POPS IN THE BALTIC

A review of Persistent Organic Pollutants (POPs) in the Baltic Sea

POPs IN THE BALTIC

A review of persistent organic pollutants (POPs) in the Baltic Sea

Authors:
Michelle Allsopp
Bea Erry
David Santillo
Paul Johnston

Greenpeace Research Laboratories
Department of Biological Sciences
University of Exeter
Prince of Wales Road
Exeter EX4 4PS
UK
www.greenpeace.org

The writing of this report was funded by Greenpeace International

ISBN: 90-73361-71-0

This report is printed on 100% recycled processed chlorine-free paper

April 2001

TABLE OF CONTENTS

SUMMARY

Persistent organic pollutants (POPs) are a group of chemicals which are very resistant to natural breakdown processes and are therefore extremely stable and long-lived. POPs are not only persistent in the environment but many are also highly toxic and build up (bioaccumulate) in the tissues of animals and humans. Most do not occur in nature but are synthetic chemicals released as a result of anthropogenic activities. Vast amounts of POPs have been released into the environment and due to long-distance transport on air currents, POPs have become widespread pollutants and now represent a global contamination problem.

In the Baltic and in other regions, certain POPs have been responsible for some catastrophic impacts upon wildlife, ranging from interference with sexual characteristics to dramatic population losses. POPs are suspected of causing a broad range of adverse health impacts in humans and there is evidence that current levels of POPs in women in the general population of some countries is sufficient to cause subtle, undesirable effects in their babies due to transfer of these contaminants across the placenta and via breast milk.

In recent decades, numerous POPs have been produced in large quantities worldwide and many are still in production and use. Some POPs, such as dioxins and furans, are not produced intentionally but are generated as by-products of many industrial processes, particularly combustion processes and industrial processes involving chlorine chemistry. Several POPs, notably certain organochlorine pesticides such as DDT and technical grade HCH, have been completely banned in industrialised countries and banned from agricultural use in most of the less industrialised countries. Due to the persistence of these pesticides, however, high levels remain in many regions of the globe. In the Baltic, there have been substantial inputs of POPs into the sea from numerous sources over the past 50 years, including discharges from industry, such as effluent containing organochlorines from pulp and paper mills, agricultural run-off, marine paints, and dumping of wastes. The Baltic Sea and its biota consequently became highly polluted and even today, levels of POPs remain comparatively high. Presently, the major contributor to dioxin releases to atmosphere in the Baltic region is considered to be the incineration of wastes, in particular municipal solid waste incineration.

This report draws together published scientific literature on levels of POPs that have been recorded in the sediments and biota of the Baltic Sea and studies of their possible impacts upon the highly specialised ecosystem of the region. It also documents studies on the levels of POPs recorded in humans from Baltic nations and of possible effects upon health. The report reveals that the Baltic environment is still suffering from contamination by POPs and that adverse impacts upon the health of some of its animal species are still occurring. With these factors in mind the report demonstrates that there is an urgent need for the complete phase out of all releases of POPs to the environment.

What are POPs?

POPs encompass many different and varied groups of man-made chemicals. Some POPs have been listed by national and international organisations as being chemicals of concern. For instance, the United Nations Environment Program (UNEP) has listed certain POPs, which are organochlorines, as being chemicals of clear concern. Organochlorines are substances containing chemically combined chlorine and carbon. This is a huge group of chemicals that includes many POPs. The UNEP list comprises 12 organochlorines – known as the dirty dozen. These are: the chlorinated dioxins (PCDDs) and chlorinated dibenzofurans (PCDFs) - chemicals that are formed as unintentional by-products of combustion and processes involving the manufacture, use and disposal of organochlorines. For example, they are produced as by-products of municipal waste incineration and other types of incineration, by open burning of wastes, in landfill fires and during the production of the chlorine containing plastic PVC as well as in other industrial processes involving chlorine chemistry; PCBs - industrial chemicals that have been banned but are still released to the environment in significant amounts from old sources and as unintentional by products of combustion and from processes involving the manufacture, use and disposal of organochlorines; HCB – a chemical used as a pesticide, as an intermediate in the manufacture of pesticides and produced as an unwanted by-product of various industrial processes involving organochlorines; organochlorine pesticides, including- DDT, chlordane, toxaphene, dieldrin, aldrin, endrin, heptachlor and mirex. Use of these organochlorine pesticides is banned or is severely restricted in most countries, but not in all.

POPs included in the above list are of immense concern given that they are ubiquitous environmental contaminants and are demonstrably toxic. Most research on POPs is limited, however to a few of these chemicals only. There are numerous other POPs which are also widespread environmental contaminants and are, therefore, also of great concern. These include pentachlorophenol, brominated flame retardants, HCH isomers - such as the organochlorine pesticide lindane, organotin compounds (used as anti-fouling agents for ships), short chained chlorinated paraffins (used in cutting oils and lubricants) and certain phthalates – DBP and DEHP, which are not particularly persistent but are nonetheless hazardous (main uses are as plastic softeners, especially in PVC).

Where are they Found?

All environmental media can become contaminated by POPs once they are released into the environment. For instance, spraying persistent pesticides onto crops can contaminate vegetation and soils, direct discharges from POPs manufacturing facilities may contaminate rivers and releases of POPs from the stacks of incinerators and industrial facilities contaminate air. Consequently, POPs can contaminate local areas close to where they are released. However, some POPs are volatile/semivolatile and may evaporate from soil or water to air. Subsequently, they may be transported for thousands of kilometres on air currents and contaminate regions remote from their source. These POPs migrate on air currents from warmer regions of the globe towards colder polar regions. Once they reach colder temperatures they condense and are deposited again on the Earth’s surface. POPs may also be transported for long distances by rivers, ocean currents and as contaminants in wildlife. Due to the extensive releases of POPs and long distance transport they have become global contaminants and even attain high levels in remote regions, such as the arctic.

POPs in Food Webs

Many POPs which pollute the environment become incorporated into food webs. They accumulate and persist in the fatty tissues of animals and humans because they are soluble in fats and are not easily broken down in the body. Even low environmental levels of POPs can lead to high levels in the body tissues of animals and humans. For many POPs, the levels in fat increase as one animal eats another, so that the highest levels are found in predator animals at the top of food webs, such as polar bears, seals, toothed whales, birds of prey and humans. Marine mammals accumulate particularly high levels of POPs because of their large quantities of fatty blubber and a reduced capacity to break down some POPs compared to other species.

POPs in the Baltic Environment

The Baltic is a semi-enclosed sea which has large inputs of freshwater from rivers making it brackish. The Baltic became a toxic hotspot of chemical pollution during the 1960s and 1970s. At this time high levels of POPs, notably persistent organochlorines such as PCBs and DDT, threatened wildlife populations. Since that time, levels of some persistent organochlorines have decreased significantly, but today POPs contamination of the Baltic Sea environment still remains comparatively high.

This report presents and reviews some of published data on levels of POPs in fish, birds, and marine mammals from the Baltic Sea and in humans from Baltic nations. It is important to note that detailed comparisons between POPs levels reported in different studies is difficult because of inconsistencies in laboratory methods used and different laboratory quality control standards applied historically. Nevertheless, comparison between studies can give insight into the state of contamination of an area and whether levels of POPs are considered to be high or low. In the Baltic marine environment, comparison of levels of POPs with studies from marine environments in other regions indicates that contamination with these substances is high.

The most commonly monitored POPs in wildlife and humans are, in general, the dioxins, the PCBs and DDT, followed by other organochlorine pesticides. It is only more recently that other POPs, such as the organotins, and brominated flame retardants (PBDEs) have been monitored more routinely. These studies showed PBDEs to be detectable in the tissues of marine fish and seals and in human milk. Unlike most of the persistent organochlorines which have declined in human milk in recent years, levels of PBDEs in human milk have been found to be increasing exponentially and are consequently of great concern.

Numerous other hazardous chemicals, which can be considered to be POPs, are known to pollute the environment and yet others remain unidentified and their toxicity, therefore, unknown. Over the past decade, some new POPs have been identified in seal blubber from the Baltic that had previously never been reported before in wildlife. These POPs include part brominated PCBs, (possibly derived from municipal waste incineration or from contamination of PCB technical formulations). In addition, bis(4-chlorophenyl) sulfone (BCPS), (possibly occurring in the environment due to the production of high temperature polymers) has been isolated and identified. Another organochlorine, octachlorostyrene, was identified for the first time as a contaminant in human blood in Germany. In addition to these chemicals, other POPs identified recently in the environment, including the Baltic region, are polychlorinated diphenyl ethers (PCDEs) in Baltic marine birds, seals, harbour porpoises, and human tissues; tris(4-chlorophenyl) methane and tris(4-chlorophenyl) methanol (possibly a contaminant in technical grade DDT), in Baltic birds and seals and, an organochlorine compound, Q1, in Baltic seals. Little is known about the possible impacts that most of these chemicals could be having on Baltic wildlife.

Marine Sediments

On entering marine ecosystems, many POPs quickly become bound to particulate matter in the water and, in time, sink to the sea bed to be deposited in sediment. In this way sediments act as a sink for POPs deposited in the marine environment.

Research on Baltic Sea sediments has demonstrated that in general, higher levels of dioxins (PCDD/Fs), PCBs and persistent organochlorine pesticides are found compared to sediments from other marine environments. For example, levels of organochlorine pesticides were between about 5 and 30 times higher in Baltic sediments than in the Mediterranean and the Gulf of Alaska. Levels of dioxins and PCBs were higher in Baltic sediments than North Sea sediments. Of the organochlorines, PCBs were reported to be the most abundant compounds. Other POPs identified in Baltic Sea sediments were PCNs, and in German marinas, high levels of organotins.

Studies on sediment cores have indicated that trends of PCB levels with time follow a similar pattern to trends in wildlife. Levels increased from 1940 to 1970, and thereafter decreased until around 1990 when levels increased slightly. Levels of PCDD/Fs also increased after 1950 to reach substantial levels between 1970-85.

