TRANSPORT AND FATE OF PERSISTENT POLLUTANTS IN THE ARCTIC

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Introduction

The Arctic is still considered as one of the few unpolluted regions in the world. This if off course true if one compare the Arctic region with other regions more influenced by human activities. However, the Arctic region is not isolated from all human activities. Due to the global dimension of environmental pollution, all pristine, yet unpolluted areas of our globe including the Arctic and the Antarctic have to cope with various environmental threats. Most of the environmental problems remote, pristine regions like the Arctic are facing today remained undiscovered for a long time. However, the first indication for long-range transport in the Arctic is given by Fridtjof Nansen during his famous expedition (1893-1896), which led to the discovery of the trans–polar ice drift. He described large amounts of sediment cover on the trans-polar ice sheet far from coastal influences and explained this phenomenon with the transport of river-related sediments and soil from the large Russian Arctic rivers. Although this discovery is not directly describing Arctic pollution, his finding can at least be considered as a description of a potential transport medium for pollutants.

Long-range transport

In the early 1980s questions were raised for which good answers were lacking. Why were the levels of selected persistent organochlorines in Arctic organisms representing high trophic levels (Atlantic cod, gulls, Waders, birds of prey and polar bears) so high, that ecotoxicological effects can be expected?
Why were the levels of persistent organic pollutants (POPs) in mothers milk and blood of indigenous people from Canada and Greenland so elevated, although these people are living at such remote place fare away from potential source areas

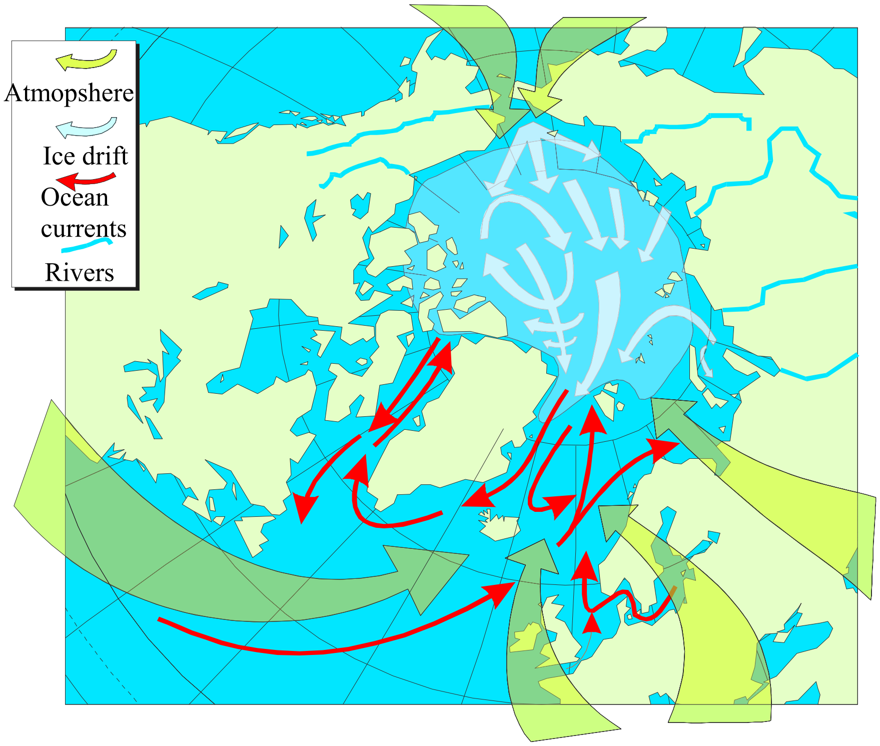


Figure 1: Different entrance routes for pollutants.

Antropogenic pollutants can be transported into the Arctic via ambient air, ocean currents, and transpolar ice movements.

(DEWAILLY et al. 1992, 1993, Muir et al. 1992, Kuhnlein und Chan 2000)? This phenomenon was difficult to explain, because the concentrations of these specific compounds were relatively low in seawater and sediment from pristine Arctic sampling sites. No significant local sources for these types of compounds exists in the Arctic, therefore, they have to be transported into the region by 'natural' pathways. For pristine regions like the Arctic, in general four possible pathways exist for the transport of persistent environmental pollutants. The Atmosphere, sea currents, the transpolar pack ice and the large Arctic rivers (Ob, Pechora, Yenisey, Mackenzie) must be considered as the main entrance way for antropogenic pollutants into the Arctic ecosystem (figure 1). Which way a respective pollutant is transported into the Arctic depends on the chemical and physical properties of the substance. In addition to the above mentioned transport routes, persistent compounds can also be transported into the Arctic via large steams of pelagic organisms (crustaceans, cod, herring, marine mammals), animals which migrate in large groups throughout different climate zones into the polar regions. These organisms can bear the contaminant freight in their lipid rich tissue and transfer the pollutants into higher organisms via the Arctic food web.

Which way is preferred by the respective POP compounds for the transport into the Arctic? The first indication for a good explanation of the above mentioned phenomenon was delivered by Norwegian and Canadian long term air monitoring programmes (Patton et al. 1989, Oehme 1991). During the Norwegian air monitoring campaign in the early 1980s, weekly high volume air samples taken at the Zeppelin mountain air monitoring station (Ny-Ålesund) revealed several atmospheric transport episodes with relatively high POP contamination (figure 2). The results of this first campaign can be extracted from Oehme et al. 1996a. Canadian research groups found comparable results for the Canadian Arctic at the Alert Air sampling station. These first results derived from the Norwegian sample series demonstrated, that during favorable meteorological conditions, a rapid atmospheric transport (~3 - 7 days) of antropogenic compounds from well-defined middle latitude source regions can occur.

Figure 2:
During year-around air monitoring compaigns since 1979 the Norwegian Institute for Air Research investigate the transport properties of POPs from source regions into the Ny-Ålesund area at Svalbard.

The atmosphere as transport medium

The atmospheric long-range transport as main transport way for persistent pollutants is off course not a new feature. Already 30 years ago sulfur dioxide (SO_2), nitrite and related compounds (NO_x) originated from the industrial zones of central and Eastern Europe made their way into Scandinavia via the atmosphere (Galloway 1995, Rodhe et al. 1995, Torseth and Semb 1998). This phenomenon is today known as 'acid rain' that caused acidification of a large number of Scandinavian lakes and ponds. Another well-known example for atmospheric transport of pollutants is the vertical transport of chlorofluorocarbons (CFC's) into the polar atmosphere, which caused the depletion of the ozone layer. While over Antarctica a huge 'ozone whole' appears in spring every year, the ozone depletion is considerably weaker and varies from year to year (Rinsland et al. 1995). A much earlier indication for atmospheric long-range transport is dated back to the 1950s when airplane pilots reported smog-like phenomena over the surface of the Arctic region during their flights across the North Pole.

Figure 3: 'Arctic haze', smog-like appearance in the polar regions caused by sulfate containing particle from industrials countries, reported for the first time from airliner pilots crossing the North pole in the 1950s.

This Arctic smog is still to be seen during intercontinental flights across the Arctic. The smog phenomenon is mainly caused by sulfate containing dust particles originated from industrial regions in Europe, USA and Asia, transported via the atmosphere for several thousand kilometers and deposited in the Arctic. The phenomenon is also known as 'Arctic haze' (figure 3). Although the potential for atmospheric long-range transport was known for quite a long time atmospheric POP transport was not immediately seen as a main cause of the high contamination of Arctic organisms including men. First after the evidence for atmospheric POP transport was presented by Canadian and Norwegian Research groups, a vivid discussion about the role of atmospheric long-range transport was initiated and is still ongoing. All circum-Arctic countries decided to conclude their national research and contaminant monitoring efforts in a common surveillance program, the so-called Arctic monitoring and Assessment Programme (AMAP). Research and monitoring group of all AMAP countries met in Tromsø (1997) in order to summarise the state-of-the-art of the contaminant situation of the Arctic. The results presented proved the importance of atmospheric long-range transport for the contamination of the Arctic (heavy metals and POPs). A comprehensive report about all monitoring and research efforts published by the AMAP secretariat, demonstrates the contamination situation the Arctic ecosystem is facing today (MacDonald et al. 2000, AMAP, 1997 and 1998).

Long-range transport as contamination source

Transport routes via atmosphere, ocean currents, rivers as well as ice transport must be considered as the largest contamination sources for Arctic ecosystem (figure 1). Practically, local contamination sources are neglectable for most of the antropogenic pollutants found in the Arctic. Only heating of buildings and other incineration processes contribute significantly to the input of certain pollutants like polycyclic aromatic hydrocarbons (PAH). The transport efficiency is one of the main differences between the above listed transport routes. At optimal weather situations a fast air transport can take place in days or weeks, from the source regions into the Arctic.

The transport via sea currents, however, can take several years. The exact dimensions of the ocean-related pollutant transport were determined for the first time by tracking antropogenic pollutants. The transport properties of radioactive isotopes released by an accident in the cooling system of the nuclear power plant in Sellafield (UK) into the Irish Sea were investigated.

