polybrominated diphenyl PolyChlorit

clohexane aphene

ethers nated Biphenyls furans

Persistent Organic Pollutants

Stiff and lean after six months curled in a den, a female polar bear squeezes herself out of her winter home. Two small cubs emerge tentatively at her heel for their first view of the world beyond a snow cave. Entirely dependent on their mother, the cubs follow obediently. Having used up most of her fat stores, the female scans the sea ice below and ponders a meal of seal blubber. But her cubs are not yet ready to travel, and her milk will have to sustain them for some time to come. The milk is rich and nourishing but today it also harbors a threat. The seals the mother has feasted on in the past, and will need again soon, are tainted by chemicals from lands far beyond her sea-ice domain. The chemicals that bind to the fat of the seals have accumulated in her own fat stores. Unwittingly, the mother passes the toxins to her young in her fat-rich milk, with effects that are still unclear.

This chapter examines organic chemicals that can affect the health of animals and people, especially those substances that accumulate in Arctic food webs and that resist degradation. These are often called persistent organic pollutants, or POPs. A review of known toxic effects and environmental levels of POPs forms the basis for evaluating whether Arctic wildlife are affected by current levels of contamination. A summary of sources and pathways indicates where the contaminants come from. Many measurements of organic contaminants have been made because of concern about high intake by people, and the human health aspects of these substances are discussed in the chapter *Pollution and Human Health*.

72 Persistent Organic Pollutants

Persistent organic pollutants: a background

In 1945, the booming chemical industry launched a new, effective tool for dealing with insect pests: DDT. It held great promise, including the hope of saving crops and of eradicating disease-carrying insects. Twenty years later, DDT and other, similar chemicals had indeed benefited agriculture and relieved some of the problems associated with insects in many areas of the world. The price of these gains, however, was becoming increasingly clear: DDT is toxic to many more organisms than those it was intended to kill. In particular, birds of prey had trouble reproducing, and their populations declined in many polluted areas of the world.

As early as 1970, when it was detected in the blubber of ringed seals, it was evident that DDT was present in the Arctic. By the mid-1970s, researchers had documented the presence of DDT and other pesticides in beluga, polar bear, and fish. Moreover, birds of prey declined in northern areas that were thought to be uncontaminated.

In addition to pesticides, most analyses in animals also found traces of an industrial oil made of compounds known as PCBs. By 1980, there was evidence that some of these contaminants had reached the Arctic via long-range transport. In the late 1980s, it became clear that human mother's milk at Broughton Island in the Northwest Territories in Canada contained enough PCBs to cause concern about effects on human health. The most likely source was the food the women had eaten.

DDT and a number of closely-related pesticides have been banned for two decades in the circumpolar countries, but long-range transport makes the issue a global one. While PCBs are no longer manufactured in any of the circumpolar countries, they are still in use in many closed systems, and leaks and disposal of old stores remain a problem. New chemicals, with unknown effects on Arctic animals, are also entering the scene.

The biological effects of POPs

Organic contaminants in the Arctic environment share many characteristics that make them especially insidious for people and wildlife.

POPs are stored in fat and are persistent

A common characteristic of most synthetic organic chemicals found in Arctic animals is that they break down very slowly. This persistence in the environment allows them to accumulate in animals, and to pass through the food web. Most of the persistent organic pollutants are also fat-soluble. They thus accumulate in the fatty tissues of animals. Storing energy as fat is crucial for survival in cold environments, and fat is therefore important in the diets of both people and animals, which also increases the intake of these pollutants.

The combined characteristics of being fatsoluble and persistent make biomagnification a major concern. Biomagnification is the increase in contaminant load as predators take on the chemicals eaten by their prey, thus further concentrating the toxic material at each successive level of the food web. For a detailed discussion of this process, see the chapter *Polar Ecology*. Indeed, the highest levels of persistent contaminants are usually found in top predators. Studies of species at different levels of an Arctic marine food web show that each step can mean a several-fold increase in body burdens of organic contaminants.

A broad attack on reproduction

Many of the most visible effects of POPs on animals are related to the ability to conceive and raise young. For example, the early declines in birds of prey were caused by thinning of their eggshells, which made it impossible for the birds to hatch their chicks successfully. POPs may also be directly toxic to developing chicks, killing them in the egg. A more subtle effect is seen in adult birds, when normal mating behavior is impaired.

The effects of POPs on mammals are well documented in polluted areas such as the Baltic Sea. Malformations in reproductive organs, fewer young, or even complete failure to reproduce are some of the detrimental signs of high contaminant levels.

One of the underlying causes of failure to reproduce is that some of the chemicals interfere with sex hormones. Such hormone disrupters can mimic or block hormones because they are similar enough in structure to fit into the body's biochemical receptors. Contaminants that block the estrogen receptor can inhibit the growth of the reproductive tract and the mammary glands, which mammals require to maintain pregnancy. In fish, the same receptor stimulates the production of a precursor to egg yolk. Under normal conditions, the yolk precursor is only present in females, but in contaminated waters it is also found in males. Some of these measurable biological effects are now used as sensitive tests to determine whether a specific compound is a hormone disrupter.

Sex hormones are important for the normal sexual development of young animals. In polluted temperate environments, high levels of hormone disrupters have been connected to malformations in reproductive organs, change of sex in some species, and abnormal mating behavior.

The immune system is very sensitive

One of the most sensitive targets for organic contaminants may be the immune system, the body's primary defense against disease. The thymus, which normally produces antibodies to fight infectious agents, can waste away and cease to function. POPs also limit cell-mediated immunity, the branch of the immune system that fights cancer cells and parasites. There are signs that animals with a high load of contaminants are more susceptible to infections.

Liver enzymes are tell-tale signs of intoxication

In the body, many toxic chemicals are converted into less toxic substances that can be excreted in urine or feces. The liver does most of this detoxification, and many organic contaminants stimulate the production of specific liver detoxification enzymes. Measurements of these enzymes are now used as biological indicators of the load of contaminants in an animal.

Stimulating the detoxification enzymes is not a problem in and of itself. However, the same enzymes are also responsible for breaking down hormones. Therefore, contaminants can indirectly increase the breakdown of hormones, potentially influencing critical hormonedependent functions, such as reproduction.

The liver-enzyme systems are different in different species, which affects their respective abilities to process contaminants. This can explain why some animals can get rid of a specific substance, while other animals accumulate it. Fish completely lack one of these enzyme systems, which makes them a carrier of many POPs in the food web.

Increased risk of tumors

Several POPs are suspected of being responsible for increased rates of tumors in wildlife in polluted areas. There are two ways by which a contaminant can increase the risk of cancer. The first is a mutation of hereditary material in the cells, the DNA, which makes the cell lose control of its growth. The second allows a cell damaged in this way to turn into a tumor. Contaminants implicated in the latter process are called promoters, and this group includes most POPs. They do not cause cancer by themselves, but can act together with DNA-damaging chemicals.

Sensitive glands and vitamin A

Several hormone-producing glands in the body are sensitive to POPs. One is the thyroid, which is responsible for the balance of thyroid hormones. A chemical disruption of the thyroid can lead to goiter and to changes in metabolism that affect growth and reproduction. Some POPs may affect the breakdown of Vitamin A. Imbalance in Vitamin A can suppress the immune system, increase susceptibility to cancer, cause skin lesions, and disrupt reproduction, growth, and development.

Many POPs can damage the adrenal gland. Seals in the Baltic Sea suffer from a series of diseases that are connected to adrenal gland effects. These diseases include malformations of the uterus, which is one of the reasons Baltic seals have had trouble reproducing.

Porphyria

Some POPs disturb production of the pigment in red blood cells, which in severe cases leads to the disease porphyria. Symptoms include skin damage after exposure to light as well as damage to the nervous system. The biochemical changes associated with porphyria, which are measurable long before symptoms appear, are used as sensitive biological markers of POPs in the environment.

Effect assessments include many uncertainties

Most of our knowledge about the toxicology of organic pollutants comes from studies with laboratory animals, semi-field studies with a few species of wild animals, and studies of the association between contaminant levels and effects in wild animals. This includes information about what levels of specific contaminants can potentially be associated with health effects in the animals. The assessments presented later in this chapter therefore include many uncertainties. For example, the assessments assume that Arctic animals in their natural environment have approximately the same sensitivity as the animals that were used in the toxicological studies. In reality, they could be more sensitive or less so. Thus, when POP levels in Arctic biota reach biological effects thresholds determined from other animals, it should be interpreted as a warning signal rather than as evidence that such effects actually do occur in the Arctic.



The peregrine falcon is a bird of prey that has been hard hit by persistent organic contaminants.

74 Persistent Organic Pollutants

A cast of characters

The term 'organic contaminants' covers a wide range of substances. Some are industrial chemicals whose toxic character is unintentional. Others are byproducts in industrial processes. The third category includes substances that are designed to be toxic, such as pesticides. Many organic contaminants contain chlorine and are also called organochlorines. Most organochlorines discussed in this chapter have no natural sources in the environment.