Marine Fish

Many POPs have been detected in marine fish from the Baltic including dioxins, PCBs, organochlorine pesticides, brominated flame retardants (PBDEs), TBT and PCNs and certain nitromusks. The highest levels of dioxins, PCBs, DDTs and HCB were evident in fish from the southern Baltic and/or north eastern Baltic. For dioxins and PCBs, high levels in fish have been found in regions subject to inputs from heavily industrialised regions. It has been suggested that local inputs of PCBs have occurred from the Estonian and Polish coasts even in recent years. Illegal use or improper storage of DDT among former USSR countries has been suggested as responsible for high levels of DDT in fish from the Gulf of Riga.

Declines in the levels of dioxins, PCBs and DDT have occurred in some fish from the Baltic Sea over the past 30 years. PCBs have decreased to about 15% of levels in the early 1970s and DDT to 5% of 1970s levels in herring. In cod, however, a decline in PCB levels is not so clear since the early 1970s although levels of DDT have declined. Little or no downward trend is generally apparent for HCB in recent years after initial declines were recorded in fish during the 1970s and 80s. Significantly, this implies a continued input of HCB into the Baltic.

Moreover despite the declines in concentrations of a few organochlorine compounds in Baltic fish, reproductive disturbances are still found in some species which are considered to be associated with pollution. Over the past 10 years, the incidence of a disease in fish known as M74 syndrome has increased, possibly in association with elevated levels of organochlorines. In addition, adverse impacts on Baltic fish are known to have occurred in the past near to discharges of bleached pulp and paper mill effluents which contained chlorinated wastes. Although these impacts have been mitigated somewhat due to changes in pulp bleaching technology, a shift to totally chlorine free (TCF) technology to facilitate closed cycle pulp mill operation and zero effluent discharge is necessary to completely eliminate adverse impacts of pulp effluent on fish.

Marine Birds

Particularly high levels of DDT and PCBs in some Baltic fish-eating birds resulted in severe population crashes during the 1960s and 1970s. DDT and its metabolites, for example, caused egg-shell thinning in guillemots and white-tailed sea eagles. Despite a substantial decline in levels of these compounds over the past 30 years in seabird and bird of prey populations, accompanied by some recovery in population sizes, adverse impacts are still evident in birds today. The thickness of guillemot egg-shells is presently less than that reported prior to 1950. High levels of PCBs and DDT are presently found in the tissues of white-tailed sea eagles inhabiting coastal regions as compared to inland birds and it has been suggested that dioxin-like PCBs are possibly the main contaminants responsible for the continued reduced rate of reproductive success in these birds.

Marine Mammals

Seals

In general, extremely high levels of persistent organochlorine chemicals have been detected in seals from the Baltic Sea in comparison to other seas. Research on samples from seals collected during the 1980s established that levels of PCBs, DDT, chlordane, toxaphene and HCH isomers were significantly higher in Baltic seals than in their counterparts from the Swedish west coast. Among the different seal species in the Baltic, namely ringed seals, harbour seals and grey seals, levels of PCBs and dioxins differed, possibly as a result of their differing feeding habits and/or differences in their ability to metabolise and excrete these contaminants.

Levels of DDT in Baltic seal species have declined continuously since the early 1970s, continuing through the 1980s and 90s. PCBs also declined in ringed seals during the 1970s and 80s, but the decline of PCBs in grey seals from 1969 to 1997 was very slow. Levels of dioxins in Baltic seals during the 1990s do not appear to have declined significantly from 1980s levels.

Baltic ringed and grey seals were severely affected by a disease-complex, known as hyperadrenocorticism during the 1970s and 80s and their numbers fell sharply. The disease complex resulted in reproductive failure, adverse effects on several organs, deformities of the claws and bone loss in the jaw. The disease complex is thought to be associated with hormonal imbalance and with suppression of the immune system induced by organochlorines, in particular PCBs. Recent research shows that grey seal numbers have increased. This has occurred concomitantly with an improvement in reproductive ability, alleviation of some symptoms of the disease complex and a decrease in tissue levels of persistent organochlorines. However, colonic ulcers, which can be fatal, are increasing in the seals. This is possibly due to the impact of “new” contaminants.

Baltic harbour porpoises

Levels of dioxins, PCBs and DDTs in Baltic harbour porpoises were significantly higher than in harbour porpoises from the less contaminated Kattegat-Skagerrak seas and the west coast of Norway. It is not possible to accurately determine trends of dioxin and PCB contaminant levels over time since the data are not sufficient for this purpose. DDT levels, however, appear to have declined since the mid 1970s.

The Baltic harbour porpoise population has drastically declined since the 1940s possibly as a result of previous hunting and due to the impacts of high tissue levels of persistent organochlorines. A recent study noted that the dioxin, PCB and DDT levels recorded in Baltic porpoises continue to be a cause for serious concern. Experimental data indicate that tissue levels of organochlorines in harbour porpoises are in the range where adverse impacts on the immune system and the nervous system would be expected to occur. Moreover, *post mortem* research on Baltic porpoises collected between 1991 and 1997 suggests that high tissue levels of organochlorines could have played a part in the high prevalence of parasitic and bacterial infections apparent in these animals.

Humans

Levels of dioxins in human milk in Denmark, Germany, Lithuania, Sweden and northern Russia, and levels PCBs in human milk in Denmark, Finland, Germany and the Russian Federation have been reported as similar to levels found in most other industrialised countries. Levels of dioxins and PCBs have been reported as decreasing since the late 1980s in Germany and Sweden by some 50% or more. However, the rate of decline of these compounds is very slow and consequently they may remain of concern for some decades into the future. Moreover, current levels of dioxins and PCBs in human milk are still a matter for concern because of the comparatively high exposure of breast-fed infants.

Levels of DDT, HCB and β-HCH in human milk were notably higher in northern Russia as compared to Denmark, Sweden and Germany. A downward trend in the levels of HCB and β-HCH has been recorded in several Baltic countries since the mid-1980s and levels of DDT have decreased considerably since the 1970s.

Edible fish from the Baltic Sea are more highly contaminated with persistent organochlorines than fish from other regions. Studies show, unsurprisingly, that dietary intake of fatty fish from the Baltic is associated with an increase of persistent organochlorine concentrations in human tissues. Levels of dioxins, PCBs and DDT were reported to be elevated in individuals who consumed large quantities of Baltic fish. These included fishermen from the east coast of Sweden, the Gulf of Finland and Latvia who consumed moderate to high amounts of Baltic fatty fish in their diet.

Concern about the possible impacts of elevated levels of persistent organochlorines on human health, particularly for the developing foetus and breast-fed infant, has prompted the Swedish National Food Administration to recommend only a very limited amount of Baltic fish in the diet. Concern has also prompted research to be carried out into the potential health impacts of a high dietary intake of Baltic Sea fish amongst Swedish east-coast fishers and their families. These studies have assessed a number of different markers of health which previous data have indicated may be adversely affected by exposure to organochlorines in the diet. Dietary exposure to organochlorines in the fisher's families was investigated by determining the amount of Baltic fish consumed, and in some instances, by the more accurate method of analysing levels of organochlorines present in blood. The results from east coast fishing families were compared with data collected on west coast fishing families and/or on the general Swedish population. Comparison with west coast fisher's families was particularly relevant because these individuals have a similar lifestyle and consume similar amounts of fish to the east coast families, but the fish from the west coast (not on the Baltic Sea) is less contaminated with persistent organochlorines. Results and conclusions of these highly important studies are summarised below:

- Fishermen's wives and sisters from the Swedish east coast gave birth to babies with significantly lower birth weights. There was also an increased incidence of low birth weight *per se*, defined as <3000g. It was concluded that a high intake of organochlorine contaminated fish from the Baltic Sea may cause intra-uterine growth retardation.
- There was a greater risk of increased time to first pregnancy among east coast fishermen's wives and an increased risk of infertility. Both effects, however, were only evident in heavy smokers. It was concluded that there may be a negative association between fertility and exposure to persistent organochlorines but more research is needed.
- No evidence was found for increased exposure to persistent organochlorines through fish consumption by fishermen's wives and increased incidence of miscarriage or congenital malformations (birth defects).
- No evidence was found for long-term impacts on intelligence (specifically, psychometric impairment) of boys born into fishermen's families by the time they were aged 18.
- A decreased overall mortality from cancer was associated with high consumption of Baltic fish contaminated with organochlorines, and was also associated with high consumption of less contaminated fish from the among west coast fishermen. However, high consumption of Baltic fish among east coast fishermen was associated with an increased incidence in stomach cancer and skin cancer.
- Increased mortality from breast cancer, and increased incidence of breast cancer and cervical cancer were found in east coast fishermen's wives.
- A study on east coast fishermen showed that higher fish consumption and higher blood levels of PCBs and DDTs were associated with lower levels of certain cells of the immune system (natural killer cells). However, a subsequent study on Latvian fishermen who consumed high amounts of Baltic fish found no effect on natural killer cell populations.

Conclusions

1. There have been substantial decreases in some persistent organochlorines such as DDT and PCBs in fish, birds and marine mammals of the Baltic Sea over the past 30 years, but overall, levels remain high. Also, in many cases the decline of some persistent organochlorines has slowed or stabilised in more recent years, a phenomenon which most likely relates to the persistent nature of these compounds and/or their continued input into the Baltic marine environment. The decline of PCBs and dioxins has been particularly slow in some species. The detection of more recently or newly identified POPs in marine biota, such as brominated flame retardants, is of great concern.

2. Despite declines in levels of persistent organochlorines, the levels of POPs recorded in Baltic marine wildlife species remain comparatively high today. Many adverse effects that are associated with exposure to persistent organochlorines are still evident in Baltic wildlife.

3. Levels of persistent organochlorines in humans from Baltic nations are not considered to be markedly higher than in other industrialised nations with the exception of northern Russia. Here, comparatively high levels of DDT, HCH and HCB were recorded in human milk. However, consumption of fatty fish from the Baltic Sea represents an important source of exposure to persistent organochlorines in humans. Consumption of high amounts of fatty Baltic fish in the diet has been associated with several possible health effects in adults and with reduced growth of the foetus in the womb (intrauterine growth retardation).