From 1975 to 1985 considerable amounts of isotopes with a long lifetime like ^{137}Cs and ^{134}Cs were released into the Irish Sea. This type of radioisotopes is exclusively originated from antropogenic sources. No natural sources are known. Thus, during long-term research programmes the transport route and residence time of these isotopes were determined during their way from the Irish Sea into the Arctic (Aarkrog et. al. 1995). For their way along the Norwegian coast into the high Arctic the following duration times of the ocean current transport of these selected radioisotopes were determined: For the 1500 km from southern Norway to North Cape a transport duration of about 2 years is estimated. For the 2200 km from southern Norway until North-Spitzbergen (Svalbard) transport duration of 4-5 year was estimated. These results represent the first investigation on the ocean-related long-range transport of environmental pollutants into the Northern Hemisphere.

Properties favorable for long-range transport

In addition to radioisotopes, there exist a large variety of antropogenic long-range transported environmental pollutants (SIMONICH & Hites 1995). All these compound groups have one common property: They express high stability in the environment, thus, these compounds must be considered as persistent (Webster et al. 1998, Gouin et al. 2000). In addition to the property of the respective compound, the special geographic and meteorological conditions must be taken into account when it comes to the estimation of long-range transport processes. Usually only the cold regions of the world are especially suitable as deposition sites. Why? Due to low average temperature microbiological degradation processes are significantly slowed down compared to middle latitude or tropic region. The half-life time of a persistent compound is increased dramatically in Polar Regions.

Mainly two types of antropogenic environmental pollutants can be described, namely inorganic and organic pollutants. The following examples shall illustrate these two groups of contaminants (see also table 1):
Inorganic pollutants: Radioisotopes, heavy metals, organo-metal compounds, 'acid rain' (NO_x, SO_2).
Organic pollutants: Industrial chemicals and unwanted byproducts of industrial production: Hexachlorbenzene (HCB), CFCs, high volatile compounds (VOC), polychlorinated biphenyls (PCBs). Incineration products (polychlorinated dioxins and furans (PCDD/F), polycyclic aromatic hydrocarbons). Crop protection chemicals or pesticides (hexachlorocyclohexane (HCH), Diphenyldichlorotrichlorethane (DDT), chlordane, toxaphene, triazine- and parathion-related compounds). Metabolites (oxychlordane, methylsulfone-PCBs heptachlor-*endo/exo*-epoxide, diphenyldichlorotrichlorethene DDE).

A large number of inorganic compounds is also known to be transported into the Arctic via the atmosphere. Examples like 'acid rain (NO_x and SO_2) and lead (Pb) from the exhaust of European cars and engines demonstrate their transport properties. However, since regulation measures for the restriction of lead used in fuel products were introduced in all western European countries, a significant decrease of the lead concentration in Scandinavian and Arctic ambient air could be observed within the last decade.

In addition to the upper mentioned components, during the recent years a continuous increase of mercury (Hg) in the Arctic is measured at the monitoring stations (MacDonald and Bewers 1996, Lu et al. 1998, Schroeder et al. 1998, Berg et al. 2001). However, the principal mechanisms of the seasonal depending HG increase are still unknown and subject for intensive investigations.

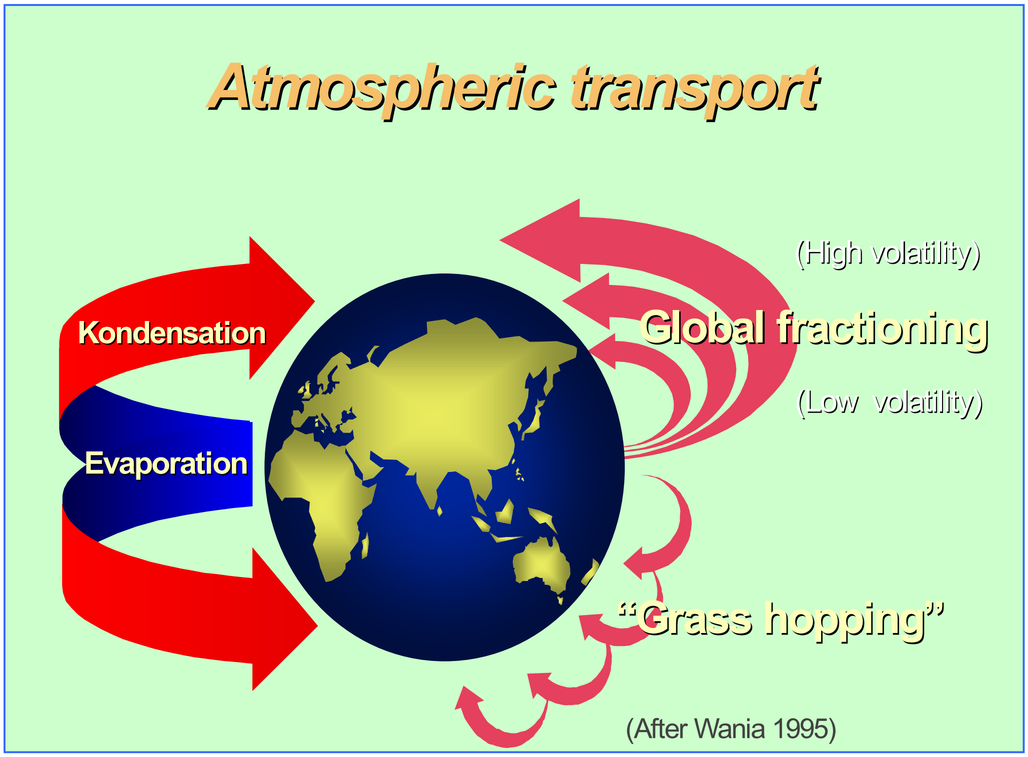
Mechanisms and pathways, which are responsible for the transport of POPs, are not so easy to grasp (Beyer et al. 2000). Large differences in chemical and physical properties like vapor pressure, volatility, polarity, solubility are the reason why the favorable medium, responsible for the transport of the respective organic compound, varies considerably.

Due to differences in solubility, one can differentiate between several categories of long-range transported organic contaminants. Lipophilic persistent pollutants like α–, β–, γ-HCH, PCBs as well as DDT and metabolites (DDE), the atmosphere is the preferred transport way into the remote Arctic. During favorable weather conditions, air masses can transport contaminants into the Arctic within few days or weeks. The above mentioned compounds adsorb on dust particles. The dust particle is then transported into the northern regions. High volatile compounds (VOC) like the low chlorinated PCBs, chlorinated benzenes etc., are directly transported in the gaseous phase For water soluble chemicals like triazine-derivatives, parathion, trifluralin, the ocean-related transport must be considered as the main transport route. As already explained, the long-range transport via the sea currents is not as fast as shown for atmospheric transport. However, also here the compounds can be transported adsorbed on particles or dissolved in the water column.

Global fractionation theory

Today, exclusively using empirical data derived by monitoring and modern analytical methods it is still inadequate to completely assess the fate of persistent contaminants in the environment. Therefore, already decades ago, comprehensive modelling tools were developed to estimate transport routes and processes (Barnicki 1994, Pacyna 1995, Liousse et al. 1996, Strand and Hov 1996, Wania and Mackay 1993, Krapivin und Phillips 2001).

Such computer based model calculations indicate that, due to its speed and effectiveness, atmospheric long-range transport can transport comparable amounts of pollutants into the Arctic, as it is known for the transport in ocean currents. The most accepted theory about atmospheric long-range transport describes the phenomenon as so-called 'global fractionation' (Wania und Mackay 1993, 1995, 1996, 1999a, Wania 1998). The 'global fractionation theory' explains atmospheric transport depending on the physico-chemical properties of the transported contaminant (e.g., solubility, vapor pressure, and molecule size). The physico-chemical properties of this substance are responsible for the length of the atmospheric transport and the subsequent deposition via rain, fog or snow in the water column, sediment or soil (figure 4). High volatile hydrocarbons, characterised by a low vapor pressure, are directly taken into the gaseous phase and immediately transported into the deposition region as shown for low chlorinated PCBs (PCB 8-99) and chlorinated benzenes. Semi-volatile compounds (SOC) like lindane (γ-HCH) and chlordanes as well as some heavier PCBs (PCB 101 – 153) are usually transported adsorbed on dust particles (Wania & Mackay 1999b). During cold weather conditions, this type of compound can be washed out and temporary deposited in the sea or



soil. During favorable warm weather conditions, those compounds are re-evaporated into the atmosphere and undergo further transport. This remobilisation is also called 'grasshopper effect' (figure 4). The role of stormy weather situations for remobilisation of semi-volatile compounds into the atmosphere is obvious but still scarcely investigated Lepri et al. 1995). Non-volatile compounds like chlorinated Dioxins and furans remain usually near the source area and are not object for long-range transport. As already mentioned, parameters like volatility and solubility are crucial in order to explain long-range transport of persistent contaminants may it by via ocean currents or atmosphere. Physico-chemical parameters like vapor pressure, polarity and molecule size, but also the overall climate situation like average medium temperature during the transport episode, deposition volume, as well as humidity are important parameters for the determination for the transport distance into the North. However, tracking of such fractionation processes is very difficult. Until recently, only laboratory experiments support the theory. A verification of repeated sea-air exchange processes was presented using the enantiomeric distribution (right- and left-handed form) of the chiral pesticide α–HCH (Falconer et al. 1995; Jantunen and Bidleman 1996; Jantunen and Bidleman 1997). A clear relation between the enantiomeric ratios (ER) found in surface water and in high-volume ambient air samples were determined. A significant deviation from the racemic distribution (ER = 1) in environmental samples can only be found when biochemical degradation occurs. Therefore, it must be assumed that α-HCH found in the ambient air was subject for microbiological degradation in the water surface before re-evaporated into the atmosphere. This is the first evidence for water-air gas exchange of α-HCH using a field experimental approach.

