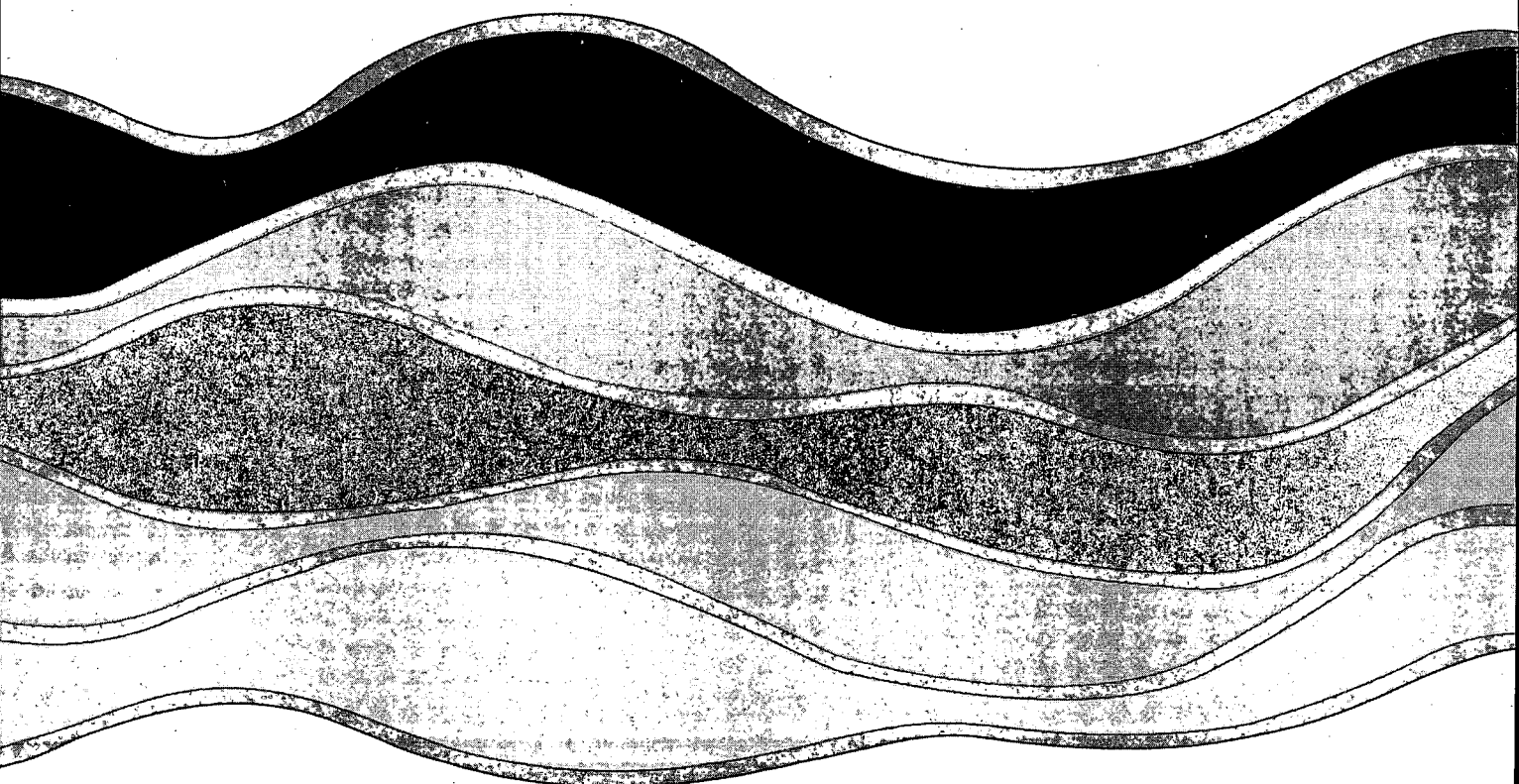NATIONAL
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TRACE ORGANIC CHEMICALS
IN THE ARCTIC ENVIRONMENT:
ATMOSPHERIC TRANSPORT
AND DEPOSITION

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TRACE ORGANIC CHEMICALS IN THE ARCTIC ENVIRONMENT:

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MANAGEMENT PERSPECTIVE

This is an invited chapter to a book titled "Pollution of the Arctic Atmosphere", edited by W.T. Sturges of the University of Colorado at Boulder and published by Elsevier Science Publishers Ltd. Of the 10 chapters in the book, this is the only one concerned with trace organic chemicals.

The chapter reviews the sources, emission processes, transport pathways and mechanisms of deposition of organochlorine pesticides, PCBs and PAHs relative to the Arctic. Although much of the atmospheric work has been conducted in Europe, actual measurements in the aquatic system (i.e., snow and surface water) is dominated by Canadian data.

Where possible, atmospheric concentrations and deposition rates for arctic regions have been compared with temperate Canadian and European sites. Aquatic concentrations of the compounds have also been compared. Data for the Canadian Arctic clearly suffer from a lack of samples as exemplified by Lake Hazen, the largest most northerly lake in the world, which has a total of six samples for organochlorine pesticides. Hemispheric comparison of results is restricted by the fact that sampling periods are dissimilar (by as much as a decade) while methods may not be comparable. It is nevertheless striking to note similarities in concentrations and fluxes to various environmental compartments between the Arctic and temperate regions.

This paper, along with others in the book, will clearly show that remoteness from industrial centres and agricultural lands does not provide adequate protection for the arctic environment from anthropogenic pollutants, especially trace organic compounds.

PERSPECTIVES DE LA DIRECTION

Ce chapitre fait partie d'un ouvrage intitulé "Pollution of the Arctique Atmosphère", compilé par W.T. Sturges, de l'université du Colorado à Boulder, et publié par Elsevier Science Publishers Ltd. Des 10 chapitres du volume, c'est le seul qui traite des substances organiques à l'état de traces.

Le chapitre passe en revue les sources, processus d'émission, trajets de transport et mécanismes de dépôt de pesticides organochlorés, BPC et HPA, dans leurs liens avec l'Arctique. Bien que la plus grande partie des travaux sur l'atmosphère aient été faits en Europe, les mesures concrètes sur le système aquatique (par exemple, neige et eau superficielle) sont dominées par les données canadiennes.

Dans la mesure du possible, on a comparé les concentrations atmosphériques et les taux de dépôt dans les régions arctiques avec ceux de sites tempérés du Canada et de l'Europe. On a aussi fait des comparaisons entre les concentrations aquatiques des composés. Les données pour l'Arctique canadien souffrent clairement d'un manque d'échantillons, l'exemple le plus marquant étant le lac Hazen, le plus grand lac des régions les plus septentrionales du monde, qui a six échantillons de pesticides organochlorés. La comparaison des résultats entre hémisphères est contrariée par l'absence de concordance entre les périodes d'échantillonnage (qui peuvent différer d'une décennie) alors que les méthodes ne sont pas nécessairement comparables. Il n'en est pas moins frappant de constater, entre l'Arctique et les régions tempérées, des similitudes dans les concentrations et les flux vers les diverses zones environnementales.

Cet article, de même que d'autres de l'ouvrage, montrera sans équivoque que, bien qu'éloigné des centres industriels et des terres agricoles, le milieu arctique n'est pas pour autant totalement à l'abri des polluants anthropogéniques, en particulier des substances organiques à l'état de traces.

CONTENTS

1. INTRODUCTION

As noted elsewhere in this volume, scientists have studied the glacial record and the atmosphere of the Arctic for several decades to investigate the residuals of anthropogenic activities in the most remote parts of the world. The phenomenon of arctic haze is a well known manifestation of hemispheric pollution in the North. Publications have generally considered increased acidity, increased sulphate and nitrate depositions, increased metals and the increase and subsequent decline of radionuclides from atmospheric bomb tests. However, there has been little interest in potential long term effects of these and other contaminants in the arctic environment.

Since the 1970's, evidence has been presented that long lived trace organic compounds are reaching the Arctic and accumulating in the ecosystem. Through measurement of body burdens of trace organic compounds in marine mammals and fish, it has become all too evident that these important biological food chains contain relatively elevated concentrations of organic chemicals. Local or regional sources could not possibly explain the presence of many of the compounds, especially the organochlorine pesticides which are for all intents and purposes foreign, in terms of use, to the Arctic. Consequently, the atmosphere, has to be an important pathway for these contaminants.

The first report of organic contaminants in the Arctic was by Holden as early as 1972 concerning concentrations of dichlorodiphenyl-trichlorethane (DDT) and polychlorinated biphenyls (PCBs) in seals[1]. Presently, there is an extensive data base of body burdens in arctic marine mammals and fish for DDT, chlordane related compounds, hexachlorocyclohexanes, toxaphene, chlorobenzenes, mirex, chlorinated dibenzo-p-dioxins and - dibenzofurans and PCBs[2,3,4,5]. The major organochlorine residue in Canadian arctic marine invertebrates and fish is toxaphene with arctic cod having toxaphene residues 5 to 10-fold higher than DDT or PCB residues[6]. Chlordane was present at levels similar to that of DDT in zooplankton and amphipods[7] and in ringed seal blubber at two east/central arctic locations[2]. In polar bears, chlordane was a major residue along with PCBs[3].