POPs – A Global Problem

The problem of global POPs contamination is set to continue because the majority of POPs from anthropogenic activities are still being released into the environment. Decreases in the levels of those POPs which have already been banned in some countries gives no room for optimism or for complacency. Levels of POPs are still high enough to be of concern, and moreover, levels of other POPs which are still widely produced, such as the brominated flame retardants and organotins, add to the already heavy burden of POPs. Because the release of POPs into the environment is continuing, there is a potential for further severe impacts on the health of wildlife and humans. Given the persistent nature of POPs there is only one way forward in order to safeguard the environment and future generations. Simply, this is to phase out the production and use of all POPs, and the processes that lead to the unintentional generation of POPs as by-products. Nothing less than an international program to achieve this and to implement clean production technologies is required. Action must be taken now to address the existing POPs problems, prevent new problems and start on the road to a Toxics - Free Future.

Greenpeace Demands

- The production and use of all POPs, and human activities that lead to the generation of POPs, must be phased out at an international and, ultimately, at a global level.

- This must be achieved through the substitution of POPs (or the processes and materials which generate them) with non-hazardous alternatives.

- Industry and agriculture must pursue clean production technologies and manufacture clean products, recognising that the only way to prevent releases of POPs into the environment is to avoid their production and use.

- As a matter of urgency, action must be taken to stop production, and eliminate all discharges, emissions and losses of those chemicals prioritised for action by UNEP.

- Presume that all chemicals are hazardous until demonstrated otherwise, i.e. until hazard identification is completed, or in those instances where hazard identification is limited by lack of information, chemicals must be assumed to present hazards of unknown proportions.

- Ultimately, measures to eliminate releases of ALL POPs and ALL OTHER HAZARDOUS SUBSTANCES to the environment will need to be taken both at a regional basis and on a global basis, because chemical contamination of the environment is a global problem and chemicals do not respect national boundaries.

1 INTRODUCTION

The building blocks of living organisms are organic compounds – that is chemical compounds that contain carbon and hydrogen (and in some cases other elements as well). These compounds are never indestructible and many break down relatively easily. On the other hand, man has learnt to manufacture organic compounds which are extremely difficult to break down. These chemicals are termed persistent organic pollutants (POPs).

A large number of hazardous chemicals have been, and continue to be, manufactured by the chemical industry both intentionally, as products, and unintentionally, as by-products and wastes. These hazardous substances include numerous POPs. Some of these POPs, notably the dioxins and furans, are also generated unintentionally as by-products of combustion processes.

The production and use of POPs, and the generation of POPs as unintentional by-products has led inevitably to the pollution of the environment with these substances. Because they are not easily degraded by natural processes, many persist in the environment for years. Therefore, even if production and releases of all POPs ceased today, they would continue to pollute the environment for many years to come. Numerous POPs have become very widespread contaminants in the environment because they can be transported for thousands of kilometres on air currents, and in rivers and oceans. As a result of this long-distance transport, some POPs even contaminate remote regions such as the deep oceans, high mountain areas and even the Arctic. Indeed, they may be considered as global pollutants.

In addition to being persistent, many POPs are, by their chemical nature, highly soluble in fats (lipophilic). Consequently they have a tendency to concentrate in the fatty body tissues of living organisms and, over time, can build up (bioaccumulate) to high levels in such tissues. In some cases the levels increase (biomagnify) as one animal consumes another in the food chain so that the highest levels are present in top predator species. Some POPs, such as organotin compounds, accumulate to particularly high levels in the liver and other tissues.

Many POPs are toxic and their long-lives in living tissues may lead to adverse effects on health. Although over time POPs may be metabolised (transformed or broken down) in the body to other compounds (metabolites), some of the metabolites produced are more toxic and persistent than the original chemical. For example, the pesticides heptachlor and chlordane are respectively broken down to heptachlor epoxide and oxychlordane which are more toxic than the original chemicals.

Man-made chemicals occur in the environment and in our bodies not as single entities but as complex mixtures. We are exposed, therefore, not to individual hazardous chemicals, but to many; not to individual POPs, but to diverse mixtures. The significance of such multiple exposure remains poorly understood. Moreover, a substantial proportion of the chemicals which occur in the environment and to which we may be exposed simply cannot be identified. This further complicates the problem.

1.1 The Chemicals of Concern

POPs may be defined in general terms as persistent organic chemicals, including synthetic substances from a range of chemical groups. A prominent and diverse group of POPs are the organohalogens, i.e. organic compounds of fluorine, chlorine, bromine and iodine. Of the halogens, chlorine has been particularly widely used by the chemical industry, in order to manufacture organochlorine chemicals for use as pesticides, industrial chemicals, solvents, cleaning agents and plastics, particularly PVC. Indeed, PVC is the largest single use of chlorine.

Indeed, all of the 12 POPs so far prioritised for action to reduce or prevent emissions under the United Nations Environment Programme (UNEP) Draft POPs Convention are organochlorine chemicals (UNEP 1995). These chemicals are described in Box 1.1.

Environmental and health problems caused by POPs included on the UNEP list have been recognised for some years and, as a consequence, the PCBs and many of the pesticides have been banned or have restricted use in most countries. However, POPs do not respect national boundaries, such that their continued production and use and generation as unintentional by-products in some countries adds to the global burden of these chemicals. In the case of dioxins, still produced unintentionally by many industrial and waste combustion processes as well as open burning, landfill fires and accidental fires in buildings, vehicles and warehouses throughout the globe. In some countries steps have been taken to reduce air emissions of dioxins from point sources, such as incinerators, but releases to air and soil from such facilities continue with little of no abatement. Moreover, few countries have established the material policies needed to address the chlorine-containing materials (e.g. PVC) that are, in effect, the dioxin sources during incineration as well as for diffuse sources, such as open burning and landfill fires.

The 12 UNEP POPs are only part of the problem we face. Many more persistent organic chemicals are still in widespread production and use, in both industrialised and less industrialised countries. A few of these are shown in box 1.2 While the chemical industry continues to manufacture such chemicals to solve day-to-day problems, they may be creating other, long-term or even irreversible problems and compromising the ability of future generations to meet their own needs. They may also be threatening the fundamental processes which support the diversity of life itself.

BOX 1.1 POPS LISTED BY UNEP

- Dioxins and furans: Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are commonly referred to dioxins and furans or collectively as “dioxins.” There are 210 individual congeners (chemicals) in the group, although some are more toxic, and some more abundant, than others. 2,3,7,8 - tetrachlorodibenzo-p-dioxin (2,3,7,8 - TCDD) is the most toxic congener, or chemical form, and is now recognised as a human carcinogen. Dioxins are produced as unintentional by-products of many manufacturing and combustion processes that use, produce or dispose of chlorine or chlorine derived chemicals. Important sources of dioxins to the environment include waste incineration, combustion of PVC in landfill fires and open burning, and many organochlorine production processes, including PVC production.

- Polychlorinated Biphenyls (PCBs): PCBs comprise of a group of 209 different congeners. Around half this number have been identified in the environment. The more highly chlorinated PCB congeners are the most persistent and account for the majority of those polluting the environment. PCBs were produced as industrial chemicals that were mainly used for insulation in electrical equipment. Production of PCBs has almost totally ceased worldwide, although there are reports of it continuing in Russia. At least one third of PCBs that have been produced are estimated to have entered the environment (Bernes 1998). The other two thirds remain in old electrical equipment and in waste dumps from where they continue to leach into the environment. Although this is the major source of PCB pollution in the environment today, some PCBs are also produced as by-products of incineration and certain chemical processes involving chlorine such as PVC production.

- Hexachlorobenzene (HCB): This chemical was previously used as a fungicide for seed grain. It is also produced unintentionally as a by-product during the manufacture of chlorinated solvents, other chlorinated compounds, such as vinyl chloride, the building block of PVC, and several pesticides. It is a by-product in waste streams of chlor-alkali plants and wood preserving plants, and in fly ash and flue gas effluents from municipal waste incineration. Its major source today remains the manufacture of pesticides (ATSDR 1997, Foster 1995).

- Organochlorine Pesticides: There are eight pesticides in this category listed by UNEP. These are aldrin, dieldrin, endrin, DDT, chlordane, mirex, toxaphene and heptachlor. The majority of these are banned or restricted in many countries, although not all. For example, DDT is still widely used in some less industrialised countries, particularly for mosquito control (e.g. Lopez-Carrillo *et al.* 1996).

Although the greatest attention to date has focused, understandably, on persistent organochlorine chemicals, the general problem of the widespread contamination of the environment with persistent chemicals extends across other chemical groups. In order to ensure protection of the environment, action must be taken to reduce and ultimately prevent emissions of all hazardous substances, particularly those which are persistent and bioaccumulative.

BOX 1.2 OTHER POPS

- Hexachlorocyclohexane isomers (HCH). γ-HCH, or lindane, is an organochlorine pesticide and a component of some shampoos for treatment of headlice. Its use as a pesticide in agriculture has declined in recent years, but it nevertheless continues to be used for this purpose in some countries of Europe (Bernes 1998), Latin America and Asia. Use of technical HCH, a mixture of HCH isomers including alpha-HCH, is yet more restricted. Nevertheless, as a result of some continued releases and its persistence in the environment, alpha-HCH remains widespread in the environment, including the Arctic.

- Brominated flame retardants. These chemicals are widely used as fire retardants in electronic equipment e.g. electronic boards in computers, radios and television sets, in plastics, textiles, building materials, carpets and in vehicles and aircraft. The production and use of some these chemicals is increasing. Brominated flame retardants include polybrominated diphenyl ethers (PBDEs), and polybrominated biphenyls (PBBs), as well as the more recently developed tetrabromobisphenol-A. It is becoming increasingly clear that PBDEs are widely distributed in the global environment and can accumulate in the tissues of humans and wildlife; similar evidence is growing for other brominated flame retardants.

- Organotin Compounds: Organotin compounds are used as active ingredients in anti-fouling agents, fungicides, insecticides and bactericides. One of the chemicals in this group, tributyltin (TBT), has been used as an anti-fouling agent in paints for boats and aquaculture nets since the 1960s, although its use is now restricted to large vessels and a global phase out for this use has been set for 2008. TBT is perhaps best know for its hormone disrupting effects in marine invertebrates, although it is also highly toxic to other organisms. It has been described as perhaps the most toxic chemical ever deliberately introduced into natural waters and has become widespread in the marine environment.