Industrial chemicals and byproducts

PCBs

The term 'PCBs' refers to a group of chemicals called polychlorinated biphenyls. They were introduced in 1929 and manufactured in the United States, Japan, the former Soviet Union, and eastern and western Europe. They are chemically stable and heat-resistant, and were used world-wide as transformer and capacitor oils, hydraulic and heat-exchange fluids, lubricating and cutting oils, and as plasticizer in joint sealants. Open use is currently banned in all circumpolar countries, but there are still substantial amounts in large capacitors and transformers. Current use and disposal are often poorly documented.

The toxic effects of PCBs depend on their chemical structure. The most toxic PCBs have a coplanar configuration, which is similar to dioxins (see next subsection). PCBs have a range of toxic effects. The most significant of these may be that they suppress the immune system, making animals more likely to become ill and more likely to die if they are exposed to infections. They can also disturb behavior and reproduction in birds, fish, and mammals. PCBs have contributed to population declines and health problems in fish-eating mammals in polluted areas, including the beluga in the St. Lawrence River estuary, seals in the Baltic Sea, and European otters. The reduced number of fish-eating birds in the Great Lakes region has also been associated with high concentrations of PCBs, especially the dioxin-like coplanar PCBs.

Other sensitive targets for the toxic effects of PCBs include the developing nervous system and liver enzymes. PCBs can also act as a cancer promoter and can cause birth defects.

Dioxins and furans

Dioxins (PCDDs) and furans (PCDFs) are a group of chlorinated chemicals, of which 17 are toxic in minute quantities. They are primarily created in high-temperature processes. Waste incinerators without efficient flue-gas cleaning systems are or have been one of the most significant sources. Wood-burning stoves and the use of leaded fuel add to the load in the atmosphere. Dioxins and furans also enter the environment as byproducts of industrial processes. Metallurgical industries are large sources to the air, while pulp and paper mills that use chlorine in the bleaching process often release contaminated water. A third type of source is as trace contaminants in chlorophenoxy acid herbicides (e.g. Agent Orange), in chlorophenol wood preservatives, and in PCB mixtures (mainly furans).

The toxic mechanism for dioxins and furans is the same as for the coplanar PCBs, but some are considerably more potent. Effects include disturbed reproduction, a suppressed immune system, and an increased risk of cancer.

Hexachlorobenzene

Hexachlorobenzene (HCB) is a byproduct in the production of chlorine gas and chlorinated compounds, including several pesticides. It is emitted to the atmosphere in the flue gas from waste incineration, and is also formed by metallurgical processes. It has had limited use as a pesticide. The major concerns are porphyria and effects on reproduction and on the immune system.

Brominated flame retardants

Many substances that are used as flame retardants have chemical properties that are similar to PCBs. Polybrominated diphenyl ethers can leach out of flame-retardant-treated textiles, electrical equipment, building material, and car interiors to cause diffuse contamination of the environment. Knowledge about the toxicity of brominated flame retardants is very limited.

Pesticides

Many of the pesticides found in animals are organic compounds that have several chlorine atoms and are very persistent in the environment. Pesticides are designed to be toxic to their target organisms. Most of them affect the nervous system and the liver, and several interfere with reproduction.

DDT

DDT is a chlorinated organic pesticide, introduced as an insecticide in 1945. Circumpolar countries restricted its use two decades ago, but it is still used in pest control programs in southern Asia, Africa, Central and South America, and Europe (e.g. Italy). In the past, it has been used to control mosquitoes and black flies in the Arctic, and there may still be some use in remote areas of Siberia, in spite of bans. In Russia, household and institutional insecticides that contain DDT and hexachlorocyclohexane are still in use. DDT enters the region via air and water currents and in migratory animals.

In the environment, DDT is converted to the metabolites DDE and DDD. These are stored in fatty tissues of fish, birds, and mammals. It is the cause of eggshell thinning in birds. DDT and its metabolites also disrupt sex hormones and affect liver enzymes.

Toxaphene

Toxaphene is a complex mixture of polychlorinated bornanes and camphenes. Until the early 1980s it was widely used to fight insects in cotton crops in the United States. Manufacture and use are now banned in the United States, but similar products are still used in some parts of the world, including Mexico and Central America.

It is difficult to measure toxaphene in the environment. The levels in the Arctic are therefore not well studied, despite the fact that toxaphene has been recognized as a persistent organic pollutant of great concern. It may be one of the most abundant pesticides in Arctic wildlife, but limited exposure information and lack of data on toxicological effects make risk assessments difficult.

Toxaphene's major toxic effects are on the nervous system. Fish are extremely sensitive, becoming hyperactive with muscular spasms and losing their equilibrium. In experiments, fish that have been injected with toxaphene at levels measured in the environment and then released into the wild have lower survival rates than uncontaminated fish.

Chlordane

Chlordane is a mixture of compounds that has been used to protect seeds from insects and for termite control. It is no longer used in most circumpolar countries, but is still produced for export in the United States. One of the components of chlordane (heptachlor) breaks down to a compound (heptachlor epoxide) that is carcinogenic, and has been found in the Arctic environment. Oxychlordane is another toxic metabolite. Chlordane affects reproduction and the immune system.

Hexachlorocyclohexane/lindane

Hexachlorocyclohexane (HCH) has a number of isomers, of which alpha, beta, and gamma are usually those found in environmental samples. The gamma isomer, also called lindane, is the most potent as an insecticide. Lindane is still used in North America, Japan, and Europe to treat seed and in Europe for other purposes. Other forms of the compound were banned in the late 1970s. In China and some other countries, technical mixtures of different hexachlorocyclohexanes are still used to treat hardwood logs and lumber, seeds, vegetables and fruit, and buildings. Lindane is a neurotoxin. It also adversely affects reproduction, the liver, and the immune system, and is a cancer promoter.

Dieldrin

Dieldrin is a soil insecticide. It is no longer used in circumpolar countries. Manufacture in Europe, especially for export to developing countries, continued until the late 1980s. Dieldrin is extremely persistent in soil and in biota. It is the strongest carcinogen of the organochlorine pesticides.

Mirex was used as an insecticide and fire retardant, mainly in the United States and Canada, until 1978. Mirex is highly fat-soluble and persistent in the environment. It is implicated in cancer and reproductive effects seen in laboratory animals.

Organotins

Mirex

Tributyltin (TBT) is an organic compound containing the metal tin. It is used as a broadspectrum killer of algae, fungi, insects, and mites. Since the 1960s, TBT has mainly been used as a marine antifouling agent. It leaches into the water from surface coatings on boats, aquaculture pens, moorings, and industrial cooling pipes. Other sources are boat repair yards, marinas, and municipal wastewater and sewage sludge. Several countries have banned the use of TBT for small boats, but international regulations still allow restricted use.

TBT breaks down rapidly at the sea surface, with a half-life of only a few days. However, in sediments and especially under cold conditions, it can remain for much longer, with a half-life of over two years. Sediment can thus remain a source long after any bans on the use of TBT have taken effect.

TBT is one of the most toxic substances that has been introduced to natural waters. A few nanograms per liter are enough to affect dogwhelk snails. Chronic effects in oysters, mussels, and crustaceans are observed at exposure levels of less than 1 microgram per liter. It is moderately fat-soluble and can thus bioaccumulate. It is a hormone disrupter that affects reproduction.

Less persistent pesticides

A large number of chlorinated pesticides are still used in circumpolar countries and elsewhere in the world. These are less persistent than their predecessors and do not accumulate or biomagnify to any great extent. Most of them have short half-lives in water, soil, and sediment. Nevertheless, many of these compounds have been found in the Arctic, especially in air, seawater, and sea ice. This reflects both their large-scale use - in some cases over a million kilograms per year - and their transport by winds in a manner similar to many of the banned pesticides. Little is known about whether low light and low temperatures may make them more persistent in the polar environment than in temperate climates.

Examples of currently used chlorinated pesticides that have been found in air, lakewater, fish, seawater, snow, and plants in the Arctic are atrazine, endosulfan, chlorpyrifos, chlorothalonil, tetra- and pentachlorophenol, and methoxychlor. Non-chlorinated pesticides include the organophosphate terbufos, the phenylamide herbicide metolachlor, and the dinitroaniline herbicide trifluralin. 75 Persistent Organic Pollutants

Sources and pathways

The picture of sources and movements of POPs to the Arctic is complex. Most POPs do not move directly from their source to their destination, but cycle around in the global environment as was described for multi-hop compounds in the chapter *Physical Pathways of Contaminants Transport*. The box below gives a further explanation of some of the peculiarities of these contaminants.

A global flux among particles, water, and air

The transport and fate of organic contaminants depend to a large extent on the physical-chemical properties of each chemical. One of the key characteristics is the environment that a particular molecule prefers. A few compounds are slightly soluble in water, with hexachlorocyclohexane as a prime example. However, most organic contaminants have an affinity for particles and fatty matrices. Depending on temperature, some of the molecules will favor the gas phase.

There is an equilibrium between these abiotic phases, so that some of each contaminant is in the air, some in the water, and some is associated with particles. Changes in temperature can alter the equilibrium, and contaminants will move from one phase to another. For example, if the temperature rises, POPs from the soil, from the snow pack, or from the surface water of the ocean will move into the atmosphere. The break-up of ice in the spring also allows for exchange between water and air. Meltwater washing over the ground and winds picking up dirt are physical processes that can move the contaminants.