Concentrations of p,p'-DDT plus p,p'-DDE and PCBs in ringed seal, which are relatively sedentary, were higher in seals from Spitzbergen[8] than in the Canadian Arctic[2]. Arctic marine fish levels of ΣPCB (sum of individual congeners), ΣDDT (p,p'-DDE plus p,p'-DDD plus p,p'-DDT) and chlordane are 2 to 5 times lower than levels of these contaminants in fish from the North Atlantic, Baltic and Pacific northwest[9].

It is only recently that efforts have been directed at understanding more fully the sources, transport pathways and controlling processes of organic contaminants in the Arctic. This

has come about largely as a result of human health concerns. Northern natives of Canada have a much higher dependence on "country foods" (i.e., fish, game and marine mammals) as a dietary staple, than most other Canadian residents. The estimated annual per capita consumption of country foods by northern natives from the eastern Arctic of Canada ranges between 108 and 597 kg with a mean of 267 kg [10]. The residents of a single community in this region have been studied in some detail with respect to dietary patterns and PCB concentrations in blood and breast milk[11]. In northern Quebec, the PCB concentrations in milk fat of Inuit women was almost 5 times higher than Caucasian women and Dewailly et al.[12] suggest that PCBs could play a role in the impairment of immunity and in the high occurrence of infection among Inuit children.

This chapter reviews the relatively limited information available quantifying long range atmospheric transport of trace organic conmtaminants to the Arctic. In order to focus the content, only organochlorine pesticides, polychlorinated biphenyls and polycyclic aromatic hydrocarbons are considered. These classes of compounds are important environmentally as they tend to be long lived in the environment, may bioaccumulate and are frequently toxic and/or carcinogenic at elevated concentrations. Emphasis is placed on work that demonstrates a pathway between the atmosphere and deposition through measuring the occurrence of these compounds in the atmosphere and the abiotic arctic aquatic environment. The marine environment will not be discussed further as it is not

possible to differentiate between long range oceanic transport and atmospheric sources to the Arctic Ocean. Finally, brief mention is made of the limited information regarding trends of these compounds in the Arctic. However, before discussing the actual data, it is necessary to summarily review the physicochemical properties of these compounds, the processes controlling their emission and transport to the Arctic and their source regions.

A note of caution is appropriate at the outset of this paper due to the fact that data are compared from numerous published sources. That is, inclusion here does not imply that the author has either investigated or concluded that the data are compatible with respect to sampling and analytical procedures and quantification methods. This is particularly problematic for complex compounds like PCBs for which different analytical methods have been used by different investigators over time and different reporting methods exist. Thus comparison of results from different studies must be accepted in this context regardless of whether or not this is explicitly stated in the related discussion. In one effort to reduce confusion with respect to PCBs, the term ΣPCB will be used to express total PCB concentrations expressed as a sum of congeners. Note, however, that the same number of congeners may not be used by all authors. The term Ttl. PCB will be used to signify concentrations determined as equivalents of a single Aroclor or a 1:1:1 by volume mixture of Aroclor 1242, 1254 and 1260. Muir et al.[9] indicated that the use of Aroclor equivalents

overestimates PCB levels in marine mammals because of the extensive transformation of congeners. Although it is desirable to conduct congenor specific PCB analyses, it is not possible to replace the Aroclor equivalent data.

The term ΣDDT can be equally confusing as different authors have included different components. The term ΣDDT will be used here to represent the summation of p,p'-DDT, the derivative p,p'-DDD and the metabolite p,p'-DDE. In order to minimize confusion when different definitions apply, the term DDT alone will be used and the compounds included will be stated.

2. SOURCES, EMISSION AND ATMOSPHERIC EXCHANGE

2.1 Potential sources

Two classes of chemicals, here called chlorinated industrial organics and pesticides (OCs) and polycyclic aromatic hydrocarbons (PAHs) are considered. OCs relevant to the Arctic are those produced in large quantitities, released in part to the environment, long lived and widely dispersed. These include PCBs, hexachlorocyclohexane (HCH), chlordane, DDT, dieldrin, heptachlor and heptachlor epoxide, endosulfan, methoxychlor, hexachlorobenzene (HCB) and polychlorinated camphenes (PCCs) or toxaphene. Production and use statisitics for the OCs are difficult to uncover

but a recent summary for some of the compounds is provided in Barrie et al.[13] using information from a variety of sources (Table 1). Current regulations regarding these compounds have also been summarized for selected countries by these authors while more detailed information is maintained by the International Registry of Potentially Toxic Chemicals (IRPTC) of the United Nations Environmental Programme (UNEP).

PCBs are a concern as most of the 209 congeners, especially those with 5 or more chlorines, are biomagnified[2,14,15]. PCBs have been used worldwide for a variety of purposes with as much as 370×10^3 kg estimated to have been released into the environment[16]. PCBs are found in the North as a result of local use in transformers, capacitors and other equipment, and the disposal of wastes at industrial and military sites. There are, for example, 21 abandoned early warning radar stations and one abandoned US Coast Guard site across the Canadian Arctic. Recently, these sites were cleaned up with respect to intact containers of contaminated oils although the possibility of buried equipment and contaminated buildings and soils at a number of these sites remains. Nonetheless, site specific contaminantion of this nature can not account for the widespread presence of PCBs throughout the Arctic[17].

Organochlorine pesticides are a concern in the Arctic because of their long half lives and proclivity to bioaccumulate in aquatic and terrestrial food chains. Because pesticides are applied

intentionally over large areas of land, their sources and pathways to the Arctic may differ from PCBs, other industrial organics and PAHs.

HCH can theoretically exist as eight different stereo isomers of which only five are found in the technical product. The dominant isomers are α-HCH at 70% and lindane at 13% of the total[18]. The insecticidal isomer lindane is currently used in pure form in North America and western Europe[19,20]. Canadian usage in the early 1980's was estimated to be 100 to 150 t a[-1] of lindane[21]. Technical HCH is widely used in southern Asia and China with estimated annual use in the late 1970's of about 6x10[4] t [22,23,24].

Technical chlordane, a multi component product, was used as an insecticide but has more recently been used only for subterranean use in Canada and the US[21,25]. Since heptachlor is more insecticidal than chlordane, efforts have been made to purify the technical product to more than 70% heptachlor. Heptachlor is mainly used as a soil insecticide and for seed dressing, and has been reported to bioaccumulate. Heptachlor epoxide, a metabolite, is common in the environment, reportedly more resistant to chemical and biological changes in the aquatic environment than heptachlor, and has a predicted half life of several years[21].

DDT has been banned or restricted in Canada, US and western Europe since the early 1970's. Its use in Eastern Europe as recently as the mid 1980's is suggested from pine needle studies[26] and it continues to be manufactured and used in southern Asia, Africa and in Central and South America[19]. Dieldrin is considered to be persistent, volatile, and bioaccumulative[21]. This product has also been restricted in Canada and the US since the mid 1970's although it is still produced and used in a number of western European countries[20]. Endrin has properties similar to dieldrin and has been applied widely to cotton and rice[27]. Its use is presently restricted or it is banned in many developed countries[20].

Endosulfan, consisting primarily of the α-isomer (70%) photolyzes in the gaseous state and, although it is bioconcentrated, it appears to be rapidly broken down in the organism or is eliminated, and there is little danger of accumulation[21,27]. It is widely used on cotton and orchards, the latter because it is not toxic toward bees[21]. Methoxychlor is not stored in fatty tissues of higher organisms[21,27] and is still approved for agricultural use in Canada, particularly for the control of blackfly larvae. Use has decreased considerably since the 1970's [21]. It is also approved for use in the US, USSR and many European countries[20].

Toxaphene was the most heavily used insecticide in the US during the 1960's and 1970's. There it was used mainly for

controlling weevil on cotton[19,28] while it has been used only in a very restricted way in Canada[21]. Use in Mexico was estimated to be about 2,200 t in 1978 and projected as 2,000 t in 1988[19]. The USSR produces toxaphene derived from turpentine rather than camphene, for use as an agricultural pesticide[29], but production and use figures are not available.

HCB, representative of a number of chlorinated benzenes, has been used variously as a fungicide, insecticide and as wood preservative and can be a by-product of many industrial activities as well as the chlorination of wastewaters. As a result it is ubiquitous in the aquatic and atmospheric environments and is a concern because it is relatively resistant to biotic and abiotic degradation. Chlorobenzenes tend to bioaccumulate[21,30].

PAHs occur naturally in the environment as a result of forest and prairie grass fires. However, the quantity of PAHs in the environment has increased remarkably as a result of incomplete combustion of fossil fuels. A number of the PAHs are potentially carcinogenic[21]. Studies of PAHs in sediments of lakes in the Adirondacks of the eastern US show that atmospheric emissions there peaked in the 1950's corresponding to the decline in the use of coal and increased use of oil for space heating[31]. Since the combustion of fossil fuels for space heating increases during the winter, it is to be expected that PAHs will be present in the Arctic during the winter season.