- Short Chain Chlorinated Paraffins: These chemicals have for many years been used to produce a range of products, including use as fire retardants and plasticisers in PVC, rubber and other plastics, varnishes, sealants and adhesives, leather treatment chemicals and as extreme pressure additives in lubricants and metal cutting oils (Campbell & McConnell 1980). It should be noted that it is not just the short chained chlorinated paraffins that are problematic but the whole group of chlorinated paraffins.

1.2 **Global Pollution and Transport of POPs**

Many POPs have become ubiquitous in the environment and can be detected at considerable levels even in remote regions such as the Arctic and Antarctic (e.g. Bidleman *et al.* 1993, Iwata *et al.* 1993). The contamination of remote regions occurs as a consequence of the long distance transport of POPs on air currents. Once in the atmosphere, POPs may be dispersed and transported across great distances on air currents before they are deposited on the earth's surface again. It is speculated that some POPs move through the atmosphere from warmer regions, where they are emitted, towards colder regions at higher latitudes. The hypothesis that explains how POPs move from warm regions to colder polar areas is known as global distillation or global fractionation. This is because once released to the environment, chemicals appear to become fractionated with latitude according to their volatility as they condense at different temperatures (Wania & Mackay 1993, Wania & Mackay 1996).

POPs are released into the environment, for example, from incinerator stacks to air, as industrial discharges to rivers, as pesticides sprayed onto crops and soil and as losses from a variety of consumer products. Subsequent movement of POPs between air, water, soil or vegetation depends on temperature, and on the physical and chemical properties of POPs. The global distillation hypothesis assumes that warmer temperatures favour evaporation of POPs from the Earth's surface to air, whereas cooler temperatures favour their deposition from air back onto soil, vegetation or water. The overall effect is that POPs volatilize to air in warmer climates and then condense and are deposited again on the Earth's surface in cooler climates. Researchers have suggested that POPs may migrate to the poles in a series of short hops by repeatedly undergoing the cycle of evaporation, transport and deposition (Wania & Mackay 1993). Others have suggested that the process is most likely to occur as a one-step process (Bignert *et al.* 1998). It has been noted that there are uncertainties about how the processes of exchange occur between air and soil/water/vegetation and that more research is needed (Addo *et al.* 1999).

It appears that the more volatile a chemical, the greater tendency it has to remain airborne and the faster and farther it travels on air currents towards remote polar regions. Conversely, chemicals of low volatility are unable to attain high atmospheric levels and are thus deposited close to where they are initially released. Therefore, POPs of higher volatility like α- and γ- HCH may migrate faster towards the poles than those of lower volatility like DDT which tend to remain closer to their source (Wania & Mackay 1993, Wania & Mackay 1996).

Observations suggest that certain POPs such as HCBs and HCHs, preferentially deposit in polar latitudes, while DDT and others primarily deposit at lower latitudes (Wania & Mackay 1996). For example, a worldwide study of persistent organochlorines in tree bark found that the relatively volatile compounds HCB was distributed according to latitude, demonstrating a global distillation effect. Conversely, less volatile compounds such as endosulfan were not as effectively distilled and tended to remain in the region of use (Simonich & Hites 1995).

It is thought that POPs in polar regions mainly originate from industrial and other human activities in nearby countries. For example, studies show that sources of POPs pollution in the Arctic are most likely to come from mid-latitudes of the Northern

Hemisphere such as Europe, Russia and North America (Barrie *et al.* 1989, Muir *et al.* 1997). However, the tropical countries are also responsible for spreading contamination to the polar regions, because some of these chemicals used in agriculture and public health like HCH, DDT and dieldrin are still consumed in considerable quantities in low latitude areas (Tanabe *et al.* 1991). It should be noted that most of the global inventory of POPs will be not eventually reach polar regions but will be retained and/or undergo degradation close to their source or en route to polar regions. Nevertheless, levels in polar regions can still be very high.

1.3 POPs in the Baltic

The Baltic Sea is a semi-enclosed sea into which many large rivers flow. It is connected to the North Sea via the Kattegat and inlets of the Belt Sea and Sound (Falandysz *et al.* 2000a). These areas are narrow and limit the inflow of saltwater from the North Sea. The Baltic Sea is consequently brackish, with the lowest salinity in the Bothnian Bay in the north and the highest in the Baltic proper in the south (Jansson & Dahlberg 1999). Being brackish, the water is too salty for most freshwater species and too fresh for most marine species. As a result, relatively few species inhabit the Baltic Sea in comparison to other seas (Jansson & Dahlberg 1999). Moreover, not many of the species which inhabit the Baltic Sea are regarded as typical Baltic brackish water species, most having previously migrated from nearby seas or freshwater (Falandysz *et al.* 2000a).

The limited number of species inhabiting the Baltic Sea means that, compared to other oceans, there are few species to perform all the basic functions in the ecosystem such as fixing solar energy, clearing the water, decomposing and recycling wastes. This has the effect of making the Baltic particularly sensitive to stress (Jansson & Dahlberg 1999), such as that caused by chemical contamination. Furthermore, studies have shown that some Baltic invertebrates (animals without backbones) are more sensitive to toxic pollution than the same species from more saline marine environments (Kautsky & Kautsky 2000). Given the vulnerability of the Baltic Sea to pollution from industrialised regions in surrounding countries, coupled with the semi-enclosed nature of the Sea, the sensitivity of the ecosystem is of great concern. Indeed, the Baltic Sea ecosystem may be seen to have suffered from large scale and long-term adverse impacts as a result of pollution.

1.3.1 Pollution of the Baltic Sea

The Baltic Sea links 9 nations along its coasts, constituting a total human population of about 90 million (Kautsky & Kautsky 2000). Discharges of pollution into the Baltic, and into rivers feeding the Baltic, has been in the form of both chemical pollution and high nutrient input. The high input of nutrients, such as organic matter, nitrogen and phosphorous inputs from industrial, municipal and agricultural discharges has resulted in eutrophication of the Baltic Sea (Jansson & Dahlberg 1999). The effects of eutrophication include increased biological production, decreased water transparency and reduced oxygen (Falandysz *et al.* 2000a). This has resulted in many adverse impacts on the Baltic ecosystem including reduction of species diversity and abundance in some areas (e.g. Jansson & Dahlberg 1999).

Over the past 50 years there have been substantial inputs of chemical pollutants into the Baltic Sea, from sources including industrial wastewaters, agricultural run-off, municipal sewage, atmospheric fallout, marine paints and dumping of wastes (Falandysz *et al.* 2000a). As a result, the Baltic ecosystem has become contaminated with numerous POPs, including many persistent organochlorines and heavy metals. Studies have demonstrated that levels of DDT and PCBs increased to particularly high concentrations in Baltic Sea wildlife in the 1960s and 1970s. The high levels of these and other POPs became a severe threat to Baltic wildlife, especially some birds of prey and marine mammals (Falandysz *et al.* 2000a). In some cases, drastic population crashes of several species have been linked to POPs pollution.

Since the 1970s, levels of several persistent organochlorines, notably DDT, have declined in different biological media as a result of their use being banned or restricted. Levels of PCBs have also declined, though generally at a much slower rate, probably reflecting, in part, their continued release into the environment from old equipment and waste dumps. Levels of dioxins have also decreased slowly during this period, though in recent years, levels of both dioxins and PCBs in some species appear to have not decreased. Moreover, levels of some more recently used POPs such as TBT (tributyltin, an antifouling agent) and PBDEs (polybrominated diphenyl ethers, primarily used as fire retardants) have increased in recent years. Furthermore, as in other regions, numerous POPs present in the Baltic ecosystem still remain unidentified or poorly characterised. For example, it is only recently that a number of chemicals belonging to a relatively new but poorly known group of chlorinated polycyclic aromatic hydrocarbons (Cl-PAHs) were identified in non-biological media from the Baltic Sea (see Falandysz *et al.* 2000a). Although there have undoubtedly been reductions from the high levels of POPs contamination seen in the 1960s and 70s, and partial recovery of some wildlife populations, adverse effects associated with certain POPs continue to persist today.

Due to the high level of chemical contamination of the Baltic Sea and its biota during past decades, it can be considered that the Baltic was a ‘toxic hotspot’ for POPs. Even today, the Baltic is highly contaminated with many persistent chemicals in comparison to other seas and may, therefore, still be considered as a toxic hotspot.

1.3.2 Sources of POPs contamination

Sources of POPs to the Baltic are numerous. Until recently, the pulp and paper industry had been responsible for major inputs of organochlorine compounds to the Baltic Sea, including a diversity of persistent compounds that have caused adverse impacts in fish near to pulp mill effluent discharges (e.g. Bernes 1998, Jonsson *et al.* 1996, Wulff & Rahm 1993). Moreover, research has shown that pollution from the paper mills was not only localised but has also spread to regions of the Baltic remote from the industry (Jonsson *et al.* 1996).

In the past decade or so, many pulp and paper mills in the Baltic region have substantially reduced their output of organochlorines by changing from the use of chlorine gas as a bleaching agent to the use of chlorine-dioxide. However, this bleaching technique does not completely eliminate persistent organochlorines in pulp mill effluent. Nor does it allow mills to recycle liquid wastes back through the process. To eradicate organochlorines from mill effluent necessitates a switch to Totally Chlorine Free (TCF) bleaching (Johnston *et al.* 1997), a method already adopted by some pulp and paper mills in the Baltic (Bernes 1998). Such a switch, in itself, does not overcome the problem that many natural components from the wood pulp (e.g. plant sterols) are also highly toxic to fish. Nevertheless, the move away from bleaching with chlorine compounds does allow the subsequent modification of mills to become “totally effluent free”, a move which achieves the ultimate goal of zero discharge of toxic wastes (Johnston *et al.* 1997).

Other sources of POPs to the Baltic Sea include both direct discharge from land-based sources as well as atmospheric deposition of POPs from local and more distant sources. In addition, sediments act as a sink for POPs and it has been suggested that re-suspension of sedimented organic matter may remain a substantial source of organochlorines in the Baltic Sea for a very long time (Jonsson *et al.* 1996). This may be particularly important in the Gulf of Bothnia where land uplift may cause erosion of contaminated sediments.