Long-range transport: a complex phenomenon

The 'global fractionation' theory describes long-range transport as a complex phenomenon depending on various meteorological, climate, geographical and physico-chemical parameters. Atmosphere as well as ocean currents and river-related transport are single elements in the complicated process describing the long-range transport of certain persistent pollutants into the remote and pristine Polar Regions. 'Global destination' theory and grasshopper effect are also able to explain unusual high levels of compounds in remote areas which are relatively polar and water soluble due to limited waterborne transport and subsequent remobilisation into the atmosphere (figure 4).

The role of characteristic meteorological conditions in the respective climate zones through those the contaminant is transported must not be underestimated. Strong seasonal differences in temperature and precipitation rule the global weather situation. The barrier between the cold northern air masses and the temperate southern air is called the polar front. The polar front plays a central role in explaining seasonal dependencies in atmospheric long-range transport processes. During winter, the polar front is expanding into the southern part of the Northern Hemisphere into central Europe, central USA, Asia, the European part of Russia, Belarus and the Ukraine. In these regions, the 80% of the main source areas for persistent pollutants are situated. Therefore, during the winter season, the polar front is not limiting the atmospheric transport from the sources into the Arctic region. In addition, often northerly wind directions are dominating during winter at the Northern Hemisphere. During summer, on the other hand, the polar front is situated far North and creates a meteorological barrier often difficult to penetrate for air masses transported from southern regions. Thus, the polar front represents an obvious transport limitation for airborne POPs into the Arctic. An additional factor supporting atmospheric long-range transport during winter is the fact, that significantly lower deposition events are reported for European, American, and Asian source regions during winter time. Thus, the probability that POPs are washed out from the atmosphere during precipitation is lower in winter than in summer. As a general conclusion, it can be stated that the likelihood of a long-range transport episode during winter and early spring is higher than during the summer.

As described in the 'global fractionation' theory, depending on the physico-chemical properties of the contaminant, a water soluble compound can be transported particle bound in the atmosphere before deposited and further transported with ocean currents or finally deposited in the marine sediments or in soil. However, water-soluble contaminants can also be transferred from the ground water in the surface water system and than be transported from the source region into the oceans. Already minor differences in volatility and solubility of the contaminants can be significant for the transport form, thus, whether the substance is transported dissolved in the water column or particle bound. This is also important for the atmospheric transport.

The Arctic as deposition area for POPs

In the Northern as well as in the Southern Hemisphere of our globe, the air masses in the temperate zones try to reach a temperature balance with the cold polar air masses. Thus, the physical law about the temperature balance between two gaseous systems is the general basis for the complex theory of the mostly polar directed atmospheric air fluxes and contaminant transport.

Due to low average annual temperature and the special seasonal daylight conditions in Polar Regions suitable conditions for the deposition of persistent pollutants are present. In addition, short, unbranched food chains related to a large number of individuals for each trophic level are characteristic for the polar ecosystem. Thus prolonged half-life times in addition with effective biomagnification and bioaccumulation in the polar food web lead to high concentrations of the long-range transported POPs in the higher trophic levels of the Arctic ecosystem, including indigenous people. High concentration levels were detected for compounds like PCBs and toxaphene as well as DDT and its metabolites as already mentioned in the introduction.

The following parameters demonstrate, why the Arctic is especially exposed to antropogenic POPs.

Daylight conditions

The polar night at 80° northern latitude lasts from October until March and changes within few weeks into the midnight sun. This constellation has a tremendous effect on the photochemical degradation of some persistent pollutants. Due to the lack of photochemical degradation processes, long-range transport of certain high volatile organic compounds is only observed during the winter. Chemicals like short-chain alkanes and alkenes (ethane, propane, ethene, etc.) are usually quick degraded by photochemical processes during the summer time. Therefore only low concentration levels are usually observed in Arctic air. In winter, at the other hand, concentrations of this type of compounds are usually elevated by one or two order of magnitudes.

Indications for a seasonal dependent photochemical degradation were also found for the persistent pesticide *trans*-chlordane. A seasonal shift in the chlordane patterns was found during a year around ambient air sampling campaign at Zeppelin Mountain (Ny-Ålesund, Svalbard). A picture of the station is given in figure 2.

Temperature conditions

The low annual average temperature of the Polar Regions is the main reason why microbiological degradation processes of organic material slow down to a minimum. Therefore, microbiological metabolisation of persistent pollutants in the Arctic practically does not exist. This extends the half-life time of these compounds exponentially during their stay in the Arctic. Compounds, which are known to be degraded in temperate regions within weeks or month, express much higher persistence in the Polar Regions. Thus, half-life times up to year are possible for those compounds.

Deposition

Due to the low annual average temperature, snow is the dominating form of precipitation in Polar Regions. The snow crystal possesses a large surface and adsorbs particles and contaminants to his crystalline surface more effective as a water droplet can do. Therefore snow precipitation can transport contaminants better to the surface (soil, water surface) as a rain droplet can do. In contrast to rain, snow crystals change the surface properties during aging on the surface. The surface of a snow crystal diminishes dramatically already after a short time on the ground due to the overall pressure of the surrounding snow as well as caused by weather and climate conditions during the aging process. Thus, due to surface reduction the adsorbed contaminant is either released into the soil/ ground, stays absorbed to the snow crystal or is re-evaporated into the atmosphere again. These three possibilities are totally dependent on the vapor pressure of the respective contaminant and the ambient climatic conditions. Therefore, high POP levels can often be found in upper layers containing new snow surface whereas in aged snow, situated near the ground, the contaminants are already released into the soil. Further bioaccumulation takes place from soil, microorganisms into the higher trophic levels.

These special Arctic climate and ecological conditions are the main reason why persistent contaminants are much better accumulated in the Arctic ecosystems, as it is the case for middle latitude regions.

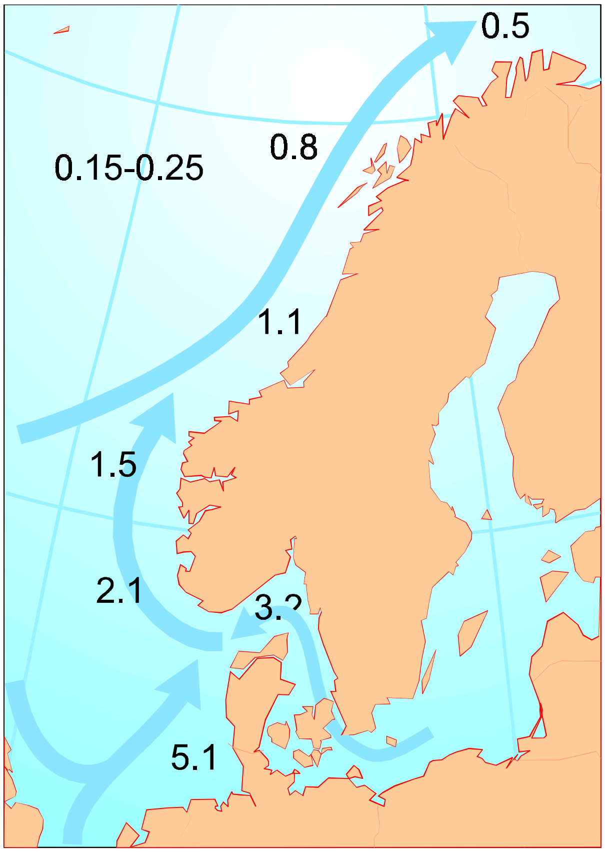


Figure 6: Transport pathway and concentration patterns determined for lindane according to investigations carried out by Gaul et al. 1992

Typical examples for long-range transported POPs.

Lindane

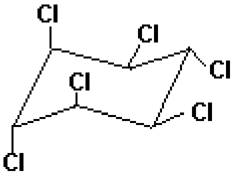


Figure 5: Lindane or γ–HCH (hexachlorocyclohexane) one of most investigated insecticides.