The sources of the compounds of concern in the Arctic are potentially global in nature and may be emitted to the atmosphere either directly through use (e.g. agricultural pesticides) or indirectly and unintentionally as part of industrial, combustion and other processes (e.g. PCBs and PAHs). Once introduced into the atmosphere, these compounds may circulate in the atmosphere and hydrosphere for long periods of time prior to going to a permanent sink or being broken down or bound up such that they are effectively removed from the system. Clearly, for all of these compounds, the major sources are outside of the arctic region although local combustion sources of PAHs will exist and improper disposal of dielectric fluids and PCB contaminated wastes has resulted in very localized contamination.

The information regarding sources and transport of trace organic contaminants to the arctic environment is inadequate at this time. However, it is possible that the more reliable information for sulphur sources can be instructive. Barrie et al.[32] estimated that 96% of the sulphur input to the Arctic in 1979/80 was from Eurasia while only 4% was from North America (see chapter 6). The Eurasian inputs were almost equally split between the Soviet Union and Europe (east and west). These authors further noted that most of the sulphur was transported into the region in the lower 2.5 km of the atmosphere. Consequently, future research with respect to sources and atmospheric pathways and processes for trace organic contaminants should emphasize Eurasia and transpolar

circulation.

2.2 Emission and atmospheric exchange

If a specific organic compound is chemically stable with
respect to, for example, photodegradation and chemical
transformation, and is semi-volatile, this compound will tend to
migrate over long distances through repeated deposition and
volatilization. Many organic compounds are likely to be
transported in this way[33]. The vapor pressure, organic content and
the concentration, size and surface area of total suspended
particles control the distribution or partitioning between vapor
and particles. Vapor-particle partitioning in turn will influence
the distance travelled by these contaminants by determining whether
the compound is scavenged by rain and snow, by dry particle
deposition, or by direct vapor exchange across the air-water
interface. Overviews of these atmospheric processes are
available[33,34,35,36]. It is sufficient here to note that compounds
with saturation vapor pressures greater than 1.3×10^{-2} Pa should
exist almost entirely in the vapor phase while those having vapor
pressures less than 1.3×10^{-6} Pa should exist almost entirely in the
particulate phase[34]. Most high molecular weight organics have vapor
pressures between these extremes, as shown in Table 2, and their
atmospheric lifetimes depend on the nature and concentration of
particles in the air mass.

PCBs have been shown to be between 80 and 100% in the vapor phase in urban atmospheres and in the area of the Laurentian Great Lakes[34,37,38]. Duinker and Bouchertall[39] investigated the partitioning of several PCB congeners and concluded that although only a very small fraction of PCBs were found in aerosols in air samples, more than 99% of the PCBs in rain were particulate associated. More highly chlorinated congeners were particle associated while the lower chlorinated congeners remained essentially in the vapor phase. Thus the method of deposition can affect the toxicity of the PCBs and can influence how readily they can be revolatilized from the earth's surface.

Particle deposition processes in polar regions have been reviewed by Barrie[40]. They depend mainly on physical factors including particle size distribution, surface roughness and wind speed. Vapor phase exchange with the surface, which may be water (fresh or saline), snow or soil, depends on the physicochemical properties of the compound as discussed above, which are in turn temperature dependent[41]. Limited summer arctic air and seawater measurements show a wide range of atmospheric saturation indices averaging about 60% and as high as 90% for α-HCH, 40% for lindane[7,41,42] and 30% for toxaphene[7,41]. Although published data do not appear to be available to confirm this, the cold dry arctic winter conditions should increase the proportion of dry deposition of aerosol associated contaminants to total contaminant deposition.

Exchange processes and rates between water or soil and the atmosphere have been researched, but usually not at arctic temperatures[38,43,44,45,46]. Only recently, have exchange processes between the surface snow and the arctic atmosphere been considered for volatile organic compounds. Results of replicate sampling of the 1985/86 snow layer on the Agassiz Ice Cap, Ellesmere Island, Canada (80° 40'N 73° 30'W) in May of both 1986 and 1987 are presented in Table 3 [13,47]. Except for Ttl. PCBs, the concentration of the OCs measured in this layer in 1986 was significantly greater than the concentration measured in the same layer in the following year. Sampling was also repeated over the two years in annual firn layers with good agreement between measured concentrations indicating that this was not an analytical artifact. Comparison of the snow accumulation layer for the winter of 1986/87 sampled in May of 1987 and again in May of 1989 shows residual concentrations of OC pesticides of about 1% in the glacier relative to the depositional concentration measured in 1987 (Gregor, unpublished data). Since melting on the glacier was limited, as indicated by the preserved specific conductance profile presented in Figure 1, and the snow layers below showed consistent low concentrations, it must be concluded that large portions of the OCs, with the exception of the PCBs, revolatilized to the atmosphere during the summer season as the air and snow temperatures rose. It is conceivable that, although a large portion of the PCBs stay, it is the lower chlorinated congeners that are lost from the snow.

Supporting evidence for revolatilization from the snow in the Arctic comes from Hargrave et al. [48] who detected HCB in snow collected in May-June, 1986 (33 pg l^{-1}) but not in snow from the same area in August-September. HCB has the highest Henry's Law Constant and the lowest water solubility of the compounds listed in Table 2 and thus it is readily volatilized from the snow. At 0°C, more than 99% of HCB was still found in the vapor phase by Bidleman et al. [49]. Similarly, chlordane was found at 91 pg l^{-1} in the snow sampled in May-June while it was undetectable (<1 pg l^{-1}) in August[48].

The knowledge base for OCs remains severely inadequate and in order to understand the processes and fate of organochlorines in the Arctic, more research is essential in polar and sub-polar conditions.

Since the combustion of fossil fuels in the northern hemisphere increases during the winter season and transport from the mid-latitudes to the Arctic is more efficient in winter, it is to be expected that the products of combustion would occur in remote arctic regions. Local sources may also be important. The sampling and analysis of PAHs is considerably more difficult than for OCs, especially in the Arctic, where local conditions and the equipment associated with sampling (e.g., aircraft) can themselves be sources. Edwards[50] notes that the extraction and recovery of PAHs is problematic, resulting in underestimates and considerable

sample variability.

Tuominen et al.[51] demonstrated that the largest proportion of PAHs in the atmosphere in Finland was associated with particulate matter and that precipitation, specifically snowfall, was an efficient scavenger of particulate associated PAH. Bidleman[33] argued that the proportion of aerosol bound PAHs increased inversely with temperature and thus both wet and dry deposition were likely to be important at ambient northern temperatures. Losses of PAHs from the arctic snowpack and air-water exchange of PAHs have apparently not been addressed in the published literature.

3. OCCURRENCE IN THE ARCTIC ENVIRONMENT

3.1 Atmosphere

There is a marked seasonal distinction between the meteorologic conditions controlling the long range transport of trace organic pollutants to the Arctic, as discussed in Chapter 2 of this volume. Episodes of pollution are much more prevalent in the late winter and early spring.

Much of the published OC data for the arctic atmosphere has come from Scandinavian countries which are more directly affected at all latitudes by both summer and winter atmospheric transport from sources to the south and east. Selected data from Scandinavia are compared to the relatively recent and geographically limited Canadian data and data from the Bering and Chukchi Sea region in Table 4. HCH has consistently been found to be the dominant atmospheric OC in the Arctic with α-HCH exceeding lindane. It has been observed[52,53] that concentrations of α-HCH in winter time are lower than in summer while higher concentrations of lindane are observed in the winter season. Air masses in the Arctic show α-HCH to lindane ratios of 10:1 in winter and as much as 50:1 and 500:1 in summer[52] and fall[53], respectively. The higher summer ratio is thought to be a result of the more rapid photo-degradation of lindane during the summer[52] and to preferential scavenging of lindane from the atmosphere.

HCB is frequently the next most dominant OC compound in the arctic atmosphere (see Table 4). Concentrations are similar from around the world and show little seasonal variation[54]. PCBs and DDT are detected throughout the Arctic but at concentrations much lower than the HCHs. Interestingly, Larsson and Okla[55] have been able to distinguish a decrease in concentration of DDT (p,p'-DDT plus p,p'-DDE) along a south-north gradient in Sweden. A gradient of this type is expected with increasing distance from the source region, however, these authors also show that the levels of PCBs and DDT in

the lower atmosphere are positively related to the mean temperatures of the sampling station. Since the amount of gas phase pollutant in the atmosphere is dependent on vapor pressure, which in turn is a function of temperature, it is conceivable that as temperature decreases northward, a decreasing amount of a pollutant is able to remain in the gas phase and thus adsorption or deposition occurs.

Chlordane related compounds were first observed in the Canadian arctic air at Mould Bay (76° 15'N, 119° 16'W) in July, 1984 [58] and more recently on the Canadian Ice Island (approximately located at 81°N, 97°W) in 1986 and 1987 at 2 to 10 pg m^{-3} (total chlordane related compounds)[41,48]. Between February and April 1988, total chlordane concentrations in air averaged 3.3 pg m^{-3} at Alert (82°30'N, 62° 18'W)[13,56]. Concentrations of α-chlordane alone have been reported generally below 5 pg m^{-3} for arctic sites in Norway[52,53,54]. These concentrations were comparable to southern Sweden but much lower than sites in the continental US in the mid 1980's[49].