It is of note that the largest source of dioxins released to the atmosphere in many European and other countries has been reported to be incineration (see Allsopp *et al.* 2001). During the 1980s and mid-1990s municipal solid waste (MSW) incineration, in particular, was identified as a major source of dioxins to the atmosphere. A summary of data from 15 countries estimated that incineration accounted for about 50% of dioxin emissions to air in 1995 (Fiedler *et al.* 1999). Even recently, notwithstanding recent improvements in technology, a study identified MSW incineration in Denmark in 1998-9 as the largest single source of dioxins to the atmosphere (Hansen 2000).

1.3.3 **Political Agreements**

At the end of the 1960s, there was already great concern about the deterioration of water and wildlife of the Baltic Sea, resulting in the formation of the Convention on the Protection of the Marine Environment of the Baltic Sea Area - the Helsinki Convention in 1974 (Falandysz *et al.* 2000a). Starting in 1981, all Baltic countries assisted in establishing assessment programmes which have since been published by the Helsinki Commission (HELCOM).

Conclusions from the 4th Periodic Assessment of HELCOM have recently been released (HELCOM 2001a). In the section concerning hazardous substances, the conclusions note that while concentrations of many hazardous substances monitored over the past 30 years have decreased throughout the Baltic, many remain cause for concern. It draws particular attention to PCBs and dioxins:-

“Concentrations of dioxins and PCBs in biota have not decreased during the 1990s in the Baltic Proper, which indicate a continuous input and/or re-suspension”.

“Decreasing concentrations of a number of organo-chlorine compounds have resulted in concomitant general improvement in the health of birds of prey and mammals. However, on-going studies show that health and reproduction are impaired, possibly indicating that the present levels of organo-chlorine compounds such as PCBs and dioxins are still too high”.

“Particular consideration should be given to the impacts of organo-chlorine compounds such as dioxins in Baltic Sea fish also with regard to the food chain”.

Contamination of Baltic Sea fish, as mentioned in the last conclusion, has recently been highlighted with regard to human health by the Swedish National Food Administration (Swedish National Food Administration 1996). It recommended that girls and women of fertile age should not consume Baltic salmon, trout or herring more than once a month. These recommendations are clearly intended to reduce overall intakes of persistent contaminants from fish by women of reproductive age, such that transfer of POPs to the foetus through the placenta, and to the newborn from breast milk, may also be reduced. In addition to this recommendation for females of reproductive age, a further protective health recommendation has been given by the Swedish National Food Administration for all other members of the Swedish population. It is recommended that individuals should not eat the above listed Baltic fish more than once a week.

1.4 POPs Conventions

In 1971, the Scandinavian states initiated a process to control marine pollution through their lobbying in the run up to the United Nations (UN) Conference on the Human Environment that was held in Stockholm 5-16 June, 1972. It was at this conference that international law making first focused on toxic substances as one of the main targets to be addressed by international regulatory efforts for the control of marine pollution. For instance, Principle 7 of the Declaration from this UN conference on the Human Environment stated that:

"*States shall take all possible steps to prevent pollution of the seas by substances that are liable to create hazards to human health, to harm living resources and marine life, to damage amenities or to interfere with other legitimate uses of the sea.*"

This process and a couple of toxic waste dumping incidents initiated a series of conventions to tackle the dumping of toxic waste at sea and/or marine pollution from land based sources (Pallemaerts 1998). The Oslo (1972, dumping at sea) and Paris (1974, land based sources) Conventions (which led to the 1992 OSPAR Convention) were the first regional results of this process. The London Dumping Convention (1972, which led to the London Convention and deals with marine sources of pollution) was the first global convention to deal with toxic pollution. The Helsinki Convention was also formed in 1974 and in 1976 the Barcelona Convention (Barcon) was established.

1.4.1 The OSPAR Convention (1992) and the "One Generation Goal"

The Convention for the Protection of the Marine Environment of the North East Atlantic (the OSPAR Convention, formed from the amalgamation of the former Oslo and Paris Conventions) entered into force in March 1998 and covers the 15 States of the North East Atlantic Region and the European Union. The Convention (OSPAR 1992) requires that all Contracting Parties:-

"…take all possible steps to prevent and eliminate pollution and shall take the necessary measures to protect the maritime area against the adverse effects of human activities…"

At the OSPAR meeting held in Sintra in June 1998, Ministers from each of the Contracting States and a representative of the European Commission agreed to a common statement, the Sintra Statement (OSPAR 1998), which sets out clear commitments to address hazardous and radioactive substances, eutrophication, the offshore oil and gas industry and the overall protection of biological diversity. With respect to hazardous substances, the Ministers agreed:-

"to prevent pollution of the maritime area by continuously reducing discharges, emissions and losses of hazardous substances (that is, substances which are toxic, persistent and liable to bioaccumulate or which give rise to an equivalent level of concern), with the ultimate aim of achieving concentrations in the environment near background values for naturally occurring substances and close to zero for man-made synthetic substances".

Note that these measures are intended to apply to a much wider range of hazardous substances than those currently under consideration within the developing POPs processes outlined above. Furthermore, the Ministers agreed to make:-

"every endeavour to move towards the target of cessation of discharges, emissions and losses of hazardous substances by the year 2020".

This latter commitment, commonly referred to as the "one generation goal", is perhaps the most important element of the OSPAR approach. It represents a substantial step forward in relation to the way in which chemical substances are regulated and a fundamental departure from the more conventional (though misguided) wisdom that chemicals can be "managed" at "safe" levels in the environment. Inevitably, meeting the target of cessation of discharges, emissions and losses will necessitate the phase out of hazardous substances (or the processes which generate them) and their substitution with non-hazardous alternatives.

1.4.2 The Helsinki Convention

In 1990 the contracting parties to the Helsinki Convention decided to extend, strengthen and modernize the legal regime of the 1974 convention. A comprehensive new Convention was adopted in 1992, the Helsinki Baltic Convention (Pallemaerts 1998). This new convention entered into force on 17 January 2000. The governing body of the convention is the Helsinki Commission, also known as the HELCOM. The present contracting parties to HELCOM are Denmark, Estonia, European Community, Finland, Germany, Latvia, Poland, Russia and Sweden (HELCOM 2001b).

Throughout the 1970s and 1980s, the work on the implementation of the Helsinki Convention was carried out by experts and attracted little political attention. In 1988 the first high level ministerial meeting within the framework of the Helsinki Convention was held and this started a process to refocus HELCOM's agenda on the central issue of toxics reduction. At that time, the HELCOM set it's main task at a 50% toxics reduction by 1995 through sector wise abatement recommendations. In 1992, the 'Baltic Sea Joint Comprehensive Environmental Action Programme'(JCP) was adopted. The JCP was developed as a practical, operational action programme which identified 132 environmental "hotspots" in the Baltic catchment area (Pallemaerts 1998).

At the1996 high level-meeting in Kalmar (Sweden), the Council of the Baltic Sea States called on HELCOM to develop an action programme for the continuous reduction of inputs of hazardous substances "towards the target of their cessation within one generation (25 years)".This prompted a new round of policy-making on toxics reduction within the Helsinki Commission, in parallel with the development of the Hazardous Substances Strategy in OSPAR. This lead to the adoption at a ministerial session of the Helsinki Commission in March1998 (HELCOM recommendation 19/5 on the HELCOM objective with regard to hazardous substances, 26 March 1998 (Pallemaerts 1998). In the document attached to the recommendation a strategy to implement HELCOM with regard to hazardous substances the provisions on implementation are contained. Like OSPAR, HELCOM now formally subscribes to the “ultimate aim of concentrations in the environment near background values for naturally occurring substances.” HELCOM is also mandated to take into account "the specific conditions of the Baltic Sea" and is guided "in particular" by the precautionary principle. According to the 1992 Helsinki Convention, the precautionary principle is to be understood as the obligation to take preventive measures where there is reason to assume that substances or energy introduced, directly or indirectly, into the marine environment may create hazards to human health, harm living resources and marine ecosystems, damage amenities or interfere with other legitimate uses of the sea even when there is no conclusive evidence of a causal relationship between inputs and their alledged effects (1992 Helsinki Convention, art. 3, para 2).

1.4.3 Stockholm Convention 2001

In 2001, three decades after the process to control marine pollution was initiated, a global convention to eliminate the production and use of POPs will be adopted in Stockholm, in May. It is expected that the POPs treaty – or Stockholm Convention as it will be called - will initially ban the global production and use of 12 persistent organic substances (POPs). But it is expected that it will also be an instrument to ban other existing substances with POPs characteristics in the future and prevent new ones from entering the markets and the environment. It should not only address intentionally produced toxic chemicals, but also toxic by-products from industrial and combustion processes, such as dioxins.

The Stockholm Convention, will be the first global legal framework to ban the production and use of toxic substances. The treaty negotiations process marks the realisation that toxic pollution is not only a local issue, but indeed of global proportion whereby POPs produced in one region contaminate and injure not only the “near field” environment, but also the “far field”, e.g. high POPs concentrations in the remote Arctic environment.

The 12 POPs listed by UNEP as priority chemicals for phase out, are chlorinated substances. Some of them – dioxins, furans – are by-products of specific industrial processes, while others are intentionally produced products (PCB’s and pesticides). A global commitment to eliminate dioxins, together with specific measures towards product and processes substitutions, are key elements that make this political process specifically important for changing today’s industrial production at a global scale, into one based on clean processes and products, and not on end of pipe technologies (filters).

However, thirty years of talking have not cleaned up the Baltic. Obviously, words are not enough and immediate action is required to halt the ongoing pollution of the Baltic and other environments. These actions should include the cessation of any expansion of sources of POPs and other hazardous substances, such as incineration and the clean up of existing ones.

This report brings together existing scientific knowledge on POPs contamination in the Baltic marine environment and in humans living in Baltic nations. Specifically, levels of POPs in Baltic sediments, birds, fish, marine mammals and humans are addressed. The report goes on to discuss possible health impacts of POPs contamination on fish, birds, marine mammals and humans.