γ-hexachlorocyclohexane (γ-HCH) is the main constituent of the technical HCH mixture (figure 5). Since the early 1980's the uses of the technical mixture as insecticide for agricultural use is prohibited in almost all western countries. Therefore, only the 99% cleaned γ-HCH (lindane) is nowadays applied for crop protection under the brand name lindane. Lindane belongs to the most investigated POPs today and can be found in the atmosphere as well as in ocean water during his transport to the Northern regions. As the name reveals, lindane is a chlorinated compound used as insecticide in global dimension. Recent estimations determined that today the Arctic Ocean contains a total amount of about 8000 t lindane. In the late 1980s, the German Hydrographic Institute performed several comprehensive investigations about the water-related transport of POPs into the Arctic waters (GAUL 1992). It could be shown that large amounts of lindane are transported into the North Sea by the big European rivers like Rhine, Weser, Elbe, and Thames. The large central European rivers cover watersheds, collecting surface water from region down to industrial centers of Eastern Europe into the North Sea region via the German bight. From the North Sea the main ocean currents transport the contaminant load along the Norwegian coast into the Arctic basin (figure 5). For lindane average concentration of 5 ng/l were determined for the German bight. However, still 200 pg/l are measured in the surface waters of the northwest Atlantic and the Arctic basin. This 200 pg/l level represent the general γ-HCH contamination level for the water body of the Arctic basin and results from the combined atmospheric and water-related long-range transport of γ-HCH into the Arctic from the source regions (AMAP 1998, Oehme et al. 1996, Bailey et al. 2000). Due to the dominating ocean current from the Atlantic Ocean into the Arctic basin it can be assumed that the main contaminant loads transported with ocean currents are originated from North America, the Arctic Russia and western Europe. Today, g-HCH still belongs to the dominant contaminats in the arctic atmosphere and biosphere (Sinkkonen und Paasivirta 2000).

Polychlorinated Biphenyls (PCBs)

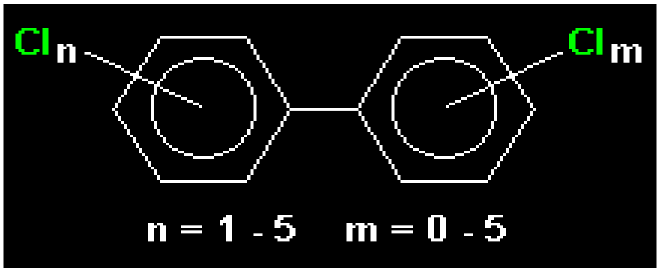


Figure 7: The 209 possible polychlorinated biphenyls (PCBs) differ in number and substitution pattern of the chlorines on the biphenyl structure.

Polychlorinated biphenyl (PCBs) is a mixture of 209 congeners. The PCB mixture is used as flame retardant in paintings, as additive for insulation purposes and dielectric fluid as for example in electric condensators. In general, between 50 to 100 congeners are released into the environment usually during destruction of old condensators, refrigerators and paintings etc. More than 1 000 000 t of various PCB mixtures are reported produced on a global basis. The different PCB congeners contain the same biphenyl structure, but differ in number and substitution pattern of the chlorines in the molecules (figure 7). Due to different synthesis ways in the production of the PCB mixture, The patterns of the technical mixture are different in PCB from Russia, Europe or USA. These characteristic differences can be used to discriminate between source areas. The distribution patterns of the PCB congeners allow also to discriminate between congeners, which are mostly transported via the atmosphere and those who prefer ocean currents as, transport vehicle. Following an air parcel several days and analysing the PCB content, usually a continuous reduction of the concentration level but also a significant change in the PCB patterns depending on the distance to the source region can be observed (Gregor et al. 1996). Mainly low chlorinated PCBs are best fit for atmospheric long-range transport.

Determination of source regions

A typical transport episode is presented in figure 8. The congener specific PCB concentrations in air masses, which moved fast from Western Europe into the Svalbard region, was measured (Oehme 1991). Within 2 days a continental air parcel was traveling from South Norway up to Zeppelin Mountain (Ny-Ålesund). To characteristic sampling

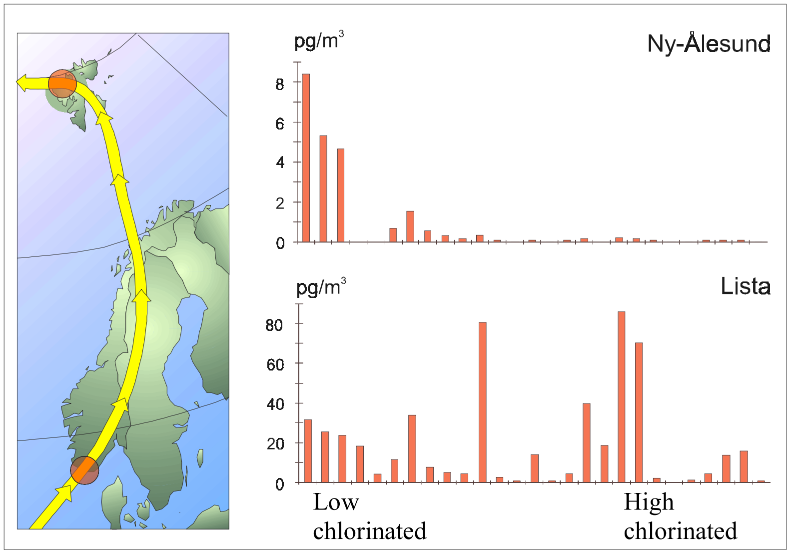


Figure 8: A typical atmospheric tlong-range transport episod. PCBs are transported form central Europe via South norway into the Svalbard archipelago. PCB content in Ambient air from South Norway (Lista fyr) and Zeppelin mountain (Ny-Ålesund, Svalbard) are compared.

sites were chosen to demonstrate the changes in concentration and patters which follow such a transport event. It is very difficult to estimate possible source areas only based on characteristic PCB pattern differences in air samples from the remote sites without. Therefore, for a serious source determination various information sources must be considered and combined.

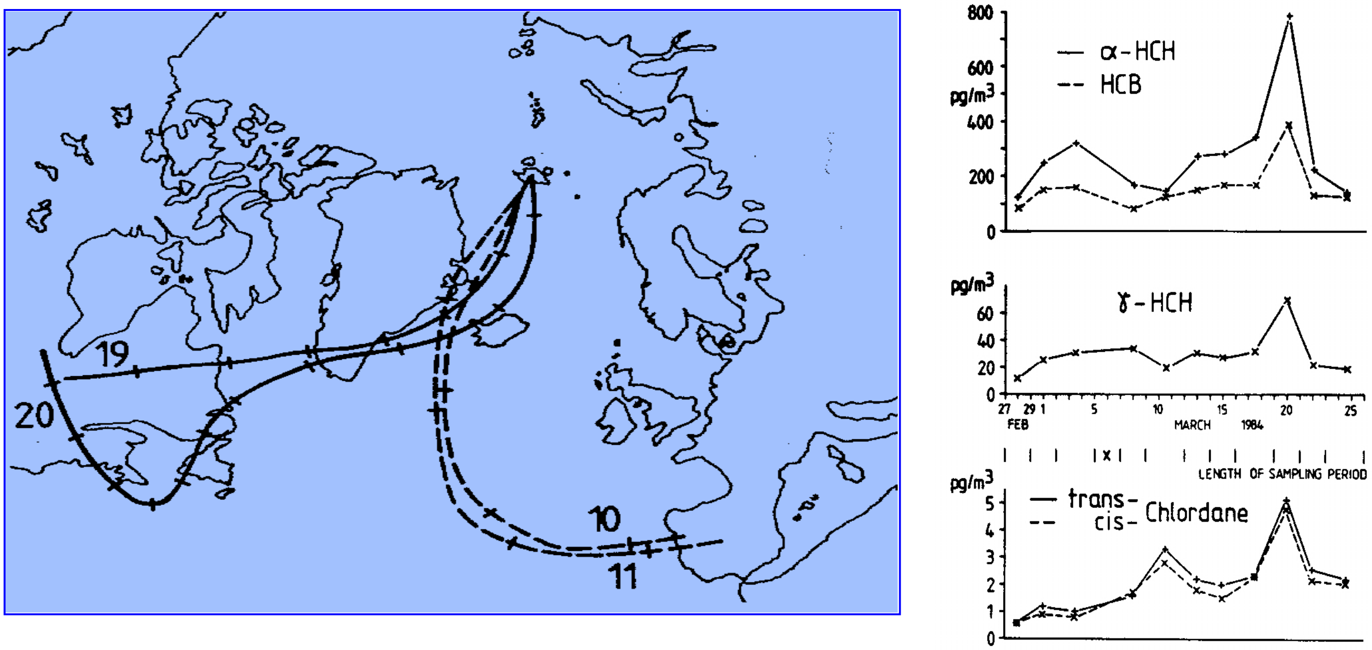


Figure 9: Typical 5-10 days back trajectories calculated for air masses moving in a certain height and velocity into the Svalbard area. Transport episodes 10. and 11. as well as 19. and 20. March 1984 are presented. As it can be derived from the level distribution, for air masses moving from the Americal content into the Svalbard area, elevated chlordane and HCH values can be determined.