Dieldrin has been detected in arctic air at between 0.1 - 5 pg m^{-3} at the Canadian Ice Island in 1986 and 1987 [41]. Only trace quantities were reported by Hargrave et al.[48] but with higher detection limits. The most abundant OC after HCH and HCB at this site was toxaphene at concentrations between 36 and 44 pg m^{-3} in the summers of 1986 and 1987[7,41].

PCCs (toxaphene) were the most abundant pesticide residues in Canadian arctic fish and marine mammals[2,3,6]. Concentrations of PCCs in the arctic atmosphere at the Canadian Ice Island were reported for the first time at 44 pg m^{-3} for August, 1986 (mean of 7 samples) and at 36 pg m^{-3} in June of 1987 (mean of 6 samples)[7,41]. Average concentrations of toxaphene at Alert in February to April, 1988 were nearly identical to the earlier summer measurements at 35 pg m^{-3} [56]. Although other published measurements are not available for north of 60°N, these measurements are comparable to atmospheric concentrations in Sweden in 1983-1985 [49].

Most studies have shown that the organochlorine compounds are concentrated in the gas phase, although the use of relative concentrations on filters (particulate associated) and absorbents (gas phase) in large volume air samplers may not be precise. Specifically, adsorption and desorption to and from particles may occur under varying conditions (e.g., temperature changes) over the course of the sampling period for large volume air samples or due to physical desorption as a result of pulling air through the filter[33,43]. Larsson and Okla[43] determined that relatively high, near source particulate PCB concentrations ranged from 0.4 to 7.3% of the total PCBs. Oehme and Mano[52] reported more than 98% of all OC compounds in the vapor phase while Bidleman et al. [49] reported less than 10% of OCs were particle associated at 20°C. These authors noted that the percentage of particle associated p,p'-DDT increased to as much as 74% at 0°C. Other OCs increased less

dramatically. HCH and HCB remained at less than 1% particle associated even at 0°C. The absence of quantifiable concentrations of OCs in aerosols was noted at the Canadian Ice Island by Hargrave et al.[48]. Further understanding of vapor-particle partitioning of trace organic contaminants at sub-zero temperatures is essential to understanding processes controlling air-surface exchanges in the Arctic.

Relatively few measurements of PAHs in the arctic atmosphere are available. Data from Alaska and Canada have been summarized in Table 5. Winter concentrations at remote sites (Alert and Narwhal Island) are similar to summer concentrations at Barrow, somewhat less than winter concentrations at Barrow and a great deal less than winter concentrations at Fairbanks. Daisey et al.[59] conclude that much of the PAH at Barrow during the winter comes from long range transport as local sources are minimal due to the use of natural gas as a heating fuel. However, the summer concentrations appear to be influenced by local emissions as suggested by the predominance of coronene and benzo(ghi)perylene which are more typical of automobile and fuel oil emissions. These authors further observe that the mean PAH distribution for March samples appears to be closely related to those observed in Norway and Sweden between December 1976 and April 1977 for periods when air masses circulate from Europe[60]. In contrast, the much higher concentrations in Fairbanks are thought to be due to vehicular exhaust combined with coal fired power plant emissions[61]. Clearly,

a better understanding of PAHs is required.

3.2 Concentrations in the arctic freshwater system

This section considers two compartments of the aquatic system. First, measurements of contaminants in snow are discussed as direct evidence of long range atmospheric transport and deposition to the arctic aquatic environment. Second, the limited data for river and lake systems are presented to link deposition to the aquatic system. In both cases, comparisons will be made with temperate regions.

The earliest known reported work on organochlorine compounds in the Canadian Arctic was undertaken in 1970 as part of the Ice Field Ranges Research Project (IRRP) in the Yukon Territory[62]. Sampling for subsequent analyses of DDT was undertaken at an elevation of 5,364 m on Mount Logan. Nineteen samples were taken in snow at depths of 1 to 15 m with DDT not detected in any of these samples at a detection limit of 5 ng l^{-1}.

The earliest measurements in northern Sweden were conducted in 1972 and 1973 using a silicone oil impregnated nylon mesh to measure airborne fallout[55]. These data (see Table 6) were compared to results from a similar study conducted in 1984-1985 and it was concluded that the overall fallout of DDT (p,p'-DDT plus p,p'-DDE)

decreased during the intervening period. The ratios of DDT to DDE were found to have decreased from 4.10 (n=17) in 1972-1973 to 2.64 (n=52) in the more recent study. Since DDT degrades to DDE in ultraviolet light in the atmosphere and by the metabolism of organisms, it was concluded that the restrictions on DDT use in Sweden in 1971 had been effective and that the DDT reaching Sweden presently has "aged" during long range transport from distant source regions. The trend of PCB concentrations over this time period was more complicated having decreased slightly in the northern part of Sweden[55].

Although still limited, recent work has been concentrated in the Canadian Arctic. The Finnish Huure Expedition to the North Pole in 1984 collected 1 1 of water equivalent snow samples for analyses of a broad range of chlorinated pesticides and chlorophenols[64]. None of the target compounds were detected in these snow samples. Based on the work of Gregor and Gummer[65] it would have been reasonable to expect to see at least lindane (α-HCH was not one of the target compounds) at the Finnish study's reported detection limit of 0.5 ng l^{-1}. The most comparable station from the 1987 study is the Arctic Ocean site which had Ttl. PCB and lindane concentrations of approximately 1.0 and 0.9 ng l^{-1}, respectively. Annual variation may well be such that these compounds in 1984 were below the level of detection of the Finnish study.

In 1979, McNeely and Gummer[66] undertook snow sampling in the area of east-central Ellesmere Island. Sampling included snow pack at 5 sites, fresh snow at 3 sites and rime frost from 2 sites between May 24 and June 19, 1979, yielding 19 samples with replicates and repeat analyses for a total of 28 analyses. The results for organochlorine pesticides are summarized and compared to selected data from the much more extensive study of Gregor and Gummer[65] in Table 7. The two stations selected for comparison were Alexandra Fiord sampled in 1986, the only station overlapping between the two studies, and the surface snow for both 1986 and 1987 from the Agassiz Ice Cap, approximately 175 km to the north. Although the results for Alexandra Fiord for 1986 were below the level of detection available to McNeely and Gummer, the observations at the Agassiz Ice Cap support HCH concentrations of the magnitude found in this earlier study.

Concurrent with the 1987 snow chemistry survey of Gregor and Gummer, Arctic Laboratories Limited undertook work at the Canadian Ice Island during 1986 and 1987 [67]. Data from these two studies are compared in Table 8 and show good agreement given the very low concentrations seen in these samples. HCH data for 1987 Ice Island snow samples are compared for three different laboratories using three different sampling methods in Table 9. The USC results are relatively high and lindane is dominant which does not agree with the other analyses and is unusual for the Arctic. The difficulty in measuring the very low concentrations in the arctic aquatic system,

as illustrated here, demonstrates the need for more extensive inter-laboratory evaluations to permit data pooling and comparisons.

Toxaphene measurements in the arctic snowpack are restricted to four samples. Two of these were collected from the Canadian Ice Island in May 1986 and two others were collected from the Devon Ice Cap on Devon Island (75° 6'N, 82° 45'W) and from the vicinity of Resolute Bay on Cornwallis Island (74° 42'N, 94° 54'W) in May, 1987. Concentrations for the two Ice Island samples were 85 and 307 pg l^{-1} while the Resolute Bay and Devon Ice Cap samples were 600 and 1700 pg l^{-1}, respectively[7]. The wide range in concentration between Resolute Bay and Devon Ice Cap was not surprising given the distance between these two stations, the difference in elevation (nearly 1,700 m) and different snow accumulation rates. There are no known published measurements of toxaphene concentrations in arctic surface fresh waters.

The most northerly data from the Soviet Union found to date although not within the definition of the Arctic used here, are the results of precipitation samples collected in the Moscow region from January 1978 to September 1979[68]. Although Bobovnikova and Dibtseva[68] did not indicate specifically which monthly samples were snow, it can be reasonably assumed for this lattitude that samples from December through March were primarily snowfall. Based on these 4 months, it can be estimated that DDT (sum of p,p'-DDT, o,p'-DDT, p,p'-DDD and p,p'-DDE) ranged from 76 to 110 ng l^{-1}; α-HCH ranged from "trace" to

25 ng l^{-1}; and lindane ranged from "trace" to 30 ng l^{-1}. The detection limit, apparently indicated by the term "trace", for any of these compounds was not stated. The concentrations of α-HCH were similar to those noted for the Canadian Arctic[65] while lindane tends to be somewhat higher, likely reflecting proximity to the source region. Similarly, DDT was an order of magnitude higher in the Moscow snow samples[65] than the arctic snow samples collected by Gregor and Gummer[65].