During the cold winter months, most POPs adhere to particles. In the atmosphere, they get carried to the Arctic on haze aerosols that follow major wind currents. In summer, when the air warms to 0° C or more, some contaminants will be transported as gases.

The further fate of the particles will depend on when snow and rain clean the air, whether contaminants become fixed in the soil or washed away by snowmelt, and whether contaminant-laden sediment particles end up in rivers that reach the sea.

Global pool of contaminants is the major source

The major source of most persistent organic chemicals to the Arctic today is the residue of widespread contamination of the global environment, the sum of past uses; see table below.

Modeling the pathways and transport of POPs gives an indication of how long this global source will continue to contaminate the Arctic environment. Such a model for DDT shows that it will be with us for many decades more. After a complete global ban, about 10 percent of the atmospheric load, 30 percent of the soil load, and only 1-2 percent of the ocean load will

Usage of selected pesticides for various periods of time.

Pesticide	Usage, tonnes	Period
Accounted for		
DDT	1500 000	1948-1993
Technical HCH	$550000\\40000\\29000$	1948-1993 1980 1990
Technical lindane	$720000\\5900\\4000$	1948-1993 1980 1990
alpha-HCH	$28000\20400$	1980 1990
gamma-HCH	$11900 \\ 8400$	1980 1990
Toxaphene	450 000	1948-1993
Interpolated		
DDT	2600 000 990 000	1950-1993 1970-1993
Toxaphene	1330 000 670 000	1950-1993 1970-1993

disappear each decade. Such models are still in their infancy, but nevertheless provide an indication of potential future environmental burdens.

The models also show that the propensity of different compounds to reach the Arctic via the atmosphere differs with their chemical characteristics. Some, such as hexachlorocyclohexane, are more likely to travel all the way, whereas others, such as dioxins, only reach the far north to a limited extent. In general, only part of the global burden will reach the Arctic, but this fraction can still create significant problems because of the tendency of organochlorine contaminants to concentrate in the fat of Arctic animals and biomagnify in the food web. This can lead to high levels in toplevel predators, including people. Cold temperatures in the Arctic also seem to create a sink for certain persistent organic pollutants, which may in some cases result in POP levels that are higher in the Arctic than in the source regions. This phenomenon is often referred to as the cold-condensation effect.

Some pesticides are still produced and used

The concern about long-range transport of POPs has led to several political initiatives to limit the production and use of these chemicals. To some extent these efforts have been successful, but it is difficult to get an accurate picture of the current global situation. International statistics are lacking and some industries are reluctant to release production numbers. An inventory made by the Governing Council of the United Nations Environment Programme, as part of a Global Action Plan, shows that DDT is still produced to control disease-carrying insects, and that it is also misused for other purposes. Chlordane is still produced for ant and termite control, while production of aldrin, dieldrin, and endrin has stopped. Mirex also appears to be out of production, as does hexachlorobenzene for use as a pesticide.

The table below gives a picture of where the pesticide hexachlorocyclohexane is still used including lindane, which is still produced and used in Europe. Available data suggest that

Estimated annual usage of alpha-HCH and gamma-HCH in 1990 for the top-consuming countries.

	<u>0</u>	
Country	Usage, tonnes	
alpha-HCH		
India	19880	
Mexico	183	
Ukraine	168	
gamma-HCH		
India	4 260	
France	1 860	
Italy	600	
Nigeria	397	
Canada	150	
United States	114	
China	100	
Spain	96	

India is the major user of lindane and technical hexachlorocyclohexane. Other countries may also be significant sources.

Pesticides have been used for insect control in the Arctic

Pesticide use in the Arctic has mainly been for insect control, which has probably occurred in all circumpolar countries. There is, for example, anecdotal evidence of spraying around military installations.

In the Yukon Territory of Canada, the use of pesticides has been well documented. Starting in 1948, DDT was applied directly into the Yukon River to control mosquitoes and black flies. Several other organochlorine pesticides were also tested. Starting in 1949, the Canadian air force sprayed DDT mixed with fuel oil around its bases at Whitehorse and at Watson Lake. In 1964, ground fogging and the use of capsules of DDT and lindane replaced aerial spraying. However, as a result of pressure from local populations, the aerial spraying of DDT was resumed and continued until 1969, when DDT was replaced by other insecticides. Drums with leftover DDT have been dumped at community landfills and on the shore of the Yukon River.

DDT was probably used for insect control around military sites in Alaska as well. In remote areas of Siberia, there may still be some use of DDT for insect control, in spite of bans. As far as can be determined, no organochlorine pesticides have been used in Arctic Norway or Sweden.

PCBs are still around in buildings and landfills

Several compounds that are no longer manufactured are nevertheless important when considering the sources of Arctic contamination. To identify environmental problems that must be dealt with in order to minimize future emissions, AMAP has tried to make a qualitative survey of sources within the circumpolar countries.

PCB mixtures have been banned from open use in all circumpolar countries, but in some countries are still allowed in closed systems that existed prior to the ban. Unintentional open use still occurs. For example, in Norway 650 tonnes are contained in products that are still around, mostly window sealing compounds and lighting equipment. Also, 400 to 600 tonnes of technical PCBs have been disposed of in such a way that they may eventually be released into the environment. This situation is probably not unique to Norway.

Several products used in buildings are leaking PCBs into the environment. In Sweden, joint sealant that was used in connecting prefabricated building elements from 1950 to 1972 contains up to 20 percent PCBs as plasticizer. This is the equivalent of 100 to 500 tonnes of PCBs in unintended open use in Sweden. Elevated levels of PCBs in the air and soil outside these buildings have been documented. Some of the PCBs have probably ended up in landfills. The joint sealant was marketed internationally and used in other countries as well. Other sources of PCBs in old buildings are floor paints.

Abandoned military sites are local PCB sources

Prior to the mid-to-late 1970s, PCBs were widely used in transformers, capacitors, and other electrical equipment. Leaks or improper disposal are known from the past, occurring, for example, at military radar stations.

In Canada, this use and the consequent local contamination have been well documented in a study of Distant Early Warning (DEW) Line Sites built to detect missiles and bombers heading toward North America. The DEW Line consisted of 63 stations across Alaska, Canada, and Greenland, roughly following the 66th parallel. At the time the stations were in use, no one was concerned about dumping PCB fluids in the environment. Of an estimated 30 tonnes of PCBs used in the stations, an unknown amount has ended up in their landfills.

Environment Canada and the Canadian Department of National Defense have attempted to clean up some of the stations by taking care of waste-containing drums, old equipment, and contaminated soil. However, many sites are still contaminated with PCBs, at levels ranging from 1 to 10 000 nanograms per gram soil. These numbers can be compared to remote background areas with 0.9 nanograms PCBs per gram soil. As is apparent from measurements in soil and plants, the severely contaminated soils have served as a source to nearby areas.

Contamination at radar stations is probably not unique to Canada. For example, some Alaskan installations have hazardous waste sites, measurements near Thule in Greenland show elevated levels of PCBs, and the Norwegians have documented elevated PCB levels at dump sites on Jan Mayen and Svalbard. In Russia, no studies to look for possible local contamination by PCBs or other POPs have been reported.

Power stations, oil platforms, mines, and trains are potential PCB sources

Electrical equipment is also used in power stations, and old equipment is still a potential source of PCBs. For example, in Canada, some temporary wartime power stations and other installations in the Yukon Territory are known to have contaminated the soil locally. An inventory of PCB contamination in the Yukon Territory has also identified the use of PCBcontaminated oil to control dust on the streets of Whitehorse. **78** Persistent Organic Pollutants

Other historical sources include electric trains, which may have spread PCBs along their tracks from leaking transformers, and hydraulic and drilling fluids from mines and oil platforms. When a fire destroyed the British oil platform *Piper Alpha* in 1988, five tonnes of PCBs were released into the North Sea. From Svalbard, there are signs that PCBs from local sources have spread into nearby fjords.

Arctic smelters and pulp mills have contributed dioxins

HCH concentrations in air, pg/m³

Emissions of dioxins and furans are associated with industrial activities that occur in the Arctic. Known local sources include iron-ore pelleting plants at Malmberget, Kiruna, and Svappavaara in Arctic Sweden, and the smelter Rönnskärsverken by the Bothnian Bay just

Ny-Ålesund, Svalbard, Norway, Apr.-Dec. 1993



gamma-HCH

alpha-HCH

200 150 100 50 0 M Α М J J Α S O N D 200 Alert, Ellesmere Island, Canada, Jan.-Dec, 1993 150 100 50 0 М М J S 0 Ν Α J Α 200 Tagish, Yukon, Canada, Jan.-Dec. 1993 150 100 50 М AMJJ Α ∣ s 0 N D Dunai Island, Lena River Delta, Russia, Mar.-Dec. 1993 100 50 0 М Α MJ J S 0 Ν D Α 100 Heimaey Island, Iceland, Jan.-Dec. 1995 50 0 D

south of the Arctic Circle. In Kirkenes, Norway, local dioxin contamination has been documented in freshwater sediment and in whitefish in a lake near the Syd-Varanger smelter works. These studies show that smelters can be significant local sources and that they can also be important contributors to background levels in the Arctic. The large smelters on the Kola Peninsula, the Vorkuta area in the north Komi Republic, and the Norilsk area are also likely sources of dioxins and furans, although this has not been documented. Other suspected sources include the secondary iron and steel industry, an aluminum industry, and a ferroalloy industry in Arctic Norway, as well as two smelters in Iceland.