- Estimation of meteorological conditions during the transport event (back trajectories).
- POP patterns.
- Concentration levels.
- The relation between different contaminants. The dominance of a certain compound indicates possible source regions.

The best indication of a source region is always the dominance of characteristic compounds known to be used in a certain region. A clear indication is given, when the meteorological calculations support the suspicion (back trajectories). Back trajectories, are theoretical calculations mainly based on empirical data about wind velocity, wind direction, temperature etc., which back calculate the way the air parcel was traveling in the past 5-10 days period prior to sampling. Elevated values of the cyclodiene insecticide chlordane (main constituents of the technical chlordane mixture: *trans-*, *cis-*chlordane, heptachlor, *trans-*, *cis-*nonachlor) which was mainly used on the North American continent were found in air masses over Ny-Ålesund (figure 9). A subsequent determination of the back trajectories confirmed the suspicion; the air parcel was originated from the North American continent (OEHME 1991, Oehme et al. 1996 a, b).

Presence and consequences of persistent contaminants in the Arctic environment

Figure 10: Polar Bears from the Svalbard Reagion and Russia are found to be highly contaminated with persistent organic pollutants.

Current information about considerable levels of persistent pollitants in Arctic organisms like polar bears but also indigenous people like inuits and saami people in the European Arctic are still worrying the scientists dealing with Arctic environmental Research. The AMAP report (AMAP 1998) documents the situation in a comprehensive way. In the meatime, new scientific reports complete the picture with comprehensive and new research results (Pellerin and Grondin 1998, Bidleman 1999, Brunstrom and Halldin 2000, Hansen 2000, MacDonald et al. 2000, Wiberg et al. 2000, Skaare et al. 2000a,b, Kleivane et al. 2000, Borga et al. 2001). New cientific methods were developed in order to include biologicla effect parameters in the comprehensive scientific evaluation of the hazardous potential of environmental pollutants (Henriksen et al. 1998). New results on the contamination status of the Arctic human populations were recently with special focus on the russian Arctic (Polder et al. 1998). For the first time, pseudohermaphrodite polar bears were found within the Svaldbard-Population (Wiig et al. 1998). The presence of pseudohermaphrodite animals in Svalbard were explainded by endocrine dysfunctions within the embryonal development. The endocrine effects were direct correlated with the high burden of persistent organic contaminants in the polar bear popualtion (Colburn et al.1997). Mainly high PCB levels in the polar bears were seen to be responsible for this physiological effects (Bard 2000, Muir 1999, Brunstrom und Hallin 2000, Skaare et al. 2000 a, b, Chui et al. 2000). Thus, for the first direct toxic effects of long-time exposure to long-rang transported halogenatic antopogenic pollutants were found in higher Organisms within the Arctic on a population level.

Already in the 1980s elevated PCB levels in breast milk from inuit mothers were found (Dewailly 1989, AMAP 1998). Thiese first results lead to new comprehensive studies about the possible effects of high POP levels on the daily life of the kanadian Inuits (VanOostdam et al. 1999). Comparable results as earlier found in Canada were also found for Greenland Inuits, were autopsy tissue samples were investigated (Dewailly et al. 1999). Today, it is clear that especially the tradional ways of food consumption by consing products produced from hunted marine mammals (seals, whales) with high fat content result in high POP body burdens. Long-range transported pollutants are deposited in the Arctic environmenta and accumulate through the food-web into high levels in top end organisms (predators like seals, polar bears and whales)

Recent investigations at the Farøy islands confirmed this hypothesis high concentration s of PCBs and trace metals were found in inhabitants mainly consuming meat from pilot whales traditionally hunted at the shores of the Farøy islands. (Weihe et al. 1996).

First studies from the russian Arcitc confirm that also the Arctic human population in Russia are exposed to considerable contaminant concentrations (Klopov et al. 1998, Polder et al. 1998). However,compared to the Greenland and Canadian Inuits the concentration status in Russia is considerably lower.

Bjørnøya, a case study

Within the past decade, multidisciplinary studies have been performed on Bjørnøya (Bear Island, Norway) elucidating the fate and the presence of persistent organic pollutants in this pristine Arctic environment. High concentrations of conventional persistent pollutants like polychlorinated biphenyls (PCB) were found in freshwater, sediments, snow, ice and biota samples reaching the region probably via long-range transport (Skotvold et al. 1998). Subsequently, bioaccumulation throughout the food webs into the top predators resulted in toxic concentration levels. Correlations between high POP burden and reduced physiological fitness were found for glaucous gulls (*Larus hyperboreus*) on Bjørnøya (Bustnes et al. 2000). This species as one of the major predators in the Arctic is a good indicator for contaminant accumulation. Within a comprehensive research programme at Bjørnøya co-ordinated by Akvaplan-niva and funded jointly by the Research Council of Norway and the State Pollution Control Authority of Norway (SFT), samples from selected species belonging to a typical freshwater food web were collected at Lake Ellasjøen and analysed for the content of POPs .In addition, gut samples from glaucous gull were taken and analysed, since guano input from seabirds like gulls into the freshwater system cannot be neglected as possible POP source. High concentration of chlorobornaes, PCBs and chlorinated pesticides were found confirming the special contamination status of the Lake Ellasjøen area at Bjørnøya. Bjørnøya is situated about 500 km North off the Norwegian mainland halfway between Norway and Svalbard (figure 11).

How strong is the influence of atmospheric long-range transport on the POP levels in the Southern Barents Sea region? Where are the sources of the persistent organic pollutants found in the Southern Barents Sea region? Based on information given by the Norwegian Meteorological institute, in average, more than 30% of the summer time, fog events are occurring. Therefore, in addition to ambient air and snow samples, also fog water was collected in order to assess fog as medium for transport and deposition pathways at Bjørnøya. Typical POP distributions in air samples from Bjørnøya (meteorological station) and the Zeppelin Mountain measuring station are presented in figure 12. In general, the POP concentration levels are similar in ambient air collected at the Zeppelin station and at Bjørnøya. HCB is dominating all air samples from both stations. However, the comparison of the PCB patterns reveals remarkable differences.

PCB concentrations in air from the "Zeppelin Mountain" station are low, and the low chlorinated congeners are usually dominating. However, the medium chlorinated PCB congeners dominate the Bjørnøya air samples. Usually, such a PCB pattern is a characteristic feature for biological samples. In addition, the dominance of the medium chlorinated PCBs (PCB 153, 138) is especially expressed in the sample taken in summer 1999 (13. - 15/7-99). During this period of the year, the cliffs, mainly south of Lake

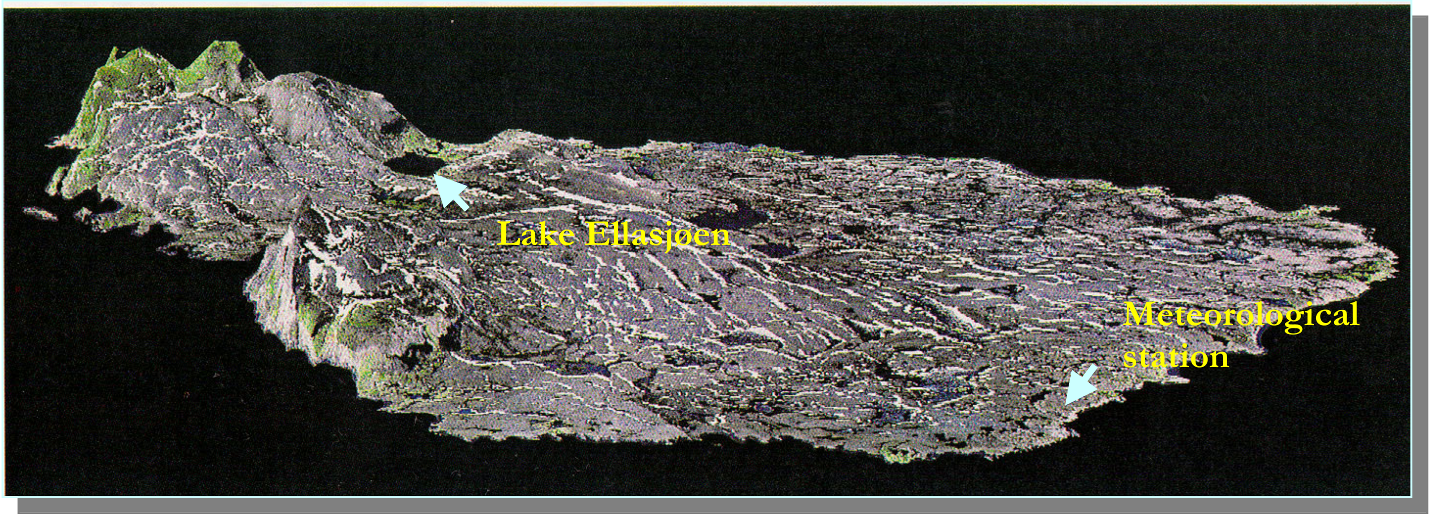


Figure 11: 3-D plot of Bjørnøya (Bear Island) 74° N, 19 ° E. including the Lake Ellasjøen sampling area in the north of the island.