2 BALTIC MARINE SEDIMENTS

POPs enter the Baltic Sea via rivers, surface run off, atmospheric deposition, direct discharges, spills and dumping of dredging material. Because of their low water solubility, most POPs quickly become bound onto fine-grained suspended particulate matter (SPM) in the water column (Dannenberger & Lerz 1996). The suspended particles sink through the water to the sea bed where they are deposited in the sediments. In this way, sediments act as an ultimate sink for POPs entering the marine environment. Nevertheless, knowledge regarding levels of POPs in Baltic Sea sediments, and their likely effects, remains limited.

2.1 Historical Trends of PCDD/Fs and PCBs

To investigate the historical pattern of PCDD/F and PCB levels in sediments of the Baltic Sea, a sediment core was taken at an offshore site in the northwest part of the Baltic proper (Kjeller & Rappe 1995). Analysis of the core revealed that low levels of PCDD/Fs and PCBs were present in sediments during the period 1882-1962 (total PCDD/F 92-234 pg/g dry weight (dw)). The authors suggested that the combustion of natural materials such as wood, coal and peat, would have been the primary source during this period. By the period 1970-85, however, the levels of PCDD/Fs and PCBs had increased substantially (total PCDD/Fs 520-1800 pg/g dw). Investigation of the congener profiles of PCDD/Fs suggested that the main source of the increased pollution was the use of chlorinated phenols such as pentachlorophenol (PCP). In Sweden, the use of chlorinated phenols as fungicides for wood protection, for textile treatment and to control slime in pulp mills began in the 1950s, and their use was ultimately banned in 1978. Kjeller and Rappe (1995) also reported that increased levels of PCBs in the sediment core reflected their increased use after 1950.

2.2 Levels of PCDD/Fs

Several persistent organochlorine compounds and organotins have been identified in sediments from the Baltic Sea.

2.2.1 PCDD/Fs

For PCDD and PCDFs, levels in the Baltic Proper ranged from 720-750 and 730 - 1052 pg/g dry dw respectively (Kjeller & Rappe 1995). In the western Baltic, levels ranged from 12.7 to 2991 pg/g dw for PCDDs, and from 2.5 to 820 pg/g dw for PCDFs (Dannenberger *et al.* 1997). A comparison of PCDD/F concentrations in Baltic sediments with those from other regions revealed lower mean contaminant levels in the northern part of the North Sea and in the Arctic (PCDDs 25 - 337 pg/g dw and PCDFs 20 - 407 pg/g dw). Lower levels were also reported for the central North Sea, though in coastal areas, for instance the Rhine estuary, higher PCDD/F levels have been reported (up to 3000 pg/g dw).

Alkyl-PCDFs are chemicals which are similar in structure to PCDFs. These compounds have been identified in connection with chlorine bleaching of paper pulp. Kjeller and Rappe (1995), identified alkyl-PCDFs in a sediment core taken from the Baltic proper, 130 km away from the nearest pulp and paper mill. Their presence at this site provided evidence of long-distance transport of contaminated sediments from the coast to the deeper ocean. The authors estimated that it took about 30 years for the contaminated sediment to travel this distance, illustrating the fact that the impacts of chlorine bleaching can be exerted at distances and over time scales remote from the mill's operations.

2.2.2 PCBs

Concentrations of PCBs have been reported to increase along a north-south gradient. For instance, Jonsson (2000) reported that mean levels of PCBs (normalised to total organic carbon content of the sediments) were almost twice as high in the south Baltic proper (latitude 54-56 degrees, mean PCBs 382 ng/g carbon) than in the Gulf of Bothnia (latitude 60 degrees, mean PCB 213 ng/g carbon).

An investigation of PCB levels in coastal sediments in Bothnian Bay and the Bothnian Sea (Gulf of Bothnia) in 1991 reported that PCBs were the most abundant persistent organochlorines, with somewhat lower levels of DDT, HCHs, DDTs, HCBs and chlordanes (Strandberg *et al.* 2000). Similar levels of PCBs have been reported for sediments from a number of locations in the Baltic region, including 9-9.3 ng/g dw (sum of 68 congeners) for the Gulf of Bothnia (Strandberg *et al.* 2000), up to 11 ng/g dw (sum of 12 congeners) in the Baltic proper (see Dannenberger & Lerz 1996) and 8.4-11.4 ng/g dw (sum of 23 congeners) for the western Baltic (Dannenberger & Lerz 1996).

Jonsson (2000) investigated PCB concentrations from 32 sites in the offshore Baltic Sea. This gave an insight into trends of PCB levels with time in the Baltic Sea. At first sight, the results appeared to demonstrate that PCB concentrations had increased from the early 1970s onwards, in direct contrast to the declining concentrations found in marine biota during the 1970s-80s. The authors noted that sediments may be re-suspended a long time after their first deposition, and that deposition rates can significantly affect the annual burial of PCBs in sediments. Re-analysis of the data, taking deposition rates into consideration, showed that PCB deposition had increased gradually in the period between 1940-1960, reached a peak in the mid-1970s, decreased substantially during the late 1970s and 80s and increased somewhat around 1990. This trend is similar to trends of PCB concentrations reported for biota from the Baltic, although concentration reductions in sediments have been somewhat less marked for sediments.

2.2.3 Organochlorine Pesticides

Chlordanes, HCHs, PCBs, HCB, DDT and dieldrin were identified in all 7 samples of settling particulate matter analysed by Strandberg *et al.* (1998b) from remote coastal and offshore stations in Bothnian Bay and the Bothnian Sea, and from a number of coastal stations. Concentrations reported are given in Table 2-1. Comparable levels were found by Dannenberger (1996) in sediment samples taken from the western Baltic (See Table 2-1).

Table 2-1: Residues of organochlorine pesticides and PCBs in sediment from the Baltic Sea (values in ng/g or ppb dry weight)

Compound HCH	HCB	DDT	Dieldrin	Chlordanes	PCB	Reference
5-23	0.49-3.4	1.9-18	0.39-22	0.31-7.2	4.6-31 (68 congeners)	Strandberg *et al.* 1998b
β-HCH 0.5	<0.01-0.75	<0.01-9			0.12-11.3 (23 congeners)	Dannenberger 1996

A study in the western Baltic showed that the highest levels of DDT and PCBs were located in coastal rather than offshore areas (Dannenberger & Lerz 1996) and specific sources were suggested. For example, the highest levels of DDT were found in the Pomeranian Bight, probably a result of the use of DDT as an insecticide (especially in forestry) in the former German Democratic Republic until the end of 1988. DDT may also have been used for agricultural applications in Poland, although few data are available to confirm this. Somewhat elevated concentrations of DDT in the Mecklenburg Bight and Arkona Sea were thought most likely to result from river discharges as well as atmospheric deposition and transport of sediment through the water column. Among the conclusions drawn were that the main sources of organochlorine input into the estuaries and western Baltic are contamination in the Oder and Peene rivers, the shipbuilding industry (Unterwarnow), shipping operations, agriculture and urban effluents. In deeper areas of the Arkona Basin and Lubeck Bight, the source of organochlorines bound to sediments is most likely long distance sediment transport.

A comparison of concentrations for some of the above mentioned organochlorines revealed that, for most compounds, levels in Baltic sediments were about 5-30 times higher than in sediments from, for example, the Gulf of Alaska and the Mediterranean (Strandberg *et al.* 1998b). There were, however, some similarities between levels of PCBs and DDTs in Baltic sediments and those from locations in southern California and, for PCBs at least, from the North Sea. For some areas of the western Baltic Sea, however, Dannenberger and Lerz (Dannenberger & Lerz 1996) recorded PCB levels around 10 times higher than mean levels in the North Sea.

2.2.4 PCNs

Polychorinated napthalenes (PCNs) are ubiquitous environmental contaminants although reports on their levels in the environment are relatively scarce (Ishaq *et al.* 2000). In the Baltic, PCNs have been detected in white-tailed sea eagles, harbour porpoise and black cormorants (see Falandysz *et al.* 1996), as well as in sediments (Falandysz *et al.* 1996, Ishaq *et al.* 2000). Falandysz *et al.* (1996) reported a concentration of 6.7 ng/g dw in a sediment sample from the nearshore sedimentation area of the Wisla River in the Gulf of Gdansk, in which up to 44 of a possible 48 congeners (tetra- to hepta-CN) were identified. Of these, tetra- and penta-congeners were more abundant than hexa- and hepta-CNs, supporting the hypothesis that atmospheric transport was the main source of PCNs in the Gulf of Gdansk Basin. This is because differences in volatility of different PCN congener groups can favour the higher environmental mobility of lower chlorinated PCN congeners. Ishaq *et al.* (2000) also reported PCNs in two samples of sediment from a coastal station in the Baltic Proper, again with tetra- and penta-CN congeners predominating.

2.2.5 Organotins

Organotins have been widely used as anti-fouling paints on ships on a global scale. A study on sediments taken from German marinas in the Baltic Sea in 1997 identified both tributyltin (TBT) and triphenyltin (TPT) in all samples (Biselli *et al.* 2000). In addition, the degradation products of TBT and TPT, mono- and di-butyltin (MBT, DBT) and mono- and di-phenyltin (MPT, DPT) respectively, were present in the samples. Levels of TBT (up to 17000 ng/g dw), TPT (up to 3800 ng/g dw) and DBT (up to 14,000 ng/g dw) were substantial at some of the sites investigated. The study noted that the levels were surprisingly high given that samples were taken eight years after a partial ban on the use of organotin antifouling formulations (for vessels under 25 m in length) in Germany.

3 POPS IN BALTIC MARINE FISH

3.1 Introduction

During the twentieth century the Baltic changed from a clear water, low nutrient sea to one of the most chemically polluted and high nutrient (eutrophic) marine environments in the world. The change in nutrient status of the Baltic has been reflected in changes exhibited by the biota that live in and around it, and it was the massive decline in the populations of some top predators that finally caused measures to be put in place to limit the amount of pollution entering the sea. As a result decreasing concentrations of environmental pollutants have been observed for biota in these waters after a peak in the late 1960s – early 1970s (Larsson *et al.* 2000).