There do not seem to be published data for organochlorine concentrations in arctic rivers and lakes except as reported by Erickson and Fowler[69] for the Mackenzie River and by this author[70,71] for Sophia and Amituk Lakes on Cornwallis Island and for Lake Hazen on northern Ellesmere Island. Data from these sites for α-HCH, lindane and HCB are compared with measurements from rivers in northern Ontario[72] and northern Quebec[73] and from Lakes Superior and Ontario[74] in central Canada in Table 10. Although the number of samples in the Canadian Arctic are few, the concentrations of HCHs and HCB are within the same ranges as those in rivers in northern Quebec and Ontario. These in turn are generally comparable to the concentrations of the two Laurentian Great Lakes reported here. The very high concentration of HCH in the surface waters of Sophia Lake is perhaps attributable to the fact that this is a meromictic lake with a very shallow surface layer resulting in trapping and minimal dilution of surface runoff. α-HCH was found above the detection limit in four of five lakes in Labrador, Canada while lindane was detected in only one of these lakes[75].

Lindane, in daylight, can be transformed to the α-HCH isomer with a half life, dependent upon temperature and other conditions, of the order of one year[53]. Although degradation rates are likely substantially different in a polar climate than in a temperate one, this ratio may be informative with respect to possible age or length of time of HCH transport. Also, chemical formulations differ. North America uses pure lindane with aerosol ratios reported at 1:7 in the Great Lakes region[34] while the HCH mixtures utilized in Europe produce ratios of 1:3-4 [52]. As shown in Table 2, the water solubility of lindane is one to two orders of magnitude higher than α-HCH. This could result in preferential scavenging of lindane by precipitation which will in turn affect the ratios of the residual compounds measured in the arctic atmosphere.

Alpha/gamma-HCH ratios in the air at the Canadian Ice Island were 6:1 and 15:1 in the spring and summer, respectively, of 1986 (see Table 4)[48]. Based on Table 3, the ratio for surface snow from the Agassiz Ice Cap in 1986 was 2:1 but 4:1 a year later. The average ratio for all surface snow samples collected by Gregor and Gummer[65] in 1986 and 1987 was 2.5:1. These low ratios in the snow relative to the atmosphere, may indicate preferential scavenging of lindane by precipitation. Whether the difference in solubility is maintained at sub-freezing temperatures is unknown. Another explanation could be that the snow pack ratios represent deposition during pollution events which carry relatively fresh HCH to the Arctic whereas the air measurements possibly represent a background

ratio of older, recycled HCH.

The ratios of α-HCH to lindane for various water bodies are also shown in Table 10. Although by no means conclusive, it is noteworthy that Hazen Lake has HCH ratios similar to Lake Ontario while Sophia Lake and Amituk Lake have ratios comparable to Lake Superior and northern Ontario and Quebec rivers. Further speculation on the significance of HCH ratios requires a better understanding of degradation processes in the Arctic as well as a more extensive data base.

Arctic snow samples collected in 1987 have been analyzed by this author (unpublished data) for 17 PAHs by capillary gas chromatography with electron impact, multiple ion monitoring mass spectrometric detection (MIM-GC/MSD). ΣPAH concentrations were highly variable ranging from non-detectable to 125 ng l^{-1}. The seven PAHs detected consistently were phenanthrene, 1- and 2-methylnapthalene, 1,2,3,4-tetrahydro-naphthalene and indene with generally minor quantities of fluorene and/or acenaphthene.

All seven of these PAHs are coal tar derivatives, practically insoluble or having low water solubilities (although having higher solubilities than many of the OCs) and of relatively low molecular weights for PAHs (Table 2). These PAHs are mildly toxic with some having a tendency to bioacumulate, to be slightly carcinogenic and even to be mutagenic. Some, such as acenaphthene and 1,2,3,4-

tetrahydro-naphthalene have commercial uses either alone or in conjunction with other products[76]. Compounds commonly associated with internal combustion exhausts (e.g., fluoranthene, phenanthrene and pyrene) were either not detected or were detected only occassionally or were believed to be sample contamination as evidenced in method blanks. Relative to other PAHs, it appears that it is the low molecular weight, high vapor pressure, more water soluble PAHs that are being found in the arctic snow.

PAHs were not detected above the limit of detection for any of the arctic snow samples from 1979 [66]. PAHs have been measured in fresh snow samples in Bavaria, F.R.G.[77] and in Switzerland[78] at concentrations ranging from about 0.2 to 2 μg l^{-1}, an order of magnitude higher than seen in the Arctic. Phenanthrene, fluoranthene and pyrene were dominant in the Swiss samples of which only phenanthrene was detected in the Arctic. Evidently, the PAHs transported to the arctic generally have higher Henry's Law Constants than the PAHs commonly found in these temperate areas.

3.3 Aerial deposition rates

The aerial deposition rates of contaminants to the land surface from precipitation is a more appropriate value than concentrations for determining spatial patterns and temporal trends. Median winter deposition rates for selected organochlorines based on concentrations in Canadian arctic snowpack in the spring of 1986 and 1987 are

presented in Table 11 [13,70]. Although these are shown as winter deposition rates, it is reasonable to assume that these represent, for all intents and purposes, annual deposition in the Arctic. There are two facts supporting this assumption. First, approximately 80% of the total precipitation in the high Arctic arrives as snowfall[79]. Second, the period of snow accumulation at these arctic stations generally ranges from September to May and this encompasses the period during which the arctic aerosol has been shown to be most heavily contaminated with anthropogenic pollutants[32,80,81].

These deposition estimates are limited by the fact that snow samples are commonly collected from remote sites and total annual snowfall has to be assumed to be equal to that recorded at the nearest weather station which may be several hundreds of kilometres distant. In addition, total snow storage in a basin generally exceeds weather station records by as much as from 130 to 300% [79,82]. It must also be remembered that deposition to the snow pack does not necessarily mean a direct transfer of this quantity of contaminant to the aquatic system following snow melt. As discussed in section 2.2, the majority of the measured pesticides disappear from the surface layer of Agassiz Ice Cap over the course of a year without substantial melting[47]. Attempts to confirm that this phenomenon occurs in snow that totally melts annually were undertaken in the vicinity of Resolute Bay in 1989 but with inconclusive results (Gregor, unpublished data). Nontheless, the concentrations of HCHs present in several arctic lakes supports the conclusion that large

quantities of trace organic contaminants are transported during snowmelt.

Loading estimates of organochlorine compounds have been made for the Great Lakes area[34,83] and for stations from across Canada[84]. The samplers in the latter study collected only rainfall, using lids to preclude dry deposition. The Canada wide data, adjusted by Strachan[84] to represent annual precipitation, are compared to ranges for the Canadian arctic data in Table 12. Interestingly, the annual deposition of pesticides to the Canadian Arctic was estimated to be nearly equal to, or only marginally less than, all regions sampled by Strachan except for the British Columbia (B.C.) station which received much more rainfall than the other sites. Total PCB deposition to the Arctic, on the other hand, was lower than all other sites. Highest PCB deposition, reported for the Lake Superior region of Canada, was approximately 10 times more than the Arctic. The high PCB deposition to the Upper Laurentian Great Lakes region is supported by the work of Murphy and Rzeszutko[85] and Swain[86].

The most northerly published data for the USSR appear to be limited to the annual fluxes in rainfall and snowfall provided by Bobovnikova and Dibtseva[68] for the Moscow area. Fluxes are estimated to be about 52 μg m^{-2} yr^{-1} for DDT (p,p'-DDT, o,p'-DDT, p,p'-DDD and p,p'-DDE) and 14 and 12 μg m^{-2} yr^{-1} for α-HCH and lindane, respectively, for the 1978 calendar year. The flux of α-HCH and lindane is about an order of magnitude greater and DDT is more than

two orders of magnitude greater in the Moscow region than in the Canadian Arctic. As suggested above, these data likely show much closer proximity to source regions. However, the Moscow data precede the arctic data by nearly 10 years and conditions could have changed considerably during this time due to different pesticide formulations, application procedures and quantities used.

At the south pole, DDT (o,p'-DDT plus p,p'-DDT) residues of the order of 100 to 2,000 pg l^{-1} have been reported in Antarctic snow for samples representing the period of snowfall between 1965 and 1969 [87]. These results questioned those of Peterle[88] which were considerably higher.

Rapaport et al.[89] analysed snow and rain samples collected at Minneapolis, Minnesota, USA from 1981 through 1983 for p,p'-DDT. Mean annual fluxes of p,p'-DDT for snow reported by these authors averaged 0.2, 0.4 and 0.13 μg m^{-2} yr^{-1} for 1981/82, 1982/83 and 1983/84, respectively. These fluxes were 10 to 100 times higher than the DDT (p,p'-DDE, p,p'-DDD, o,p'-DDT and p,p'-DDT) fluxes observed for the arctic sites in which the technical product of p,p'-DDT commonly constituted more than 75%.