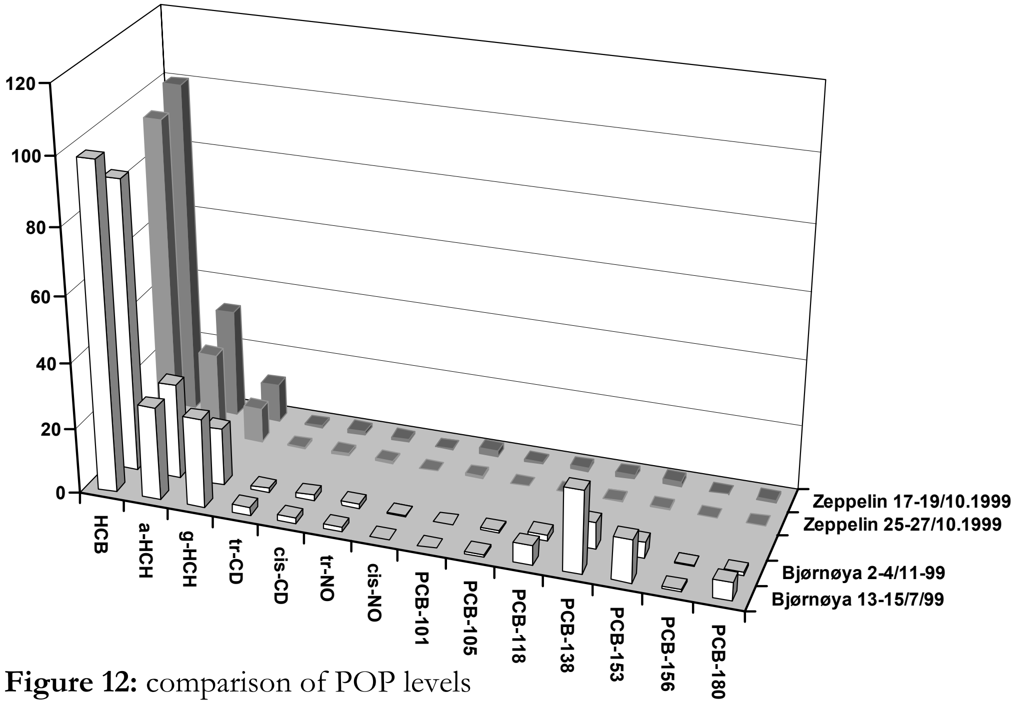


Figure 12: comparison of POP levels in atmospheric samples from Bjørnøya and Svalbard (Zeppelin station)

Ellasjøen, are populated from thousands of nesting seabirds. Therefore, one hypothesis for the unusual POP pattern is that the guano and remainings of seabirds redistributed into the air around Bjørnøya, contribute significantly to the elevated POP air levels at Bjørnøya as the dominating local source during summertime.

Fog water samples collected in Bjørnøya were also dominated by medium chlorinated PCB congeners and indicate the strong influences of local biological

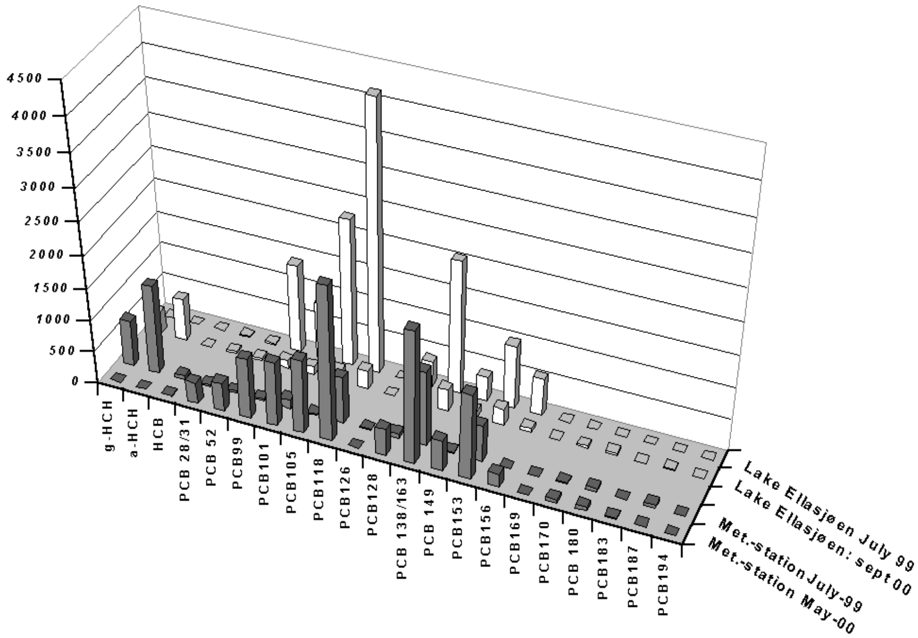


Figure 13: Comparison of POP levels in fog samples from Bjørnøya sampling sites

sources (e.g., seabird guano) on the POP patterns (figure 13). The influence of possible biological sources is also supported by the dominating γ-HCH in Bjørnøya fog. Seasonal differences are even more expressed in fog samples as found for ambient air probably due to the vicinity of bird cliffs and hatching areas in the lake Ellasjøen catchment area. The September sample from lake Ellasjøen (2000) showed low PCB concentration levels, whereas about 10 times higher concentrations of PCB (PCB 118) was found.

Quantification and Monitoring

It is obvious that only long-term monitoring in combination with highly reliably analytical methods can reveal the role of long-range transport as source for contamination in the Arctic. Therefore, AMAP suggest a list of priority compounds for monitoring in Arctic samples. In addition a guideline for trace analytical laboratory was elaborated in 1994 in order to provide high quality data for a subsequent interpretation of the data. Without the political and interdisciplinary co-operation of all Arctic nations as well as scientist working in different research fields (meteorologists, biologists, chemists, physicists, geologists, glaciologists etc.), all these efforts to control the input of pollutants into the Arctic had been of very limited value. The trace analytical determination of persistent pollutants regardless sample material consists in general of four preparation and quantification steps.

1. Sampling and storage
2. Sample preparation
3. Clean-up
4. Analysis and quantification

A quality control protocol must follow every sample to ensure and document the reliability of the data. In case of problems during sample preparation or unusual finding during quantification, the scientist can always double check these documents in order to find hints for possible faults. In addition sufficient field blanks, laboratory blanks must be included in order to show that no contamination occurred during sampling, storage, and sample preparation and clean-up. The recovery rates for all contaminants analysed must be determined for the analytical method used. If the quality documentation do not indicate an artifact, the data must be considered as real high quality values.

Sampling and storage

The importance of the right methods for storage and sampling can not be underestimated. In case of biological samples, fresh sample material must always be preferred. Sampling tissue from organisms found death since weeks must be avoided since microbiological processes in the tissues of the carcass can significantly change the pollutant pattern. In addition, lipids and proteins are rapidly degraded, thus, the tissue composition changes quickly after the death of an organism. Tissues for analysis of POPs should not be taken from organisms death longer that few days to avoid unwanted artifacts in the determination of the POP content. Samples like sediment, soil, water, or ambient air as well as biological material should always be stored frozen until further preparation is been carried out. Ambient air samples are collected adsorbed on solid phases like glass fiber filters/ polyurethane foam plugs or other adsorbent materials (silica, XAD-2 etc.). The sample material is always stored frozen at a minimum of – 20 ºC in order to avoid microbiological degradation on the samples during storage.

Sample preparation

A good sample preparation method must provide the best possible access to the content of contaminants found in the sample material. For sample types like water and ambient air, extraction methods like Soxhlet extraction for the solid materials carrying the air contaminants or liquid/ liquid extraction as well as solid phase extraction for water samples can be employed (figure 14). However, for sediment or biological material an additional step must be included prior to extraction. The sample material must be homogenised to provide a homogenous distribution of the contaminants in the whole sample and also to destroy membranes, cells etc to provide an easy access to the contaminants in the sample during extraction. Sediment and soil samples must be dried prior to further clean up under controlled conditions. Biological tissue samples represents the most difficult sample materials an environmental analyst has to cope with. Homogenisation and extraction must be modified and optimised depending on the sample. Homogenisation is carried out directly on the wet tissue for further 'wet extraction' or a high excess of precleaned sodiumsulfate (Na_2SO_4) is added for both homogenisation, drying and destruction of cellular material during homogenisation in an analytical mill or comparable instrument. Other methods like soapification, freeze-drying etc are described in the literature. The homogenated material will be further extracted by using suitable methods like Soxhlet extraction, cold column extraction, accelerated solvent extraction, micro wave assisted extraction etc., depending on the sample matrix and the contaminants to be anaysed.