Pulp and paper mills located within the Arctic Ocean's drainage area that use elemental chlorine in the bleaching process can contribute to the load of dioxins and furans. For example, studies in the Arkhangelsk area show local contamination along the Severnaya Dvina River and its tributaries. The levels are fairly low, however, and the pulp mills along the river are probably not major sources to the Arctic Ocean. In North America, studies along the Peace-Athabasca River system have shown that pulp mills have in the past released dioxins and furans, some of which have accumulated in the sediments of Great Slave Lake.

Other sources of dioxins and furans in the Arctic include burning wood for heating and waste incineration. Forest fires may be an additional natural source.

Levels in the air, snow, and rain

Air currents are the most important transport routes by which organic contaminants reach the Arctic. Measurements of POPs in air can thus be used to identify source regions. Measurements of organic contaminants in precipitation give important information about how much of the air contamination is scavenged and further transported to terrestrial, freshwater, and marine environments.

Air measurements point to mid-latitude POP-use as source to the Arctic

Hexachlorocyclohexane is the predominant organochlorine in air. The figure left shows concentrations from five different stations. A summer decrease in some measurements is probably due to precipitation, which cleans hexachlorocyclohexane out of the air. Spring peaks may be related to volatilization from the snow pack.

Other organochlorines that occur in substantial amounts in Arctic air include PCBs, toxaphene, and chlordane-related compounds.

Elevated levels of pesticides, especially lindane and chlordane, have been correlated with

Month

Hexachlorocyclohexane levels in air showing concentrations of alpha-HCH (height of light green bar) and gamma-HCH (height of dark green bar). long-range transport from use areas farther south. These results, summarized in the map at top right, demonstrate that current and past use of organochlorines in the mid-latitudes of the northern hemisphere is the most likely source to the Arctic environment. The compounds are resistant to environmental degradation and have high enough volatility to continue to cycle.

Russian snow and rain have unexpectedly high levels of PCBs and DDT

Measurements of contaminant levels in snow provide an indication of how much of the airborne material stays in the Arctic, since snow is very effective in scavenging particles from the atmosphere.

Concentrations of DDT and PCBs in snow from the Taimyr Peninsula and Laptev Sea, Russia, in 1995 were about ten times higher than in the Canadian Arctic, with PCB concentrations averaging 10 nanograms per liter. These high levels are especially alarming considering that only seven PCB components were measured; total PCB levels can be much higher. There may be problems with unintentional contamination during sampling, and there is a need to confirm the high values.

The tables show some examples of precipitation measurements.

POP concentrations in Russian samples, ng/liter.

	НСН	DDT	РСВ
Snow, May 1995 Taimyr Peninsula	5.61	2.06	5.3
Precipitation, August 19	94		
Taimyr Peninsula	0.88	3.2	11.9
Laptev Sea	< 0.1	1.1	1.1
Barents Sea	0.67	0.34	4.3

Deposition of POPs to snow in Canada, mg/m²/season.

		НСН	DDT	PCB
Alert	1992/93	0.88	0.05	0.19
Alert	1993/94	0.50	0.05	0.37
Eureka	1991/92	0.78	0.01	0.26
Mould Bay	1993/94	0.25	0.03	0.23
Cape Dorset	1993/94	0.22	0.06	0.51
Dawson City	1993/94	0.19	0.04	0.41
Whitehorse	1993/94	0.12	0.02	0.20
Tagish	1992/93	0.28	0.01	0.24
Tagish	1993/94	0.20	0.04	0.35

Levels in terrestrial environments

The main concern in the terrestrial environment is that contaminants that end up in plants can be carried through the food web, as the plants become food for grazing mammals and birds. Plants, especially perennial mosses and lichens, can also give a good indication of how much of the airborne contaminants are deposited to the surface within the Arctic; see the figure to the right.



Except for locally polluted sites, levels in plants are generally low compared to industrialized areas farther south. PCBs, DDT, and hexachlorocyclohexane dominate. Although toxaphene was measured in only one place, Ellesmere Island, it was the most abundant of all POPs. Even across small geographic areas, there is a gradient from higher concentrations of organochlorines in the south to lower concentrations in the north. This gradient appears, for example, in data from Finland and from Ellesmere Island.

Measurements in soil have mostly been carried out at sites known to be contaminated, such as the Canadian DEW Line sites. These local levels are high enough that the soil can serve as a source of PCBs to the surrounding ecosystem.



Pathways and source regions for POP-contaminated air.

Average concentration of PCBs in moss and lichen.

Caribou and reindeer have low levels of POPs

Caribou and reindeer feed on ground vegetation and can potentially accumulate contaminants from the plants. The major contaminants ending up in the animals are hexachlorobenzene and hexachlorocyclohexane, reflecting the predominance of these contaminants in atmospheric deposition to their grazing areas. The levels seem to be fairly uniform across the Arctic; see the figure below. How-



HCH in caribou/ reindeer liver ng/g wet weight 10-

Λ

Hexachlorocyclohexane levels in caribou/reindeer liver. Russian data are based on one individual per site.

Biomagnification of PCBs from three sites in Canada.



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ever, Russian results from two consecutive years differ considerably. Excluding Russia, mean hexachlorobenzene concentrations in the dataset for Canada and Svalbard range from 0.3 to 3.7 nanograms per gram liver (wet weight) and hexachlorocyclohexane concentrations range from 0.9 to 8 nanograms per gram liver (wet weight) in caribou, which are extremely low levels. For Russia, mean hexachlorobenzene levels range from 0.09 to 7.6 nanograms per gram liver and hexachlorocyclohexane levels from 0.2 to 7.5 nanograms per gram liver, depending on the year.

For DDT and PCBs, the levels in Russian reindeer liver (for both years) are higher than in other areas, which is also true for other ground-feeding animals such as lemming, ptarmigan, and brant.



Levels in North American caribou show a more varied geographic picture for PCBs and DDT than for other compounds. This may reflect a greater influence of contaminants from regional sources in North America relative to global distribution.

There is no information about biological effects of organic contaminants in reindeer/ caribou, but in general, the levels are several orders of magnitude lower than those expected to cause effects.

The levels in caribou have also been used to examine the biomagnification of contaminants in the terrestrial food chain from lichen to caribou to wolf. The diagram at the bottom of this page shows that PCBs accumulate for each step in the chain and thus that biomagnification occurs in the terrestrial environment. A closer look at the different components of PCBs reveals that wolves are able to break down some substances, but that the most persistent remain.

Waterfowl carry contaminants from overwintering areas

Waterfowl and terrestrial game birds and their eggs are food sources for people as well as for birds of prey, such as the peregrine falcon. A study of Canadian waterfowl shows that some of the birds probably carry substantial burdens of contaminants from their overwintering areas farther south. For example, birds from eastern North America have much higher levels than birds from farther west. The traditional overwintering areas for eastern birds are the Great Lakes, the Gulf of Mexico, and the eastern American seaboard, which are all relatively polluted environments. Mirex, which is a typical contaminant of the lower Great Lakes, only shows up in the eastern birds.

In general, the levels in waterfowl are low, in the nanogram per gram range (whole body), but with some exceptions. Birds feeding on fish and mollusks have higher levels than those eating plants, which reflects their higher position in the food web. Also, oldsquaw, pintail, and some individuals of semipalmated plover have PCB levels that exceed the thresholds for reproductive system effects in other birds.

The Canadian peregrine falcon still suffers from eggshell thinning

The peregrine falcon is a predatory bird that has suffered badly from high levels of pesticides in the environment. This includes the populations of the subspecies that breed in the North American Arctic, Falco peregrinus tundrius, which feed on waterfowl and small rodents. Studies in the Canadian Arctic show average PCB levels of 8.3 micrograms per gram and DDE levels of 4.5 micrograms per gram egg.

It is difficult to know whether Arctic populations are recovering as fully as those in

northern temperate areas. The number of birds is naturally low, and large fluctuations in population size make recovery hard to monitor. However, in the early 1990s, contaminant levels in the birds were still high enough to reduce eggshell thickness. In 1991, 28 percent of the clutches showed thinning equal to or greater than the threshold level that has been associated with failure to reproduce.

Since the early 1980s, in spite of bans on DDT in all the circumpolar countries, eggshell quality in this population of tundra peregrines has not improved. Levels of DDE, dieldrin, and heptachlor epoxide were lower in the early 1990s than in the early 1980s, but female peregrines had higher levels of PCBs, four times the maximum value reported in the 1980s. Moreover, chlordane seems to be increasing again in the eggs, which is probably a result of an increased use of this pesticide.