3.4 Deposition trends

To date there has been limited conclusive research regarding trends of trace organic contaminant deposition in the Arctic despite the extensive work conducted on glacial records for other anthropogenic contaminants (see chapter 3). Periodic measurements in air and fresh snow have been too restricted temporally to provide any time series information with the exception of the Swedish study of Larsson and Okla[55] noted in section 3.2. Stengle et al.[62] sampled at depth on Mount Logan but could not quantify DDT. Thus the only work intended specifically to determine temporal trends of OC deposition is from the Agassiz Ice Cap on north-central Ellesmere Island.

The Agassiz Ice Cap has been sampled at a remote site at an elevation of approximately 2,000 m asl in both 1986 and 1987 to include 17 years of samples between 1969/70 and 1986/87 [13,47]. Intuitively, this cold glacier should provide a credible historical record of anthropogenic pollutant deposition because (i) average summer melt affects only about 3% of the winter snow layer resulting in negligible redistribution of ions between adjacent snow layers[81] and (ii) annual snow accumulation at this lattitude and elevation represents, on average, more than 75% of the total annual precipitation[79]. The seasonal specific conductance record of the firn (Figure 1) is clearly preserved and is consistent with that reported by Barrie et al.[81]. As noted by these authors, this profile reflects the trace constituent composition of the atmospheric aerosol which

undergoes a strong seasonal variation with maximum ion concentrations, and therefore specific conductance, during the arctic winter[90,91].

The large volume sample required for trace organic chemistry restricts the profiling for these compounds to annual layers. At first glance, it appears that annual quantities of chlorohydrocarbons in the firn at the Agassiz Ice Cap decreased from winter of 1969/70 through to the winter of 1986/87 as follows:

- α-HCH   from >300 to 70 ng m^{-2};
- lindane   from 90 to <20 ng m^{-2};
- heptachlor
  epoxide   from 20 to 5 ng m^{-2};
- dieldrin   from >50 to 20 ng m^{-2};
- chlordane   from 20 to <5 ng m^{-2}; and,
- Ttl. PCB   from >300 to 100 ng m^{-2}.

The consistent decline among all compounds is quite remarkable. However, as noted in section 2.2, the firn does not seem to be an effective permanant trap of the volatile trace organic compounds with the exception of PCBs. The residues in the firn therefore are not quantitatively representative of the annual flux of these pesticides to the glacier. It must then be concluded that the trends seen in the glacier for lindane, α-HCH, heptachlor epoxide, dieldrin and chlordane are not entirely the result of a trend in annual loadings, but rather are being controlled, to some degree, by annual meteorological conditions influencing the snow/atmosphere exchange.

Until the processes controlling deposition and revolatilization are better understood, it appears that only Ttl. PCBs demonstrate a real trend in annual deposition to the ice cap. The residue masses of Ttl. PCBs in the glacier will be influenced somewhat by the annual variability of summertime losses, nevertheless, it is conceivable that the actual annual deposition is dominant. Since meteorologic conditions that control atmospheric transport to the Arctic and deposition to the snow vary from year to year, it is necessary to consider this variability and to attempt to adjust for it. Gregor et al.[47] normalized the Ttl. PCB trend for annual meteorologic variability with ^{210}Pb flux measurements which were also determined for the glacier. The radioactive isotope ^{210}Pb is derived from the decay of ^{222}Rn which emanates from the earth's surface where it is produced as a member of the ^{238}U decay series. ^{222}Rn decays in the atmosphere to ^{210}Pb with a half life of 3.8 days. The charged daughter ions produced are chemically reactive and soon become irreversibly associated with aerosol particles[92]. Since the source of ^{210}Pb (i.e., the earth's surface) remains more or less constant with time, it can be hypothesized that the deposition of ^{210}Pb to the glacier would be uniform from year to year if the air mass movements and the complex interaction of factors controlling transport and deposition to the glacier remain constant. Thus, the differences of measured decay corrected fluxes in the snow layers can be assumed to represent, simplistically, the year to year differences in atmospheric transport and deposition of terrestrially derived anthropogenic pollutants. Under these assumptions, any trends in these adjusted trace organic

substance flux data should therefore be real changes in the flux of contaminants to the glacier.

The adjusted Ttl. PCB residues in the annual layers of the Agassiz Ice Cap are compared to measured quantities in Figure 2. These results imply that the tripling of the aerial mass of PCBs in the glacier beginning about 1975/76 is largely an artifact of variable meteorologic conditions affecting transport and deposition. The quantity of PCBs in the glacier has evidently remained stable since 1976/77 at a level approximately one half of that which prevailed between 1970/71 and 1975/76.

Although this decline in the glacier agrees in general with trends in PCB body burdens in ringed seals from the western Arctic between 1972 and 1981 [93] and for the Lancaster Sound area between 1975-76 and 1984[2] it is possible that correspondence among these limited data is merely fortuitous and a great deal more work is required to thoroughly evaluate trends in contaminant deposition in the Arctic.

4. Summary and Conclusions

The arctic atmosphere and polar glaciers have received considerable research over the past several decades for the purpose of measuring the extent of anthropogenic contamination of remote regions. But only relatively recent research has been concerned with the transport, deposition and accumulation of trace organic contaminants. Extensive data on contaminant body burdens of marine fish and seals have been compiled. However, information on the sources, atmospheric pathways, present deposition rates, deposition trends and processes controlling the fate of these contaminants in the Arctic is sparse.

If the source areas and atmospheric pathways for OCs and PAHs are similar to those for sulphates, then it is conceivable that Eurasia is a major contributor to the concentrations seen in the Arctic. This, however, needs to be confirmed. Local sources in the Canadian Arctic are not significant at the regional scale. Significant quantities of OCs and PAHs are found in the arctic atmosphere during the winter season indicating that large scale, long range transport does occur. In addition, some evidence exists that large percentages of the high vapor pressure compounds may be revolatilized to the atmosphere from the snow as temperatures rise. These processes warrant more research under arctic conditions.

Evidence of deposition and incorporation into the arctic aquatic system is provided by the rather limited data on fluxes and river/lake concentrations. Annual deposition of OCs in the Canadian Arctic are of the same order as for southern Canada. PAHs, which are more variable and more readily influenced by local activities, are more difficult to assess. Overall, there are no great differences between the concentrations of HCHs and HCB in arctic aquatic systems than more southerly rivers and lakes of Canada. However, some of the less volatile OCs do not seem to be present in northern waters.

Despite extensive work utilizing glacial ice cores for monitoring pollutant trends for acid deposition and climate change, there has been only limited research on the glacial record of OC deposition. The only glacier in the northern hemisphere for which published data exist is the Agassiz Ice Cap on Ellesmere Island. This research, although limited to 17 years of record at this time, indicates first that the glacial contaminant residues are not necessarily indicative of actual deposition. Only the PCB record appears relatively intact and these data, after correction for annual meteorologic differences, suggest that Ttl. PCB deposition has decreased by about a factor of 2 since 1970.

Clearly, more comprehensive research is required to expand the information base for trace organic contaminants in the Arctic. This research must seek a broader geographical and temporal coverage. Equally as important, though, is the research directed at

understanding the processes and fate of these contaminants in a polar environment. This must come from both laboratory and field based research. Only with this information, combined with ecological effects assessments, will an understanding of the potential of trace organic contaminants to impair the delicate arctic ecosystem be attained. Protection of the balance of this ecosystem, which is so intertwined with the support systems of the indigenous peoples, may depend on rapid aquisition of essential information to demonstrate the need for more extensive, hemispheric restrictions of the contaminants considered here.

List of Figures

specific conductance (usie cm^{-1}at 25°C)

FIGURE 1

FIGURE 2

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Table 1   Global production and use estimates of major organochlorines (modified from Barrie et al.[13])

compound	use	period of manufacture	estimated global production (megatonnes)			ref.
			global	USA	Other	
PCBs	dielectric and hydraulic fluids	1930-present	1.2	0.6	0.4	19
DDT	insecticide	1940-present	1.5	≈1	≈0.2[a]	28
toxaphene	insecticide	1947-present	-	0.5	≈0.2[b]	19,28
chlordane	insecticide	1945-present	-	≈0.25	-	25
hexachlorobenzene (HCB)	fungicides and inadvertent production	1915-present	-	≈0.1	-	30
hexachloro-cyclohexane (HCH)	insecticide	1945-present	-	≈0.4	≈1[c]	22,23
aldrin/dieldrin	insecticide	1950-present	-	0.11	0.04[d]	19

[a] based on reported annual usage in Mexico, South America and India[19]

[b] based on use in Mexico and Central America[19]

[c] estimated from reported annual use of 0.06 Mtonnes in India and China during the 1970's and 1980's

[d] estimated from reported annual use of 0.001 Mtonnes of aldrin in Central America and Mexico in the period 1978 to 1988

Table 2 Physicochemical properties of selected organochlorine compounds and PAHs[a]