Clean up

A variety of tissue components have the potential to disturb the correct quantification of a sample extract. Therefore, further clean up is essential to provide correct data about pollutant levels in the respective material. Usually chromatographic methods are employed for the clean up of the sample extract (figure 10). If biological material is involved (biological tissues, plankton samples etc) often so-called gel permeation chromatography is used since this method is especially developed for the removal of lipids and proteins from a sample extract. For sediment, soil, water or ambient air sample extracts, standard column chromatographic clean-up steps are sufficient to provide good results. Only, if these methods do not result in sufficiently cleaned eluates, sulfuric acid treatments can be included. However, since the treatment with sulfuric acid can also destroy a large number of contaminants, the application of this method must be carefully considered.

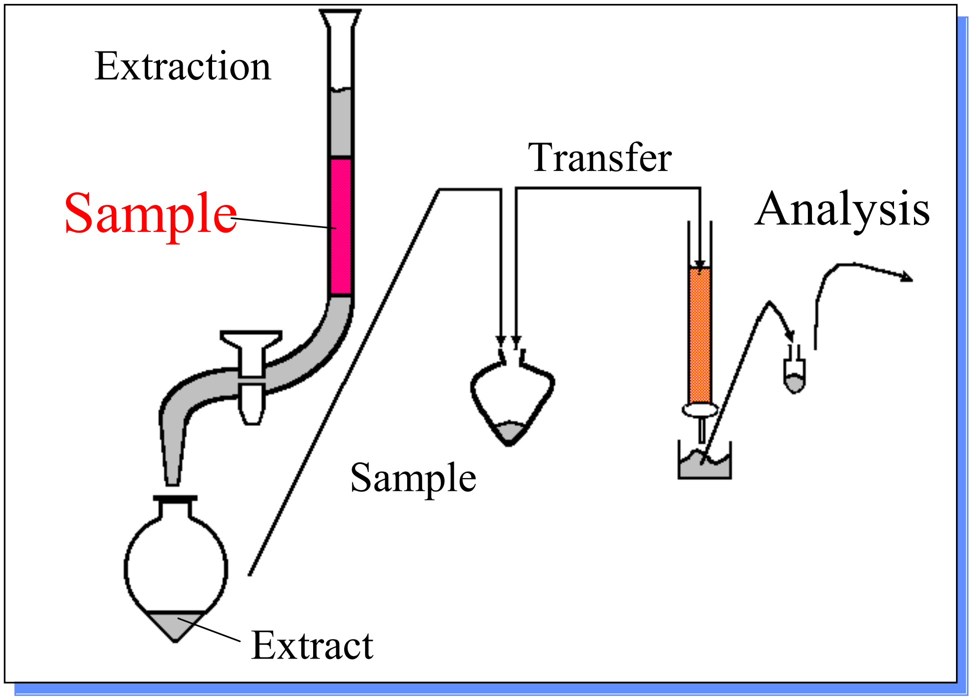


Figure 14: Scheme for sample preparation and clean-up prior to analysis . Several chromatographic steps arre involved in order to provide clean sample eluates.

Analysis and quantification

For analysis of the clean sample eluate, also chromatographic methods in combination with high sensitive detectors are usually employed. Today mainly high performance liquid chromatographic (HPLC) separation and gas chromatographic (GC) methods are used depending on the physico-chemical properties of the contaminants investigated. For the determination of chlorinated semi-volatile persistent pollutants (PCBs, chlorinated pesticides and metabolites) gas chormatographic methods are best suited. Various detectors are available for the determination of this type of contaminant. A halogen sensitive detector like the electron capture detector is mostly used for the determination of chlorinated pollutants. However, since the compounds only can be identified by comparing elution times of standard and sample, for security reasons two independent GC-analysis should be carried out on two different types of capillary columns (quality assurance). One of the best suitable detectors for the gas chromatographic determination and quantification of semi-volatile contaminants is the mass spectrometric detector (MS). In addition to elution time mass specific information are derived by this method. Thus, only one GC-run is needed to determine the concentration of the respective compound.

HPLC methods in combination with spectroscopic detectors (diode array detector, UV- detector, and mass spectrometric detector) are used to determine polar, water soluble compounds. However, due to the poor sensitivity of these detectors, the determination of ultra-trace concentrations is often impossible with these methods.

A liquid or gas chromatographic separation and a subsequent detection with suitable detector results in a so-called chromatogram (see figure 15). Prior to analysis, the skilled analyst has determined the response of the single compounds, detection limits, limits of quantifications. Based on this information he has calculated response factors and determined the linearity range of the compounds to be analysed with the respective detection method. The height or the calculated are of the single peak is directly proportional to a certain concentration of the contaminants. Thus, he has sufficient information to calculate the concentration levels of the single compound. The concentration values are basis for further considerations about biomagnification, accumulation, toxicity in co-operation with other research groups (see the following sections).

Monitoring

Based on the above given considerations, it should be clear, the priority list given by AMAP reflects not necessarily the most important contaminants to be monitored in the Arctic environment (table 1). This list is more to be seen as a mirror for the

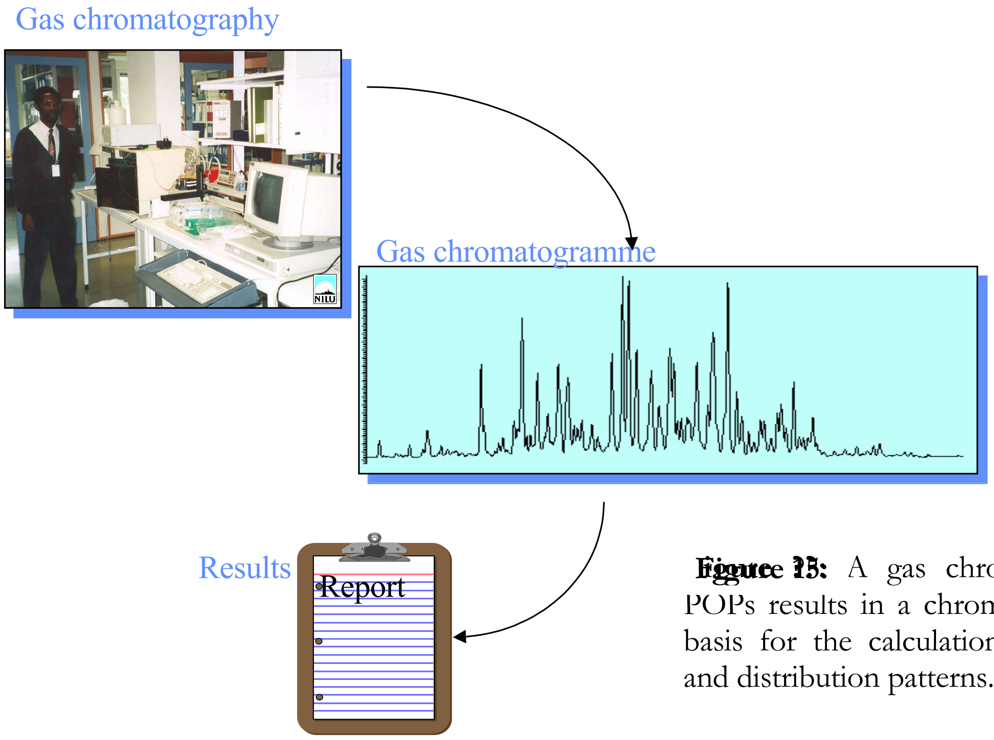


Figure 15: A gas chromatographic analysis of POPs results in a chromatogramme which is the basis for the calculation of concentration levels and distribution patterns.

possibilities and limitations modern trace-analytical methods are facing today. Only those compounds, analytical chemists are able to analyse and monitor with a sufficient quality are listed here. Therefore, one can not assume that the compounds represented here are covering all relevant contaminants to be found in the Arctic. A realistic conclusion should be, all those compounds are listed on the AMAP list which can be analysed in trace amounts and are important contaminants in the Arctic environment. Therefore, comprehensive monitoring programmes like AMAP reflect the state-of-the-art in trace analysis and must be further expanded if new methods prove the presence of new compounds in the regions.

Group	***Individual Compound***
Chlorobenzenes	Hexachlorobenzene (HCB), Pentachlorobenzene, Tetrachlorobenzene
Hexachlorocyclohexanes	α–,β–,γ–HCH
Chlordanes	*cis-/trans*-chlordane, *cis-/trans*-nonachlor, oxychlordane, heptachlor heptachlor-*endo-exo*-epoxide
DDT	4,4'-DDE,-DDD,-DDT 2,4'-DDE,-DDD,-DDT
Mirex	Mirex
Toxaphene	Toxaphene
Dieldrin	Dieldrin, endrin
PCDD/F	2,3,7,8-tetra- to octachlorodibenzo-p-dioxins and furans
PCBs	30 selected polychlorinated PCBs specified in the AMAP report
Current used pesticides	Atrazine, chlorpyrifos, α–/γ-endosulfan, pentachloroanisole.
Other POPs	Pentachlorophenole, Brominated flame retardants, Polychlorinated diphenyl ethers (PCDPEs), PCB sulfones
Polychlorinated naphthalenes	PCN
Short-chain chlorinated paraffins	C10-C13 chlorinated alkanes

Table 1: Detailed list of POPs to be analysed under the AMAP monitoring programme, adapted according AMAP II draft version.