The source of the contaminants in peregrine falcons is the migratory waterfowl on which the peregrines prey. A study at Rankin Inlet showed that levels of PCBs in oldsquaw (long-tailed duck) and pintail were high enough to lead to reproductive effects in peregrine falcons. The threshold levels for DDE were exceeded in oldsquaw, water pipit, and semipalmated plover. These birds are especially important in the diet of female peregrines. Several other contaminants were also detected in the prey, but not at levels high enough to affect peregrine reproduction. Predatory birds that feed mostly on animals resident year-round in the Arctic have much lower levels of contaminants.

The Committee on the Status of Endangered Wildlife in Canada previously considered the tundra peregrine falcon threatened. The status has been changed to vulnerable because the chemical threat did not appear as great as before. These recent results suggest that the threat is still present.

Icelandic gyrfalcon accumulate POPs with age

A study of gyrfalcon in Iceland shows that these birds of prey accumulate organic contaminants over their lifetime. Among birds that were found dead in Iceland, the levels in recently hatched birds were about 100 nanograms DDT and DDE combined per gram muscle. In ten-month-old birds, the concentration was a hundred times greater; in 20-month-old birds, it was a thousand times greater than in the hatchlings. Since most of the birds in the study were young, the average levels in the population could have been even higher than the levels reported here. The gyrfalcons are yearround residents in Iceland but probably feed on migratory birds that are contaminated.

The study also showed that the concentrations of PCBs and DDT were higher in leaner birds than in birds with more fat. Most organic contaminants are normally stored in fat, and using fat reserves may have released enough DDT and PCBs into the birds' vital organs to contribute to their deaths.

Fennoscandian birds of prey are recovering

In Fennoscandia, white-tailed sea eagle, osprey, merlin, peregrine falcon, and eagle owl have all suffered from high concentrations of organic contaminants. From the 1950s to 1970s, eggshell thinning and lowered reproductive capacity led to population declines. For most of the birds, the trend has now reversed and their populations are starting to recover. This recovery coincides with the decline in environmental levels of DDT and PCBs.

While the situation has improved, the problems have not disappeared. Merlin from Alta in Norway still have about 10 percent eggshell thinning on average, and in some cases their DDT levels remain high enough to cause concern about reproductive success. Peregrine falcons also have PCB levels above the lowest that cause reproductive effects in other birds; see diagram on page 87. There is some concern about current contaminant levels in the food of Norwegian white-tailed sea eagles: PCB (diagram, page 88) and DDT levels in a range of fish species from Arctic sites exceed several of the guidelines for protecting fish-eating wildlife.

Studies of a population of peregrine falcons on the Kola Peninsula also conclude that contaminant levels are high. Dioxin was found in concentrations that are associated with embryonic mortality in other bird species.

American mink and marten have low contaminant loads

American mink and marten feed on small mammals and fish throughout the forested region of North America and can potentially accumulate organic contaminants from both terrestrial and freshwater environments. Mink are known to accumulate PCBs, to which they are extremely sensitive. Even levels as low as 72 nanograms per gram food can lead to a failure to reproduce.

The levels of PCBs in American mink and marten in the Arctic are low in most places that have been investigated. The exception is mink from Grand Baleine, Quebec, in eastern Canada, which have PCB levels that are just below the no-effect level for litter size. Also, fish from several lakes have PCB levels that may be high enough to cause reproductive effects if they are eaten by mink.

Scandinavian mink and otter are bouncing back

The American mink was introduced to Scandinavia in the 1940s. At first, the population increased rapidly but then leveled off in the 1960s **82** Persistent Organic Pollutants

and declined in the early 1970s. The chief cause of the decline was most likely high exposure to organic contaminants, particularly PCBs. After a 50-percent reduction in environmental levels of PCBs from 1975 to 1978, the mink population began increasing again.

Otters in the Swedish Arctic are now experiencing a similar recovery. The population declined rapidly from the 1950s to the 1970s, and by 1980 only a few isolated groups survived. From the end of the 1980s and into the 1990s, the northern population has suddenly been increasing again.

By analyzing PCB levels in otter muscle, it has been possible to determine a threshold concentration of contaminants above which an otter population will suffer. Muscle tissue levels above 7.5 to 25 micrograms per gram lipid lead to population declines, whereas concentrations below 7.5 micrograms per gram lipid allow a population to recover. Norwegian otter populations that live along the Arctic coast fall in the low range and have indeed remained constant or even increased slightly. The levels of PCBs may, however, be high enough to affect neurobehavioral development. Levels of dioxins and dioxin-like PCBs in Swedish otter are above those associated with immunosuppression in seals.

Red fox and wolf

The food habits of fox and wolf differ greatly depending on geographic area and time of year. Red fox typically prey on smaller mammals, but also eat insects, reptiles, fish, and berries. Wolf packs often follow and hunt caribou. Red fox and wolf samples from Canada contained measurable levels of POPs, of which PCBs were most prominent. The levels were highest

Hexachlorocyclohexane levels in river water.



in fox, probably reflecting generally higher contamination levels in the northern Quebec environment in which they were studied.

Levels in freshwater environments

The freshwater environment reflects the combined input of contaminants from the air and from runoff. Most of this contamination is the result of long-range transport from temperate industrialized regions. Local contamination is a problem in some lakes in the Arctic.

Some Russian rivers and lakes seem to be very contaminated with POPs

Hexachlorocyclohexane is the main POP detected in river water. The diagram below shows levels of hexachlorocyclohexane in rivers from across the Arctic. Most noteworthy are the exceptionally high levels in some Russian rivers, especially the Ob. A high ratio of gamma-hexachlorocyclohexane to alpha-hexachlorocyclohexane indicates input of the pesticide lindane.

DDT, too, was detected in river water, with concentrations ranging from 0.03 nanograms per liter in rivers draining into Hudson Bay to 5 nanograms per liter in the Ob River.

Russian data also include analyses of suspended particles. Some of the contaminants are present on particles at levels 10 to 100 times higher than have been recorded from Canadian or Norwegian rivers. For example, PCB concentrations up to 26.6 micrograms per gram dry weight and DDT concentrations up to 2.75 micrograms per gram dry weight have been recorded. Further investigations are needed before drawing any final conclusions, but if verified, these data show that some Russian rivers draining into the Arctic Ocean are more contaminated than surface waters in urban areas of North America and western Europe. These rivers could constitute a significant source to the Arctic Ocean.

Studies of lake water are only available from a few lakes in Canada and from two lakes in Russia. Hexachlorocyclohexane, hexachlorobenzene, and toxaphene have been detected, as have several less-persistent organochlorines. This suggests that lake water can be an important reservoir for water-soluble organic contaminants. PCB levels in lake water in Canada exceed USEPA quality guidelines for protection of aquatic wildlife (0.017 nanograms per liter).

The data from Russian lakes, from the Taimyr Peninsula, mirror the high levels of hexa-

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chlorocyclohexane, DDT, and PCBs that were found in river water. PCB levels are high enough to exceed Canadian environmental quality guidelines (1 nanogram per liter). Again, because of possible quality assurance problems, these measurements need to be verified before firm conclusions are drawn.

Lake sediments have low levels of POPs

The figure to the right shows the levels of DDT and PCBs in lake sediments from remote areas of the Arctic. Persistent organochlorines appear in most of the sediments, with concentrations ranging from 0.01 to 40 nanograms per gram dry sediment. These concentrations are similar to or lower than those in sediments from midlatitude lakes in North America and much lower than those in industrialized areas. One exception is Wonder Lake in central Alaska (240 nanograms PCBs per gram dry sediment), which may indicate local contamination. The levels in Wonder Lake are above the median effect range in environmental guidelines for aquatic life. Five lakes in Canada, one in Norway (on Bear Island), and one in Russia (on the Taimyr Peninsula) exceeded the minimal effect range.

The south–north comparison of available data from Canada, Alaska, and Finland shows that PCB levels in lake sediments decrease sixfold from 46°N to 81°N.

Core samples of lake sediments give a picture of how the deposition of contaminants has changed over time. PCBs first appear in the 1940s and their input peaks in the 1970s and early 1980s, which corresponds to the maximum use of PCBs in industrialized areas. In the Canadian High Arctic, the onset as well as the peak come somewhat later. This supports the idea that time trends in deposition of persistent, semi-volatile organic compounds in the polar region will be delayed and prolonged compared with areas closer to the sources.

Dioxins and furans have also been analyzed in some lake sediments. The levels in all sediments exceeded the Canadian environmental quality guidelines for protecting aquatic life (0.09 picograms per gram dry weight). In Great Slave Lake, Canada, the chemical signatures point to two different sources: combustion and the effluent from bleaching in pulp mills in the Peace-Athabasca-Slave River basin. The peak in deposition in the 1950s coincides with increased industrial activity in the region to the south of the lake. with the introduction of two chlorine-bleached kraft pulp mills within the drainage basin, and with the use of pesticides contaminated with dioxins and furans. The mills have since switched to elemental-chlorine-free bleaching.



Sediments from a few lakes in northern Finland, Norway, and Sweden also contain dioxins and furans. Air transport from combustion sources seems to be the only logical source. The levels in these lakes are about ten times higher than in the lakes studied in Canada, and similar to background levels in some other areas of Europe.

Canadian data point to toxaphene as a major contaminant in freshwater fish

The general picture for animals in freshwater systems is that levels of organic contaminants are higher than in the terrestrial environment, but in most cases below levels that would pose problems for fish. In a few cases, however, the contaminant levels are high enough to affect the quality of the fish as food.