Cod, herring and sprat are the most economically important fish species in the Baltic Sea (Jansson & Dahlberg 1999). Stocks of these species declined up to 1990, except for an increase in cod stocks between the late 1970s and mid-1980s. Since 1990, the spawning stock of sprat has increased significantly. However, the cod stock is presently still below biologically safe limits (Jansson & Dahlberg 1999) .

Cod (*Gadus morhua*) is a predatory, migratory fish of the continental shelf that lives in waters of the northern hemisphere in depths up to 600 m and generally near the sea bed. Cod accumulates lipophilic organochlorines in its liver and, because it moves around considerably, it accumulates pollution from throughout the area in which it lives. As a result it is a good indicator of pollution near the sea bed over a wide area (Falandysz *et al.* 1994b). The Baltic Sea is generally much shallower than 600 m – very little of it is deeper than 200 m and in theory, where salinity and oxygen status allow, most of the Baltic is a suitable feeding ground for this species (Jansson & Dahlberg 1999).

Atlantic salmon (*Salmo salar*) is found throughout Europe from the Arctic circle in the north to Portugal in the south. Stocks of salmon in the Baltic Sea have been in steady decline since the 1900s due to exploitation of rivers for hydroelectric power and an intensification in salmon fisheries. The introduction of aquaculture has seen a change in composition of the salmon stock. Originally some 50 Baltic Sea rivers accommodated spawning salmon populations. Today very few still harbour reproducing stocks. Whereas in the 1940s all salmon in the Baltic were wild, today 85-90% of salmon are from artificial hatcheries (Bernes 1998, Jansson & Dahlberg 1999). This has an enormous effect on the genetic make-up of the salmon stocks, reducing the gene pool and making the fish more susceptible to disease than previously (Bernes 1998, Jansson & Dahlberg 1999).

Herring, (*Clupea harengus*) is also a commercially important Baltic species that has undergone a significant population expansion over recent decades primarily as a result of the fall in predatory cod numbers (Bernes 1998).

Recent evidence suggests that the deep-sea is the final global sink of semivolatile pesticides. A survey comparing the burden of persistent organic pollutants in different species of fish, graded by the depth at which they live, found significantly higher burdens of these contaminants in bottom-dwelling species (Looser *et al.* 2000).

As detritus from the surface, be it whole organisms, or their waste, sinks through the water it is inevitably eaten by something else. This process happens a number of times before material reaches the sea bed. Alternatively, the pollutants may become adsorbed to falling dead matter. Once at the sea bed, or in the deeper parts of the ocean basin, they are again able to biomagnify in the food web that exists there (Looser *et al.* 2000, Skei *et al.* 2000). Fish from the sea bed were found to have up to 600 times the concentration of DDT when compared with surface living species and although this data was not collected in the Baltic Sea it may also be applicable there (Looser *et al.* 2000).

3.2 **Levels in fish and plankton**

3.2.1 **Dioxins and PCBs**

3.2.1.1 ***Dioxins (PCDDs)and Furans(PCDFs)***

The Baltic sea has been the subject of considerable academic research into dioxins and furans, but this has tended to concentrate on the top predators such as birds of prey and marine mammals. There are few data on concentrations of dioxins and furans in marine fish and molluscs.

Considerable variation was found in the concentrations of PCDD/Fs in Baltic herring from Finnish coastal areas. Much higher concentrations were found in fish caught in polluted estuaries and coastal areas (those near heavy industry) when compared with fish caught at less impacted sites. PCDD/F concentrations ranged from 1-3.6 pg/g (ppt) iTEQ fresh weight (fwt) with lower concentrations (around 1 ppt iTEQ fwt) in unpolluted areas in the Gulf of Bothnia. Highest concentrations (around 3.6 ppt iTEQ fwt) were found in fish from the Gulf of Finland where there is considerably more industrial activity. Bioaccumulation factors were highest in pike, approximately 10 times that in herring. Herring had lower concentrations of PCDD/Fs than did other fish species sampled, probably because of the dilution effect caused by their greater fat deposits (Korhonen & Vartiainen 1997).

Higher concentrations of PCDD/Fs were found in herring from the southern Baltic where values of 81 ppt iTEQ fat were reported. Similar values were found in salmon (up to 84.9 ppt iTEQ fat) (See table 3-1) (Becher *et al.* 1998, Falandysz *et al.* 1997d). These values appear to be associated with high levels of industrial pollution although they are not directly comparable with the fresh weight concentrations reported in the previous paragraph. It should, however, be noted that dioxin concentrations in herring vary considerably from one individual to another, even within a very small area, so that comparisons can only be made with confidence when very large data sets are used (Bernes 1998).

Table 3-1: PCDD/Fs in Baltic Marine Fish

Species	Compound PCDD/Fs	References
flounder (*Platychthis flesus*)	8.4 ppt iTEQ fat	Falandysz *et al.* 1997d
pike (*Esox lucius*) (N. Baltic)	0.5 ppt iTEQ fwt	Korhonen & Vartiainen 1997
pike (*Esox lucius*) (Gulf of Finland)	1 ppt iTEQ fwt	Korhonen & Vartiainen 1997
herring (*Clupea harengus*)	81 ppt iTEQ fat	Falandysz *et al.* 1997d
herring (*Clupea harengus*) oil	250 ppb fat	Öberg *et al.* 1999
herring (*Clupea harengus*) (N. Baltic)	<1 ppt iTEQ fwt	Korhonen & Vartiainen 1997
herring (*Clupea harengus*) (Gulf of Finland)	3.6 ppt iTEQ fwt	Korhonen & Vartiainen 1997
cod (*Gadus morhua*)	7.8 ppt iTEQ fat	Falandysz *et al.* 1997d
salmon (*Salmo salar*)	16.7-84.9 ppt iTEQ fat	Becher *et al.* 1998

The amount of dioxin biomagnification in the Baltic Sea food chain is very dependent on the level of chlorination of the dioxin congener concerned. In this context, the most heavily chlorinated (those with eight chlorine atoms) are not necessarily the most prone to biomagnification. The most toxic dioxins are those which contain four or five chlorine atoms, and these are the ones that are also found in increasing concentrations in predator species such as herring and cod (Bernes 1998).

3.2.1.2 ***Polychlorinated biphenyls (PCBs)***

PCBs are found throughout the Baltic and in most fish and mollusc species. PCB concentrations in fish may vary considerably depending on the age or size of the fish and on other factors such as migration and length of exposure (Chiu *et al.* 2000). Nevertheless, highest concentrations appear in areas with the heaviest industrial pollution, around the coast of Poland, and in the north eastern Baltic in the areas impacted by states of the former USSR.

A 1994 study found that cod liver oil from the Baltic was up to five times more contaminated with PCBs (up to 10 ppm fat) than comparable samples from the Icelandic Shelf (Falandysz *et al.* 1994b).

Seasonal PCB concentrations varied with fat content in mussels from less polluted areas of the southern Baltic, increasing during the autumn and decreasing during early and late summer. Reduction in residues during the middle of the summer appeared to be related to spawning. This relationship was much less pronounced in mussels taken from a polluted harbour area. Stress caused by organochlorine pollution is known to affect filtration rate in mussels, and thus their ability to feed (Lee *et al.* 1996b). PCBs concentrations were strongly elevated during the winter, much more than could be accounted for by changes in lipid content alone, in mussels from the most polluted site (Kiel Innenförde, 487.1 ppb dry weight (dwt), winter compared with 200.4 and 297.8 ppb dwt during the previous and subsequent summers at the same site). Only slightly elevated levels were observed in mussels taken from less polluted harbours (e.g. Travemünde 266 ppb dwt, winter, compared with 237.1 and 130.5 ppb dwt during the previous and subsequent summers at the same site) (Lee *et al.* 1996b). Levels of PCBs were also strongly elevated in mussels from the harbours (131.4-487.1 ppb dwt) when compared with samples taken from the beach (23.6-87.2 ppb dwt) or out to sea (36.9-66.7 ppb dwt) (See table 3-2) (Lee *et al.* 1996b). An unusual fluctuation in the concentrations of congener #101 in mussels collected from a site out to sea (Kiel Leuchturm) was attributed to a recent local input of PCB to that area during the sampling period (Lee *et al.* 1996b).

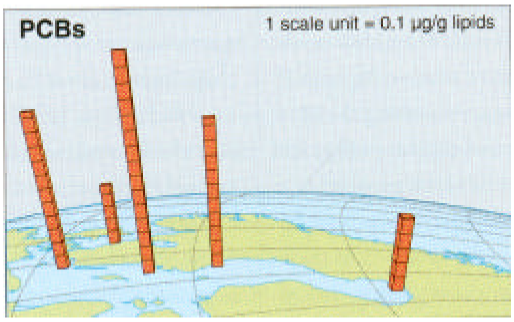


Figure 3.1: PCB levels in herring from five stations in the Baltic in the mid 1990s (from Bernes, 1998)

Research in some areas of the Baltic has suggested local input of PCBs, even in recent years. High concentrations of lower-chlorinated PCBs in perch (*Perca fluviatilis*) sampled at Daugavgriva on the Estonian coast appeared to correspond to the congener profile of a Russian-made PCB formulation, Solvol. There was concern that the high water exchange at Daugavgriva could lead to further pollution by PCBs both of the Gulf of Riga and of the Baltic proper (Olsson *et al.* 1999). A similar situation pertains in Poland, where the PCB congener pattern exhibited by flounder collected close to the estuary of the Vistula River was characteristic of freshly discharged PCBs, unlike the congener patterns shown in other fish samples taken from Gdynia and Mikoszewo which were characteristic of environmentally 'aged' PCBs. Concentrations of PCBs in fish were also very high in this southern part of the Baltic, up to 9400 ppb fat in flounder, and 11000 ppb fat in eelpout (See table 3-2) (Falandysz *et al.* 1997c, Falandysz *et al.* 1998a). Fig 3.1 gives the data from the Swedish Contaminant Monitoring programme which provides further evidence of high levels of PCBs in fish from the southern Baltic. The highest concentrations shown, however, appear to exist close to the Stockholm archipelago (Bernes 1998).