Compound	Henry's Law Constant Pa(mol m^{-3})		vapor pressure over liquid (25°C) (KPa)	water solubility[b] (ug l^{-1} @ 25°C)
	freshwater(°C)	seawater(-2°C)		
organochlorines				
lindane	0.32 (25)	0.03	5.5x10^{-2}	2000-12000
α-HCH	0.55 (22-25)	0.10	2.7x10^{-1}	1200-2000
toxaphene	0.61 (20)	0.09		300
hexachloro-benzene	83.0 (25)	7.8	1.3x10^{-1}	<20
α-endosulfan	1.09 (25)	0.10	6.1x10^{-3}	150-600
dieldrin	1.12 (20)	0.125	1.0x10^{-2}	186-200
chlordane	4.92 (25)	0.47	5.7x10^{-3}	56-1850
DDT	5.3 (25)	0.5	5.1x10^{-4}	<1-25
DDE			3.3x10^{-3}	1-140
2,4,5,2',4',5'-PCB	42.9 (25)	4.1		
PAHs				
pyrene	1.2 (25)	0.11	1.4x10^{-2}	
phenanthrene	4.0 (25)	0.38	1.0x10^{-1}	1000-1800[c]
napthalene	43 (25)	4.1	3.8x10^{1}	26000[c]

a modified from Barrie et al.[13]
b from Arctic Laboratories[67]
c from CCREM[21]

Table 3   Comparison of 1985/86 season concentrations for
           selected chlorohydrocarbons in Agassiz Ice Cap as
           measured in 1986 and again in 1987

compound	concentration (pg l^{-1}) (mean±standard deviation)		
	1986 (n=5)	1987 (n=3)	residue (%)
lindane	4080±909	128±43	3
α-HCH	6576±1441	497±90	8
heptachlor epoxide	358±55	47±15	13
chlordane (cis + trans)	760±89	40±26	5
dieldrin	1346±238	213±75	16
α-endosulfan	1094±156	ND	nil
Ttl. PCB	972±344	648±118	67

ND = not detectable

Table 4 Selected Arctic (north of 60°N) air concentration data (ranges except where noted as arithmetic means) from published literature (n = number of samples, α-HCH = α-hexachlorocyclohexane, HCB = hexachlorobenzene, ΣPCB = total PCB as reported by individual references, ΣDDT = DDT + DDE, ND = not detectable)

Location	Time	n	α-HCH	lindane	HCB	ΣPCB	ΣDDT	Ref
					(pg m^{-3})			
Norway								
Bear Island	Fall'80	5	217-516	15-42	29-45			52
	Winter'81	5	185-270	12-16	59-188			
Spitzbergen	Fall'80	4	708-1290	25-50	50-85			52
	Winter'81	5	161-274	13-17	71-95			
Hopen	Fall'82	11	250-1500	ND-6	100-230			53,54
	Winter'83	11	200-750	20-90	90-210			
Norway								
Ny Alesund	Fall'82/83	@	407-1416	0.1-67	75-227			54
Winter/spring'83/84		@	121-787	12-102	29-389			
Summer'84		@	260-774	24-100	20-201			
Bjornoya	Fall'82/83	@	277-1550	0.1-32	78-200			54
Winter/spring'83		@	110-469	23-80	87-201			
Summer'84		@	38-305	5-41	42-149			
Sweden	'84/85	#				≈20-140	≈10-120	55
Canada								
Ice Island	Spring'86	6	451[*]	70[*]	73[*]	<2[*]	2[*]	48
(81°N,97°W)	Summer'86	4	253[*]	17[*]	63[*]	<9[*]	<1[*]	
	Summer'86	7	417-731	24-37	131-233	9-20	0.5-1.4	40
	Summer'87	6	283-377	36-53	44-155	15-28	0.8-4.3	
Alert	Feb-April'88	5	100-270	15-31	100-140	29-60	0.6-3.5	56
Bering &	'88	17	140-410	52-84	120-270			57
Chukchi Sea								

@ number of samples is relatively large and is stated to be based on 48 or 72 hour sampling over sampling periods each year of about 30 days

sampled over a period of one year but number of samples not indicated

* mean of the indicated number of samples

Table 5   Concentrations of selected PAHs in the arctic
           atmosphere expressed as ranges for the period of
           samples (pg m^{-3})

| | Canada | Alaska | | | |
	Alert Feb-April 1988	Barrow March 1979	Barrow August 1979	Narwahl I. April 1979	Fairbanks Nov 1976 to Apr 1977
n	5	3	5	2	21
phenanthrene	6.6-130	80-170	4-30	4-420	100-21,600
2-methyl-phenanthrene	0.16-3.2	NA	NA	NA	NA
fluor-anthrene	7.7-74	120-520	8-55	23-95	500-29,000
pyrene	1.9-50	50-540	5-72	150	400-39,800
benzo(a)-anthracene	0.75-8.3	NA	NA	NA	NA
chrysene	4.0-24	40-170	ND-15	ND	600-44,700
benzo(b) fluoranthene	6.4-23	NA	NA	NA	NA
benzo(e) pyrene	1.8-8.0	120-370	ND	ND	300-20,700*
benzo(a) pyrene	1.0-3.4	7-44	ND-35	ND	
benzo(ghi) perylene	1.2-8.6	70-74	ND-40	ND	2900-154,700
reference	56	59	59	59	61

* sum of benzo(e)pyrene and benzo(a)pyrene
NA = not analysed
ND = not detected

Table 6   Comparison of published contaminant deposition for northern Sweden and Moscow with Canadian Arctic data (μg m^{-2} yr^{-1})

compound	Moscow[a]	Sweden			Arctic[d]
	1978-79	1972-73[b]	1974-76[c]	1984-85[b]	1986-87
alpha-HCH	14				0.02-9.1
lindane	12				0.01-2.0
DDT	52[e]	≈1.2-13.2[f]		≈0.1-8.79[f]	0.01-3.0[g]
Ttl. PCB		≈1.2-16.6	24-60	≈24-154	0.04-0.6

[a] from the Moscow area as reported by Bobobnikova and Dibtseva[62]
[b] estimated from figures published by Larsson and Okla[55] for stations north of 60°N only. Actual data were reported for a monthly time period and these data have been multiplied by 12 to provide a comparable annual value.
[c] from Lunde et al.[63] only for stations north of 60°N
[d] from Barrie et al.[13] and determined from annual snow pack samples representing September to May and here assumed to represent the annual deposition
[e] sum of p,p'-DDT, o,p'-DDT, p,p'-DDD and p,p'-DDE
[f] sum of p,p'-DDT and p,p'-DDE
[g] includes only 1987 data, sum of p,p'-DDE, p,p'-DDD, o,p'-DDT and p,p'-DDT

Table 7   Summary of pesticide snow chemistry data collected in 1979 by McNeely and Gummer[66] from east-central Ellesmere Island compared with selected data from Gregor and Gummer[65] for Alexandra Fiord (1986), and the surface snow layers from Agassiz Ice Cap in 1986 and 1987

pesticide	1979 range (ng l^{-1})	confirmed detections[a]	unconfirmed detections[b]	Alexandra Fiord[c] (ng l^{-1})	Agassiz L-1[d] (ng l^{-1})	Agassiz L+1[c] (ng l^{-1})
α-HCH	<1 - 18	21	5	0.43	6.7	19.1
lindane	<1 - 8	10	5	0.22	4.4	6.8
chlordane	<1 - 4	3	7	0.17	0.74	0.23
DDT[e]	<1 - 6	4	10	0.07	0.01	0.13
α-endo sulfan	<1 - 2	1	2	0.24	1.1	1.1
dieldrin	<2 - 4	5	12	0.83	1.4	1.0

[a] Includes both detections above the detection limit and below the detection limit (i.e. not quantified) but confirmed to be present by MID-GC/MS

[b] Indicates instrument responses below the detection limit but of insufficient concentration to permit confirmation by MID-GC/MS

[c] Single sample

[d] Represents the mean of 5 samples from this layer

[e] sum of p,p'-DDD, p,p'-DDE, o,p'-DDT and p,p'-DDT

Table 8 Comparison of 1987 Ice Island samples for
organochlorine residues as reported by Arctic
Laboratories[67] with results from Gregor and Gummer[65]
based on 3 replicates for each laboratory

| | Concentration (pg l^{-1}) in melted snow | |
| | Gregor & Gummer (mean±1SD) | Arctic Laboratories (mean±1SD) |
compound		
Aroclor (1254)[a]	129±37	185±160
hexachlorobenzene	11±6	12±3
heptachlor	<3[b]	6±3
aldrin	Int	0.5±0.5
o,p'-DDE	ND	2.8±2.4
p,p'-DDE	ND	20±14
Mirex	ND	<4.0
trans-chlordane	8±1	20±15
cis-chlordane	<12[c]	25±18
p,p'-DDT	23±19	<47±26
heptachlor epoxide	30±11	46±26
alpha-endosulfan	21±1	97±58
dieldrin	54±5	<140±55
endrin	spike	27±11
methoxychlor	55±35	37±26

SD = standard deviation
Int= interference on chromatograms
ND = not detectable
[a] For this comparison, the PCB data from Gregor and Gummer
are reported as Aroclor 1254 whereas in all other Gregor
and Gummer references in this report PCBs are reported as
the 1:1:1 mixture of Aroclor 1242, 1254 and 1260.
[b] One of the three samples was ND. Therefore the
concentration is reported as less than the maximum
concentration observed with no standard deviation.
[c] Two of the three samples were ND. Therefore the
concentration is reported as less than the concentration
of the single value with no standard deviation.