Based on measurements mostly from Canada, the major contaminant in fish is toxaphene. There are no known current sources of toxaphene in the Arctic, and the contamination probably derives from long-range transport. Levels are highest in fish that feed on other fish, such as lake trout and burbot. Burbot liver is of special concern because its high fat content seems to increase its ability to accumulate organic compounds. The toxaphene levels in burbot measured in Canadian lakes range from 40 to 2300 nanograms per gram liver. These levels are close to those known to affect bone development and reproduction in other fish.

PCBs, ng/g	DDT, ng/g
<mark> </mark> < 2	<mark>O</mark> < 0.25
2-7	0.25-1
7-15	O 1-2
15-25	• 2-4
25-40	• 4-5.25
>40	

PCBs and DDT levels (dry weight) in surface sediments from remote lakes.

35

30

40



POP levels in landlocked and migratory Arctic char. Freshwater fish also have PCBs, DDT, some chlordane-related compounds, hexachlorocyclohexane, and hexachlorobenzene in their bodies. Excluding a few hot spots, the levels of PCBs in lake trout, a predatory fish, range from 9 to 450 nanograms per gram. The map above shows the contaminant levels in landlocked and migratory Arctic char.

Lake Laberge has high levels of all POPs

Some hot spots of contamination cannot be explained by local sources. One such example is Lake Laberge in Canada, downstream from the community of Whitehorse. The lake is used for commercial, sports, and native subsistence fishing. Analyses of lake trout, burbot, and lake whitefish revealed that the levels of PCBs, DDT, and toxaphene were 30 times higher than in other lakes in the Northwest Territories, and comparable to contaminant levels in the severely polluted Great Lakes.

There have been numerous suggestions about the sources of these contaminants. Current scientific opinion is that long-range air transport is the major pathway, which in combination with changes in food-web structure has led to significant biomagnification. For example, lake trout in Lake Laberge eat only fish and have much more fat in their bodies than lake trout in other lakes in the region. Changes in the food web have been attributed to overfishing and to increased productivity due to nutrients from the Whitehorse sewage lagoon.

Levels in marine environments

The Arctic marine environment collects contaminants from the air, but also from ocean currents, rivers discharging into the Arctic Ocean, and sea ice that transports POP-laden particles.

Seawater measurements reflect pathways of contaminant transport

The table below presents some examples of contaminant levels in seawater. Hexachlorocyclohexane dominates the picture, except for Russian waters where PCB levels are high, up to 15 nanograms per liter in the Kara Sea. These high levels seem to mirror the high input of PCBs from Russian rivers.

Hexachlorocyclohexane levels are highest in the Canadian Archipelago. The permanent ice

Organochlorine concentrations in seawater, pg/liter.

a	lpha-HC	H DDT	РСВ
Norwegian Sea, 1985	2750	<50	<500
Barents Sea, 1992	477	3	38
Laptev Sea, 1994	260	760	2540
Kara Sea, 1994/95	120-560	50-1250	510-3940
Pechora Sea, 1992	330	270	550
Canadian Archipelago, 1992	4180	1.0	-
Bering Sea, 1990	1500	1.0	12
Bering Sea, 1993	1990	-	-
Chukchi Sea, 1990	1400	0.3	8.4
Chukchi Sea, 1993	2060	-	-



cover in the Canadian Basin does not allow dissolved hexachlorocyclohexane to outgas very easily, and the high concentrations in surface water probably reflect higher hexachlorocyclohexane concentrations in the atmosphere a decade or so ago. Toxaphene is similarly trapped under the polar ice cap as a 'ghost of the past'. These contaminants will slowly drain through the Canadian Archipelago over several decades. A closer look at seasonal changes in concentrations also reveals that some toxaphene probably follows particles settling on the sea bottom during the summer peak of productivity.

Levels in seawater can also be used to shed light on the mechanisms that transport contaminants to the Arctic. Detailed measurements in the Bering and Chukchi Seas (see the figure above) show that hexachlorocyclohexane levels in the water increase along a south-north gradient. This has been suggested as evidence for a cold-condensation theory; that semivolatile contaminants condense out of the atmosphere as temperatures drop. Less volatile contaminants, such as PCBs, DDT, and chlordane, were present at lower levels in the Bering and Chukchi Seas than in more temperate latitudes.



Marine sediments are comparatively clean

Concentrations of organic contaminants in Arctic marine sediments are, in general, extremely low compared with freshwater sediments, and ten to a hundred times lower than in the Baltic Sea. The most apparent geographic trends are that concentrations of PCBs, hexachlorocyclohexane, and hexachlorobenzene are higher closer to the shore along the Norwegian coast than in the open sea. They are also higher in gulfs and river mouths along the Russian coast, and around Svalbard; see figure below. Industrial and municipal effluent as well as runoff and river water are probable explanations in some areas.

In some places, dumping influences local sediment levels. Two examples of PCB-pollu-



Contaminant budget for the Arctic Ocean

Information about levels of contaminants in seawater and river water along with equations describing exchange with the atmosphere have been used to calculate budgets for hexachlorocyclohexane, toxaphene, and PCBs. The results for hexachlorocyclohexane are presented here. The picture that emerges is that the Arctic Ocean is in a steady state for gamma-hexachlorocyclohexane and that it is exporting alpha-hexachlorocyclohexane. Therefore, we can expect a decline in the large pool of alpha-hexachlorocyclohexane in the Canadian Basin.

For toxaphene, lack of data prevents a detailed mass balance calculation, but the available information suggests that inputs and outputs of toxaphene are roughly equal. The atmosphere accounts for slightly more than 40 percent of the input. Volatilization is an important loss process, though most toxaphene is removed by ocean currents via the Canadian Archipelago and Greenland.

The mass balance for PCBs is preliminary and based on some results from Russian rivers, that have to be verified. The current picture indicates that inputs exceed outputs by 50 percent. 24 percent of the overall input is via rivers. Some portion of PCBs is removed by settling particles, but most of the export is via ocean currents.

Ålpha-hexachlorocyclohexane concentration in seawater increases moving from south to north, illustrating the cold-condensation effect.

PCB concentrations in marine surface sediments.



PCB concentrations in seabird eggs.

ted military sites are Cambridge Bay harbor off Victoria Island in Canada and Thule in North Greenland.

Dioxins and furans also show up in marine sediment, but at low levels. Concentrations in the Barents Sea, for example, are ten to twenty times lower than those in the North Sea. The chemical signature suggests that combustion is the major source.

Marine food webs are not well studied

One of the major concerns in the marine environment is that POPs are incorporated in the fat of small invertebrates low in the food web and subsequently accumulated and biomagnified by larger animals. Levels of persistent organic pollutants in marine food webs are poorly studied. Available measurements show that contaminant levels in fish are generally lower in the Arctic than in temperate seas.

The highest levels occur in predatory fish, high on the food chain. Greenland halibut from Canada have levels of toxaphene that are close to those known to affect bone development and reproduction.

Military radar sites may have contaminated marine fish

PCB and DDT levels are high close to several military sites and accompanying communities. At Cambridge Bay off Victoria Island in Canada, a DEW Line radar station has contaminated the local marine environment. At this site, DDT levels in benthic predatory fish such as fourhorn sculpins averaged 25 nanograms per gram muscle and PCB levels averaged 50 nanograms per gram muscle. However, the nearby bays were not contaminant-free, and an analysis of the pattern among the POPs showed that rivers can also be a significant source of contamination along the coast.

Some seabirds have very high PCB and DDT levels

Seabirds breeding in the High Arctic are contaminated with the same suite of compounds as those breeding in temperate regions. The levels vary depending on feeding habits; see the diagram above. The common eider and king eider, which feed on mussels, have the lowest levels, whereas fish-eating cormorants (shag) have relatively high levels. Other birds with high levels are the glaucous gull, black-legged kittiwake, and puffin. A study of seabird eggs showed that glaucous gulls from Prince Leopold Island in the Canadian High Arctic had organochlorine levels four to ten times higher than other birds in the area, reflecting their position as a top predator in the marine food web.

In migratory seabirds, the load of contaminants mirrors migratory habits. Eiders in Low-Arctic Canada that overwinter off Newfoundland and in the Gulf of St. Lawrence, in waters that are known to be contaminated, have a contaminant load that is considerably higher than that of populations that overwinter in northern, cleaner waters. There is also a general geographic pattern with cleaner birds in the west of Canada than in the eastern Low Arctic.

A study of birds from Svalbard, the northern Norwegian coast, and the Kola Peninsula found no major differences between the three areas. The birds may share the same overwintering areas, and contaminants are probably well mixed throughout the Barents Sea.

Do Arctic marine birds suffer from their load of toxic compounds, as has been clearly shown in birds from polluted environments? In general, contaminant levels in the Arctic are lower than in, for example, the Great Lakes region, but a study of glaucous gulls from Svalbard brings some disturbing observations. Birds that were found dead of unknown causes had high concentrations of PCBs in their fat. The birds were probably starved and stressed before they died, and it could be that the elevated levels of PCBs in their remaining fat were high enough to affect vital functions in the birds. The levels of contaminants in eggs from species on which the glaucous gull feeds are higher in Svalbard than in, for example, the Canadian High Arctic.