Biomagnification in the food web

Once transported to the Arctic POPs are taken up by organisms in Arctic food chains. Being highly persistent, the lipophilic POPs tend to bioaccumulate in body lipids (MacDonalds and Bewers 1996). Due to the high seasonal variation in primary production and food availability, organisms living in the Arctic region accumulate lipid reserves for energy storage. The lipids are mobilised at times of food shortage, resulting in a seasonal variation in the organisms lipid composition and quantity, and a redistribution of the POPs within the organisms (Henriksen et al. 1996, Jørgensen et al. 1997). Transfer of lipids and energy through the marine Arctic food chains is rapid (Falk-Petersen et al. 1990). Lipids originating in the spring bloom can be traced in the secondary consumer capelin within six months. Thus, there is a great potential for transfer of lipid-associated POPs through the food chains in the Arctic. The main mode of entry of POPs in the terrestrial food chain seems to be atmospheric deposition on the surfaces of plants followed by uptake by herbivores. However, the levels of POPs in organisms higher in the terrestrial food chain, such as Arctic caribou and ptarmigan, are low (Elkin 1993, Severinsen and Skaare 1997). In the aquatic environment, the mode of uptake and elimination of POPs in the lower end of the food chains (phytoplankton, zooplankton, fish) is mainly by passive diffusion across the respiratory surfaces (Thomann 1989). However, since the most lipophilic compounds are found adsorbed to particulate matter in the water, uptake of POPs through the diet is also considered as an important way of entry in zooplankton and fish. In the upper end of the marine food chains (seabirds and marine mammals) uptake of POPs is exclusively through the diet. Lower aquatic organisms may release POPs by passive diffusion to maintain an equilibrium with the POPs in the environment (Thomann, 1989). Seabirds, marine mammals and terrestrial species, however, can eliminate POPs only by metabolism followed by excretion, or by transfer to their offspring through the placenta, lipid rich milk or egg (Addison 1982, Nakata et al. 1995). In contrast to terrestrial food chains, high levels of POPs have been found in the top predators of marine food chains such as Arctic fox, glaucous gull (figure 12) and polar bear. The data available for Arctic fish have shown levels which are two to three orders of magnitude lower than in birds and mammals, indicating a very high biomagnification factor between these two trophic levels in the Arctic. Indigenous peoples, who depend heavily on lipid rich food from the sea, occupy the niche of top-predators in the marine ecosystem and are therefore especially vulnerable to biomagnifying contaminants. Elevated levels are found and are further evidence that POPs not only can be transported to and accumulated in remote area but also may pose a health risk for people living from organisms on top of marine food chains (figure 16).

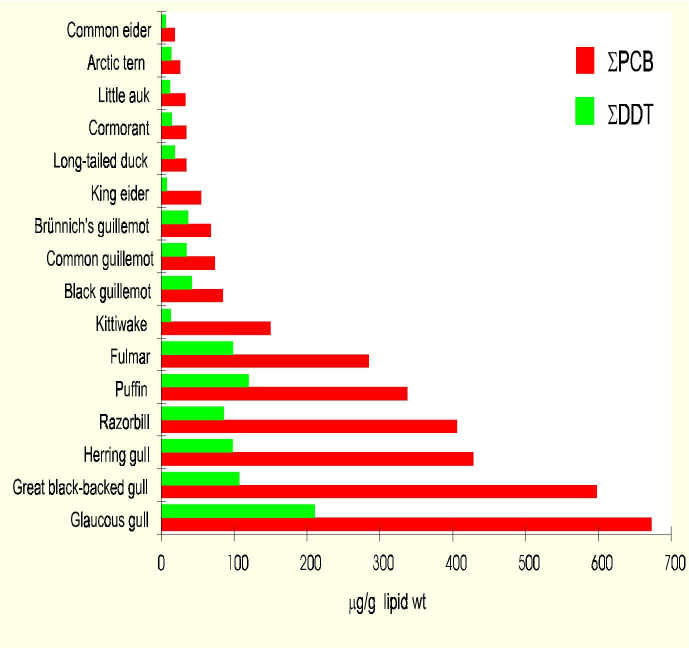


Figure 16: Levels of PCBs and DDTs in Arctic birds reflecting the differences in accumulation pattern and trophic levels (according to GABRIELSEN 1998).

Effects

The presence of organochlorines in Arctic ecosystems in general, and in many top-predators including humans in particular, has become a cause for concern. Due to their lipophilic character, POPs can easily cross cell membranes and thereby activate specific biological processes by binding to arylhydrocarbon (Ah)-, cholinergic- and/or steroid-receptors.

Binding of POPs to the Ah-reseptor may lead to increased activity (induction) of specific enzymes in the hepatic P450 (CYP-family) system, and thereby metabolisation of the pollutant. Aliphatic and simple aromatic hydrocarbons are readily metabolised, whereas polyaromatic and polychlorinated compounds are metabolised much slower and thus are accumulated (Varanasi 1989). Metabolisation of PAHs may have serious consequences for the animal since toxic and carcinogenic derivatives are formed. Of the different POPs that have been detected, PCB is by far the one that has been found in highest concentrations. The affinity of PCB for the Ah-receptor raises serious concern for possible immunotoxic effects, since such effects are mediated through the binding of the pollutant to the Ah-receptor (Safe 1984). Invertebrates have very limited capacity to metabolise POPs, and toxic effects are only manifested when they are eaten by vertebrates. The dose-dependent induction of CYP1A enzymes by POPs, which has been demonstrated in both fish (Goksøyr 1995), fish-eating sea birds (Walker 1992), as well as in seals, whales and polar bears (Norstrom and Muir 1994, Wolkers et al. 1998), may become a useful diagnostic tool for environmental pollutant biomonitoring in aquatic ecosystems.

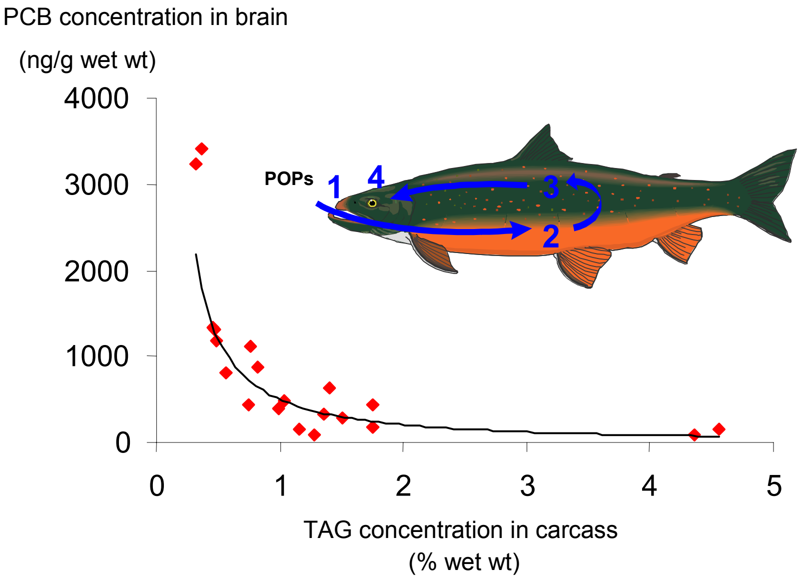


Figure 17: Redistribution of PCB (congeners 101.105.153,180) in Arctic char (*Salvelinus alpinus*) brain following the mobilisation of lipids during starvation (according Foshaug et al. 1998)

Although the levels of some POPs detected in top-predators may exceed the thresholds associated with reproductive disturbance and immunosuppression in laboratory animals (AMAP 1998); there is still a lack of knowledge about the possible threats imposed by POPs in Arctic ecosystems. The temporal changes in lipid deposition and mobilisation may have implications for toxicokinetic mechanisms of deposited POPs and in both fish and fish-eating sea birds, emaciation has been shown to be accompanied by a strong redistribution (figure 17) of lipophilic pollutants from adipose tissues to more sensitive tissues (*e.g.* the brain) (Henriksen et al. 1996, Jørgensen et al. 1997). Arctic organisms may hence be considered to be extra sensitive to POPs with high lipophilicity. The dependency of Arctic animals of deposited lipids for reproduction may facilitate the transference of POPs to the gonads. Since many POPs have the ability to bind to the oestrogen receptor and to interfere with many important enzyme-regulated steps in the biosynthesis of hormones, reproduction in Arctic animals may be particularly vulnerable.

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