Table 9 Comparison of 1987 Ice Island snow samples for hexachlorcyclohexane (HCH) residues among Gregor and Gummer[65], Arctic Laboratories[67] and University of South Carolina (USC) as reported by Arctic Laboratories

	Concentration (pg L^{-1}) in melted snow		
	Gregor & Gummer[a] (mean±1SD)	Arctic Labs[b] (mean±1SD)	USC[c] (mean±1SD)
α-HCH	143±13	290±130	315±25
lindane	83±9	192±96	365±15

[a] Based on 3 samples collected June 6, 1988
[b] Based on 3 samples collected June 8,9 and 15, 1988
[c] Based on 2 samples collected June 7 and 9, 1988

Table 10   Comparison of selected organochlorine pesticide concentrations (pg l^{-1}) in arctic lakes and the Mackenzie River with rivers from northern Ontario and Quebec and Lakes Superior and Ontario in central Canada

	Sample depth (m)	Number of Samples	α-HCH	lindane	HCB	α-HCH/ lindane	reference
L. Hazen	2	3	590	170	90	3.5	70,71
L. Hazen	35	3	340	100	120	3.4	70,71
Sophia L.	2	2	13,620	1,740	140	7.8	70,71
Amituk L.	2	1	3,270	450	50	7.3	70,71
Amituk L.	20	1	3,350	420	70	8.0	70,71
Amituk L.	39	1	2,220	210	30	10.6	70,71
Mackenzie R. at delta	NA		3,000– 4,900	400– 500	NA		67
N. Ontario Rivers	NA	60	5,000– 8,400	400– 1,500	100– 600	7.1[*]	72
N. Quebec rivers Hudson Bay	NA	87	8,000	1,100	ND	7.6[*]	73
N. Quebec rivers Ungava Bay	NA	87	5,800	800	ND	7.3[*]	73
L. Superior	1	19	5700– 11,000	800– 1,430	20–40	7.4[*]	74
L. Ontario	1	31	1,170– 5,920	310– 2,280	20– 110	3.1[*]	74

[*]Ratio has been determined from means as reported by respective authors
NA = not applicable
ND = not detected

Table 11   Median and range of concentrations and
            corresponding winter season deposition of
            trace organic compounds from Canadian Arctic
            snow chemistry surveys in 1986 and 1987[13,70]

| compound | 1986 (n=12) | | 1987 (n=15) | |
	concentration (pg l^{-1})	deposition (ng m^{-2} yr^{-1})	concentration (pg l^{-1})	deposition (ng m^{-2} yr^{-1})
Ttl. PCB	745 (20-1760)	73	907 (257-1773)	95
alpha-HCH	3185 (430-8120)	285	910 (143-42700)	95
lindane	1160 (220-4446)	104	700 (83-10500)	73
heptachlor epoxide	97 (ND-362)	10	117 (ND-1600)	16
alpha-endosulfan	345 (100-1094)	37	175 (ND-4880)	24
dieldrin	540 (205-1402)	58	400 (ND-4420)	56
HCB	28 (10-70)	3	16 (ND-104)	2
DDT[a]	18 (10-50)	2	93 (ND-1380)	8
CBZ[b]	NA	NA	240 (ND-915)	30
PAH[c]	NA	NA	19000 (190-149000)	4246

a DDT is the sum of p,p'-DDD, p,p'-DDE, o,p'-DDT and p,p'-DDT
b sum of all detected chlorobenzenes excluding HCB
c sum of all detected PAHs
NA = not analyzed
ND = not detectable
HCB = hexachlorobenzene

Table 12 Comparison of annual contaminant deposition in precipitation in non-arctic Canada[84] with Canadian arctic (September to May)[70] deposition estimated from concentrations in snowpack (μg m^{-2} yr^{-1})

compound	Lake Superior[b]	Central Ontario (1984)	North Saskat-chewan[c]	South Alberta (1985)	South B.C.[d]	Arctic[e]
α-HCH	3.9-11	14	2.1-6.5	3.3	22-36	.01-2.3
lindane	2.0-3.7	7.3	.39-1.9	1.3	4.8-6.2	<.01-1.0
dieldrin	.15-.50	.30	.01-.12	.022	.03-.34	0.0-.25
DDT[a]	.08	.02	.002-.01	.007	.03	<.01-.2[f]
Ttl. PCBs	1.2-4.9	1.6	1.5-1.7	1.2	1.0	.01-.3
HCB	.04-.05	.08	.003		.05	<.01

a Strachan[84] reported only p,p'-DDE as opposed to p,p'-DDD, p,p'-DDE, o,p'-DDT plus p,p'-DDT for the Arctic
b The Lake Superior data are from 1981, 1983 and 1984.
c The station located at Cree Lake in north Saskatchewan included data collected in 1984 and 1985
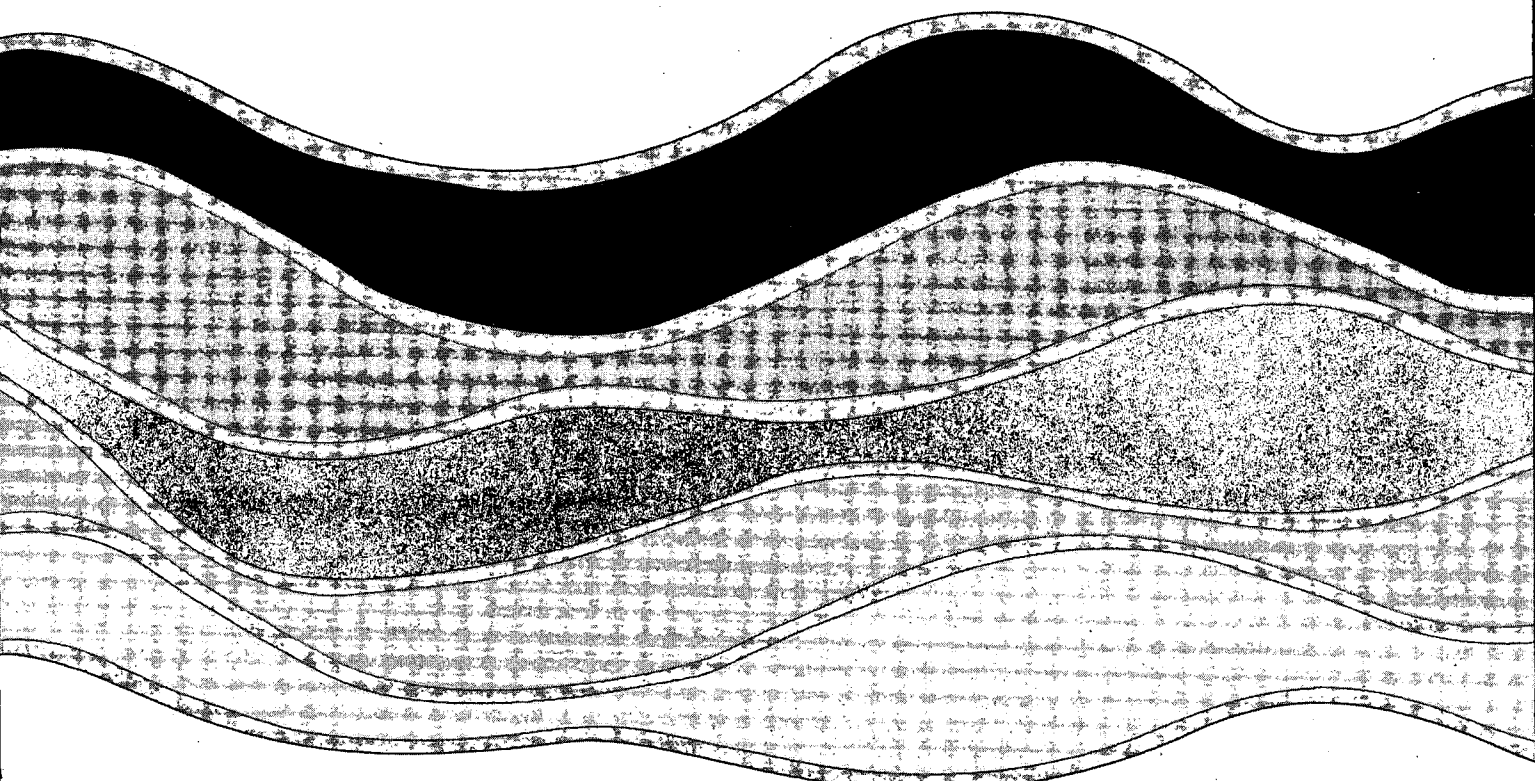d The station at Kanaka Creek in southern B.C. included data from 1985 and 1986
e The Arctic data are ranges for the two years, 1986 and 1987 for all sites except Alexandra Fiord
f Represents the range of loadings for 1987 sites only