A comparison to known low-effect levels in other bird species also causes concern. The diagram at the top of the opposite page shows that PCB levels in seabirds from the Canadian and Norwegian Arctic approach or exceed lev-

els that are known to affect reproduction in other bird species. PCB levels in cormorant (shag) and fulmar are at or above the loweffect level for embryo deformities and the low- or no-effect level for hatching success. Puffin, thick-billed murre, common guillemot, black guillemot, and kittiwake have levels that exceed the upper no-effect level for hatching success. PCB levels in glaucous and herring gulls are even higher and approach or exceed the low-effect level for hatching success.

Toothed whales have high levels of POPs

The levels of contaminants in different species of whale also reflect their feeding habits. Beluga, narwhal, and harbor porpoise eat fish and have higher levels of contaminants than minke whale, which mostly eat invertebrates.

Harbor porpoise from Northern Norway had the highest levels of PCBs and DDT of any of the whales in Arctic waters and were in the same range as animals from southern Norway; see the map below. These PCB levels exceed the threshold for immune toxicity. In North American waters, PCB levels in beluga range from 2.6 to 6.0 micrograms per gram blubber.

The levels of contaminants in whales and other Arctic marine mammals are higher in males than in females and also depend on the age of the animal. Adult female whales usually have lower levels than adult males. In males, unlike females, the levels increase with age. Studies of cow-calf pairs show that whales get a large portion of their load of organochlorines very early in life, from their mother's milk. Females have accumulated large burdens by the time they begin to reproduce, and pass this on in marine mammal milk which is very rich in fat. In the newborn calf, this initial dose is further concentrated as the young animal uses the fat for energy. Only when it starts feeding on its own will the contaminants from the milk be diluted in its body. For the female, nursing the young rids her body of some of its contaminant load, which is why reproducing females have much lower concentrations of organochlorines than males of the same age.

How do the contaminants affect the whales? One study shows that when whales are starved, PCB levels may affect their livers. The whales in this study had entered a freshwater lake system when the water was free of ice and were trapped when winter set in. Their breathing holes became smaller and smaller. By mid-winter, native hunters decided to harvest the animals. At this time they weighed about 200 kilograms less that healthy whales of the same length. This starvation probably forced the whales to use their blubber fat for energy, which in turn increased the concentration of PCBs in the remaining fat. A correlation was seen between PCB levels and the amounts of liver detoxifying enzymes. One conclusion is that currently-observed body burdens of contaminants can be associated with



subtle effects when a lack of food forces whales to burn fat reserves for energy.

For assessing effects other than on the liver, we must rely on comparisons with known effects levels in other species and other tissues; see the diagram on the next page. The major concern seems to be PCBs, the levels of which range in blubber from 1.9 to 6.3 micrograms per gram lipid. This is higher than the no-effect and low-effect levels associated with subtle neurobehavioral effects (assuming that levels in blubber reflect levels in blood serum), but below the no-effect level for reproduction and kit survival in mink. PCB levels in fish from some sites are also high enough to cause concern about effects from the whales' diet.





to different thresholds for biological effects. * no-observed-effect level (NOEL) ** lowest-observedeffect level (LOEL) or lowest-observedadverse-effect level (LOAEL). + refers to average values



PCB concentrations in blubber from harbor porpoise and minke whale from Norway.



Geographical trends for DDT and PCBs in harp and ringed seals (range averages). The locations are arranged from west to east.

Left: PCB levels in food items compared to thresholds for biological effects and to environmental quality guidelines to protect fish-eating aquatic wildlife. Right: PCB levels in mammals compared to different threshold for biological effects.

* lowest-observedadverse-effect level ** no-observed-effect level or no-observed-adverse effect level EC_{50} = concentration at which half of the animals in the study were affected.

Seals in the Barents Sea are more contaminated than in other Arctic regions

The most important contaminants in seals are PCBs, chlordane, and DDT. The figure above shows the levels of PCBs and DDT in harp and ringed seals from Arctic waters. The mean PCB levels range from 0.24 to 5.7 micrograms per gram blubber. These are higher than in the fish they eat, which shows the role of biomagnification in the Arctic food web, but are still much lower than in polluted areas such as the Baltic Sea. PCB and DDT levels in seals seem to increase from west to east, with the highest levels seen in seals from Svalbard, northern Norway, and Russia. The geographic trend for hexachloroxyclohexane is the opposite, with the highest levels in Canadian ringed seal.

The loads of PCBs in seals are considerably lower than those associated with poor reproductive success in seals from polluted areas, but exceed the no-effect and low-effect levels for subtle neurobehavioral effects in laboratory animals; see below right. Parts of the diet of some seals are also contaminated enough to cause concern. If seals are as sensitive to PCBs as mink, on which most diet studies have been done, several fish species exceed no-effect concentrations for dietary intake; see left panel below.

POP levels in walrus reflect their feeding habits

Walrus feed mostly on bottom-dwelling animals that have low levels of organic contaminants. However, high levels of PCBs in some animals suggest that there are important exceptions. Nineteen of 53 walruses from across the Canadian Arctic had more than 1 microgram PCBs per gram blubber. These results suggest that some walrus probably have ringed seal as an important part of their diet. The high levels were recorded in walrus from east Hudson Bay, Foxe Basin, and east Baffin Island. Walrus from Svalbard have levels of PCBs as high as walrus from east Hudson Bay.

No one has studied the biological effects of POPs on walrus. However, a comparison of known-effects levels from otter and mink show that walrus from east Hudson Bay and Svalbard exceed the no-effect levels for reproduction and kit survival. PCB and dioxin levels in seal-eating walrus are similar to the lowest levels that have been associated with immune suppression in seals.

Polar bears are at risk for immune and reproductive effects

As a top predator in the marine food web, the polar bear could be the species most exposed to contamination of the Arctic environment.



O3 Persistent Organic Pollutants





There have been several studies of POP levels and possible biological effects in polar bear, especially to examine whether POPs damage the ability of bears to conceive and to rear their young.

Polar bears do indeed accumulate significant amounts of many organic contaminants. The maps above show the results of a recent circumpolar study on which biologists and conservation managers from Canada, USA, Greenland, Russia, and Norway collaborated. The contaminant levels are high compared with most other Arctic animals. The median values are 7.2 micrograms PCBs per gram fat, 2.0 micrograms chlordane per gram fat, 0.19 micrograms DDE per gram fat, and 0.15 micrograms dieldrin per gram fat. These levels are lower than those in seals from highly polluted waters such as the Baltic Sea.

In six of the regions included in the study, PCB concentrations in polar bears are in a range that is known to affect the reproduction of mink, and in some other areas they are approaching these levels. Even if mink, when compared with other animals, are extremely sensitive to organic contaminants, one cannot discount the risk that PCBs at current levels affect the reproductive ability of some polar bears. One of the areas where levels in polar bears are highest is Svalbard, but so far it has not been possible to relate contaminant loads in individual female bears on Svalbard to their ability to reproduce.

One concern is that polar bear cubs receive a high dose of POPs from their mothers' milk. The levels of the fat-soluble compounds are indeed very high in cubs of the year, higher even than in yearlings. This means that young cubs are exposed at a period of growth and development when they may be most sensitive. One study found that the mortality of young polar bears on Svalbard was higher than in other areas.

In 1996, two female cubs accompanying their mother on Svalbard were noted to have abnormal external genitalia, making them pseudo-hermaphrodites or hermaphrodites. The causes behind this abnormal development are unknown. It could be a rare natural event, or the result of a hormone-producing tumor in the mother, or a consequence of toxic substances.

For PCBs, the levels in adult bears are high enough to exceed the no-effect and low-effect levels associated with subtle neurobehavioral effects in offspring of laboratory animals. At four of the study sites - Svalbard, east Greenland, McClure Strait, and eastern Hudson Bay - they exceed the PCB levels known to affect kit survival in mink. At other sites they are close to this limit. Polar bears from east Greenland, Svalbard, and McClure Strait also have enough PCBs in their bodies to raise serious concerns about immune-system effects. Some bears on Svalbard have levels exceeding those associated with poor reproduction in seals. Levels of dioxin-like substances in polar bear from Resolute Bay, Canada, and Svalbard may be high enough to affect the immune system.

Polar bears in Canada have increased liver enzyme activity, which has been correlated with the levels of coplanar PCBs. There are also some indications that PCBs have affected the thyroid hormone and vitamin A levels. These biomarkers indicate that the diet of polar bears is probably contaminated enough to affect the functioning of sensitive hormone systems.

A look at the diet of polar bears supports the concerns raised by POP levels in the polar bears themselves. The levels of PCBs and dioxin-like substances in ringed seal blubber from all sites exceed environmental guidelines for protecting wildlife. DDT levels exceed some of the guidelines.

Arctic fox often scavenge seal carcasses left by polar bears. They might therefore get just as much contamination from their food as do polar bears, and be at similar risk for reproductive and immune effects. So far, contaminants in Arctic fox have only been studied on Svalbard, where their levels are as high or higher than those of polar bear.

The circumpolar polar bear study makes it possible to look at geographic variations and to infer possible underlying causes. There are POP levels in polar bear and Arctic fox.

clear signs of extensive transport of POPs to all areas of the Arctic and subarctic. There are also some notable peaks in concentration. For example, PCB levels in polar bears are highest on the east coast of Greenland, on Svalbard, and in the Arctic Ocean. The Greenland and Svalbard peaks could be caused by combined long-range atmospheric transport of PCBs from North America and from Europe. Another source could be ice that has received contami-

Food-web structure, selective break-down, and stress related to biological effects

The most striking feature of organochlorine levels in Arctic biota is the biomagnification that occurs in food webs. The highest levels are usually recorded in top predators in environments with long food chains, such as polar bear, which are third-level carnivores in the marine food chain. Extremely high contaminant levels have also been recorded in bottom-dwelling amphipods that feed on the carcasses of dead animals. This can be contrasted to the short terrestrial food chain, where wolves are only firstlevel carnivores, and have correspondingly lower contaminant loads.

Many Arctic animals are opportunistic feeders, and may occupy more than one position in the food web. Walrus feeding on seals have much higher contaminant loads than walrus that feed on mussels. Also, changes in food-web structure can affect contaminant intake. Lake Laberge in Canada illustrates how overfishing and higher nutrient input have changed feeding habits of lake trout, resulting in elevated contaminant levels.

Not all contaminants biomagnify. In fact, the chemical signatures in top-level predators, such as the polar bear, reveal that some compounds are effectively degraded along the food chain. For example, polar bears can break down DDT, so that their DDT levels are sometimes lower than those of ringed seal, their main food source.

Much of the contaminant load is stored in fatty tissues. This fat is used for energy. When a substantial proportion of the fat is used up, as happens in starvation periods, the concentration of contaminants in the remaining fat increases, with a corresponding increase in the concentration in blood and in vital organs. There are some indications from predatory birds that the combination of a high body burden of contaminants and prolonged periods of starvation may contribute to the death of an animal.

> nants from atmospheric deposition as well as from the Russian continental shelf. Such transport would also bring PCBs to the Barents Sea and expose the bear population on Svalbard. There are parallel observations of high levels in ringed seals, which is the main diet of the polar bear.

The peak in the Arctic Ocean is more difficult to understand. It could be that the food web in the permanent ice pack has a different structure than that of coastal and ice-margin areas, and is based more on the microscopic animals that live in ice, where they can accumulate large amounts of contaminants.

The levels of chlordane in polar bears show a peak in southeastern Hudson Bay in Canada, which also has the highest DDE and dieldrin levels. This is probably a sign of atmospheric transport to this area in summer from eastern and central North America.

Time trends in pike from Lake Storvindeln, Sweden.



Hexachlorocyclohexanes are highest in bears from the Bering and Chukchi Seas, which reflects continuing input from Asia. Other POPs are generally lower in this region.

Time trends

Is the burden of contaminants in Arctic animals increasing or decreasing? Will contaminants used in the past continue to move to the Arctic even if use is reduced or discontinued? The results from a few long-term studies of temporal trends in Arctic biota indicate that PCB and DDT levels in the Arctic environment have declined in the past 20 to 25 years, since the first controls began on DDT and on the use of PCBs in open systems. Evidence from dated sediment cores also shows input declines in subarctic latitudes, as discussed under *Lake sediments have low levels of POPs*.

Sharp declines in PCB and DDT levels were seen in the 1970s and 1980s, and some time trends indicate continued declines during the late 1980s and 1990s, whereas others are unclear. There are no long-term standardized data sets for the High Arctic. Levels in biota are extremely variable, and observed levels are still near the thresholds for biological effects. These uncertainties and variabilities make it difficult to speculate about the future, and reinforce the importance of careful sampling programs and the need to archive samples for future analysis.

Less is known about time trends for hexachlorocyclohexane, hexachlorobenzene, chlordane, toxaphene, dieldrin, and dioxins and furans. Of these, hexachlorocyclohexane has been followed most closely. In air, there was a nine-fold decrease from 1979 to 1993 in measurements from the Bering and Chukchi Seas and from several locations in the Canadian Arctic Archipelago. In the European Arctic, however, alpha-hexachlorocyclohexane levels seem to have declined two-fold, but lindane levels have increased from 1984 to 1992, reflecting different regional inputs.

The remainder of this section presents the results of some specific time-trend studies.

Fish and moss in Scandinavia show a decreasing contaminant load

Some of the few studies that have been designed specifically to look at time trends in biota have been conducted in Scandinavia, which is probably more representative of the subarctic than of the High Arctic. The studies also may not reflect the load in long-lived species such as polar bear and beluga. Fish provide the most detailed information.

In Sweden, a study of POP levels from 1967 to 1995 shows a promising trend; see graph to the left. The levels of DDT, PCBs, dioxin-like substances, hexachlorocyclohexane, lindane, and hexachlorobenzene have all declined, in lakes as well as in the Baltic Sea. The decline of PCBs is further verified in a Finnish time trend study.

The start of the declines in PCBs and DDT, in the 1970s, coincides with decisions to reduce environmental pollution, and shows the value of such political efforts. Even in remote areas of Arctic Sweden, the sudden decline in the Russian economy in the late 1980s shows up in the data, as do reductions in the production and use of pesticides. As a reminder of the consequences of renewed use of pesticides, spraying of DDT in the former East Germany in the summers of 1983 and 1984 caused readily-detectable increases of the contaminant in Swedish lakes.

PCBs continue to be a concern. PCB levels have not declined as fast as those of pesticides. This indicates a continuing release of PCBs into the environment, and thus a need to take action against potential sources.

In Norway, moss has been used to look for time trends in the deposition of PCBs. In coastal areas, the mean concentration fell from 21.2 nanograms per gram in 1977 to 6.9 nanograms per gram in 1990. Because moss takes up all its nutrients from the air, this decline mirrors a decrease in atmospheric PCB concentrations.

Marine animals show a varied geographic picture

Results from monitoring marine animals provide good evidence for declining concentrations of the major organochlorines in both the European and the North American Arctic. For example, seabirds breeding in northern Norway had 60 percent lower levels of hexachlorobenzene, 85 percent lower levels of DDE, and 78 percent lower levels of PCBs in the period 1983 to 1993 compared with 1979 to 1981.

In the Canadian High Arctic, PCB and DDT levels in eggs of migratory seabirds declined from 1975 to 1993, mostly in the late 1970s and early 1980s. This decline may reflect an overall reduction in organochlorine levels in the North Atlantic. The decline is not uniform, however. In one of the birds, the ivory gull, PCB and chlordane levels in the eggs have increased.

Declines in POPs in seals and whales from the western Canadian Arctic are not as steep as those observed in seabirds and whales from eastern Canada. From 1972 to 1991, PCB concentrations declined five-fold and DDT concentrations three-fold in marine mammals from the western Canadian Arctic.

Over the past 10 to 12 years, seals and walrus from Eastern Canada and Greenland have shown no declines in contaminant levels, nor have there been any declines in DDT, PCBs, chlordane, or toxaphene in female ringed seal or in male narwhal from Lancaster Sound from the mid 1980s to the early 1990s.

PCB levels in Arctic fox from Svalbard seem to have been similar in the 1970s and in the 1980s.

Summary

All persistent organic pollutants in AMAP's monitoring program have been found in the Arctic. The levels are generally lower than in temperate areas, but for several substances they are still in concentration ranges in which effects on some animals are expected. These include reproductive effects in birds from DDT and in some marine mammals from PCBs and dioxinlike compounds. Current concentrations in several Arctic species are also close to or above thresholds known to be associated with immunosuppressive and neurotoxic effects. The most vulnerable animals are those high in the food web, such as polar bear and birds of prey.

Biomagnification is one major factor contributing to the high levels and biological effects of persistent organic pollutants in Arctic animals. Another biological pathway is via migratory birds that overwinter in polluted environments.

The major source of persistent organic pollutants in the Arctic is long-range transport via air currents, as demonstrated by monitoring of air concentrations. There may also be significant sources of some contaminants, such as PCBs, DDT, and hexachlorocyclohexane, within the AMAP region, but these are not well documented. Data from river water and sediments indicate a substantial input from Russian rivers into the Arctic marine environment, but these data must still be verified.

Available data point to some geographic trends. In general, the levels of PCBs and DDT seem to be higher around Svalbard, in the southern Barents Sea, and in eastern Greenland than in, for example, the Canadian High Arctic. Levels of hexachlorocyclohexane appear to be higher in the Canadian Arctic than in Eurasia. Very limited data from Russia and Alaska were available for this assessment. The lack of circumpolar data limits our understanding of sources, transport pathways, and mechanisms for focusing contaminants. The role of sea ice in transporting contaminants and then releasing them during melting warrants further investigation.

A few studies of time trends point to a decreasing load of PCBs and DDT in subarctic regions from the 1970s to the 1980s, after use of these substances was restricted or banned. However, it is not clear whether this decline has continued from the 1980s to the 1990s, or if similar declines have occurred in the High Arctic. The decline seems to be slower for PCBs, which may indicate continued low-level leakage to the environment from unknown or poorly studied sources.