Table 3-2: PCBs in Baltic Marine Fish

Species	Compound PCBs	References
mussels (*Mytilus edulis*) – beach	23.6-87.2 ppb dwt	Lee *et al.* 1996b
mussels (*Mytilus edulis*) – harbour	131.4-487.1 ppb dwt	Lee *et al.* 1996b
mussels (*Mytilus edulis*) – open sea	36.9-66.7 ppb dwt	Lee *et al.* 1996b
stickleback (*Gasterosteus aculeatus*)	4.8-8.2 ppb fat	Falandysz *et al.* 1997a
three-spined Stickleback (*Gasterosteus aculeatus*)	2700-4200 ppb fat	Falandysz *et al.* 1998b
eelpout (*Zoarces viviparus*)	11000 ppb fat	Falandysz *et al.* 1997c
perch (*Perca fluviatilis*)	3000-6400 ppt fat	Falandysz *et al.* 1998a
perch (*Perca fluviatilis*)	570-1900 ppb fat	Olsson *et al.* 1999
pikeperch (*Stizostedion lucipercа*)(Lake Ladoga), Russia	1.04 ppm fat	Kostamo *et al.* 2000
pikeperch (*Stizostedion luciperca*)	11000 ppb fat	Falandysz *et al.* 1998a
smelt (Lake Ladoga), Russia	0.68 ppm fat	Kostamo *et al.* 2000
vendace (*Coregonus albula*) (Lake Ladoga), Russia	0.69 ppm fat	Kostamo *et al.* 2000
lamprey (*Lampetra fluviatilis*)	100-1700 ppb fat	Falandysz *et al.* 1998a
lamprey (*Lampetra fluviatilis*)	1700 ppb fat	Falandysz *et al.* 1997c
flounder (*Platychthis flesus*)	4800-9400 ppt fat	Falandysz *et al.* 1998a
flounder (*Platychthis flesus*)	910-9400 ppb fat	Falandysz *et al.* 1997c
herring (Clupea harengus)	1 ppm fat (1980s-1990s)	Korhonen *et al.* 1995
herring (Clupea harengus)	<1-3 ppt iTEQ fwt	Korhonen & Vartiainen 1997
cod (*Gadus morhua*) liver (tinned)	ΣPCBs 1200-2600 ppm f.wt 290-530 ppt iTEQ	Falandysz *et al.* 1992
cod (*Gadus morhua*) liver oil	ΣPCBs 6,600-17,000 ppb fat	Falandysz 1994
cod (*Gadus morhua*) liver oil	7.5-10 ppm fat	Falandysz 1994
cod (*Gadus morhua*)	1400 ppb fat	Falandysz *et al.* 1997c
cod (*Gadus morhua*) liver	PCB-153 109-254 ppb wwt	Fromberg *et al.* 2000
salmon (*Salmo salar*)	ΣPCBs 1462-3090 ppb fat	Atuma *et al.* 1998a
salmon (*Salmo salar*)	PCB-153 8-20 ppb wwt	Fromberg *et al.* 2000
salmon (*Salmo salar*)	3.5-5 ppm fat	Korhonen *et al.* 1995

3.2.2 Organochlorine pesticides

3.2.2.1 DDTs

Concentrations of DDTs have fallen considerably in plankton collected from the Baltic Sea when compared with levels in the early 1980s. Residues measured in 1992 were five to eight times lower than in comparable samples analysed in 1983 and were reported as being in the region of 300 ppb fat in recent studies (Falandysz *et al.* 1997e, Falandysz *et al.* 1998c). As the base of the Baltic sea food chain, the presence of DDT compounds in plankton will result in the transfer and accumulation of these residues to each successive predator. The data presented in table 3-3 give a good example of this process of biomagnification. Reported concentrations of DDT are larger at each successive level in the food chain rising from around 300 ppb fat in plankton to 1000-2000 ppb fat in predator species such as herring, flounder and cod.

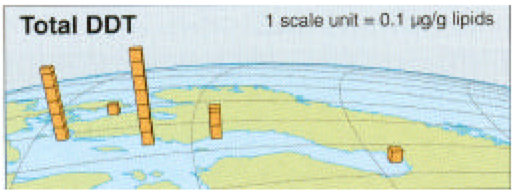


Figure 3.2: Average levels of DDT in herring from five stations in the Baltic for 1994-6 (from Bernes 1998)

Mussels from the southern part of the Baltic exhibited different concentrations of DDTs depending, as expected, on the nature of the environment from which they had been collected. Animals from harbours had higher concentrations (up to 88.3 ppb dwt) than those taken from less polluted areas such as an island out to sea (26.4 ppb dwt) or the beach (6.6-16 ppb dwt) (see table 3-3) (Lee *et al.* 1996a).

Three-spined stickleback are one of the smaller fish to inhabit coastal areas of the Bay of Gda sk on the Polish coast. Fish collected from four sites on a beach contained DDT levels ranging from 1300-2600 ppb fat (see table 3-3). The authors reported that these concentrations were elevated when compared with concentrations in other fish species from the northern part of the Baltic Sea although these comparative data were not given (Falandysz *et al.* 1997a). They are, however, similar to those reported for DDT concentrations in perch (*Perca fluviatilis*) (up to 1100 ppb fat) from the Gulf of Riga in the north eastern Baltic (Olsson *et al.* 1999). The authors suggested that illegal use or improper storage of this compound continued among former USSR countries, allowing local input of DDT into the Gulf of Riga. High levels (1400 ppb fat) (see table 3-3) too were found in this species collected on the Baltic south coast (Falandysz *et al.* 1999b). In this case the authors attributed the high concentrations to past usage of DDTs in Poland and their continued presence at elevated concentrations in Polish soils. In addition they suggested that use of DDT continued in neighbouring countries up until the early 1980s and that this might have resulted in aerial transport and deposition onto Polish soil (Falandysz *et al.* 1999b).

Fig 3.2 shows the situation that pertained in the mid-1990s for DDT residues in herring. As with so many other compounds, the highest values around the Swedish coast were found in the Stockholm archipelago (700 ppb fat), with lowest levels in Bothnian Bay (100 ppb fat) and the straits joining the Baltic with the North Sea (<100 ppb fat) (Bernes 1998).

Samples of cod liver oil (see table 3-3) taken from the southern Baltic sea had maximum DDT concentrations of 6300 ppb. This was twice as high as in samples from the western Baltic (3400 ppb fat) and more than three times the level found in samples from the North Sea (1700 ppb fat) (Falandysz *et al.* 1994b).

Table 3-3: DDTs in Baltic Marine Fish

Species	Compound DDTs	References
plankton	300 ppb fat	Falandysz *et al.* 2000a
plankton	270-340 ppb fat	Falandysz *et al.* 1997e
mussels (*Mytilus edulis*)	5.1-59.1 ppb dwt	Lee *et al.* 1996b
mussels (*Mytilus edulis*)-harbour	26.9-88.3 ppb dwt	Lee *et al.* 1996a
mussels (*Mytilus edulis*)-beach	6.6-16 ppb dwt	Lee *et al.* 1996a
mussels (*Mytilus edulis*)-sea	26.4 ppb dwt	Lee *et al.* 1996a
blue mussel (*Mytilus edulis*)	910 ppb fat	Falandysz *et al.* 1999b
three-spined Stickleback (*Gasterosteus aculeatus*)	1800 ppb fat	Falandysz *et al.* 1999b
three-spined Stickleback (*Gasterosteus aculeatus*)	1300-2600 ppb fat	Falandysz *et al.* 1997a
perch (*Perca fluviatilis*) (Gulf of Riga)	180-1100 ppb fat	Olsson *et al.* 1999
perch (*Perca fluviatilis*) (S. Baltic)	1400 ppb fat	Falandysz *et al.* 1997a
perch (*Perca fluviatilis*)	1300-1500 ppb fat	Falandysz *et al.* 1998d
perch (*Perca fluviatilis*)	1400 ppb fat	Falandysz *et al.* 1999b
pikeperch (*Stizostedion luciperca*)	1900 ppb fat	Falandysz *et al.* 1999b
pikeperch (*Stizostedion luciperca*) muscle (Lake Ladoga)	350 ppb fat	Kostamo *et al.* 2000
smelt muscle (Lake Ladoga)	480 ppb fat	Kostamo *et al.* 2000
vendace (*Coregonus albula*) muscle (Lake Ladoga)	380 ppb fat	Kostamo *et al.* 2000
lamprey (*Lampetra fluviatilis*)	980-1400 ppb fat	Falandysz *et al.* 1998d
flounder (*Platychthis flesus*)	1500-1700 ppb fat	Falandysz *et al.* 1998d
flounder (*Platychthis flesus*)	1600 ppb fat	Falandysz *et al.* 1999b
herring (Clupea harengus)	p,p'DDE 3-30 ppb wwt	Fromberg *et al.* 2000
herring (Clupea harengus) oil	1700 ppb fat	Öberg *et al.* 1999
herring (Clupea harengus)	280-700 ppb fat	Roots 1995
herring (Clupea harengus)	1300 ppb fat	Falandysz *et al.* 1997b
herring (Clupea harengus)	1300 ppb fat	Falandysz *et al.* 1999b
cod (*Gadus morhua*) liver oil (W.Baltic)	3400 ppb fat	Falandysz *et al.* 1994b
cod (*Gadus morhua*) liver oil (S.Baltic)	6300 ppb fat	Falandysz *et al.* 1994b
cod (*Gadus morhua*) liver oil (N. Atlantic)	1700 ppb fat	Falandysz *et al.* 1994b
cod (*Gadus morhua*) liver	p,p'DDE 103-547 ppb wwt	Fromberg *et al.* 2000
cod (*Gadus morhua*)	1200 ppb fat	Falandysz *et al.* 1999b
salmon (*Salmo salar*)	p,p'DDE 25-56 ppbkg wwt	Fromberg *et al.* 2000

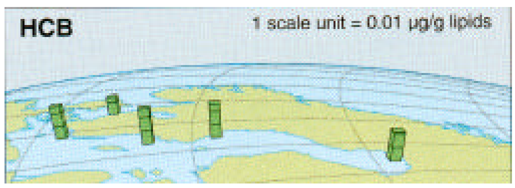
3.2.2.2 ***Hexachlorobenzene (HCB)***

Levels of HCB ranged from 10-16 ppb fat in a study that examined plankton from throughout the Baltic. HCB concentrations in plankton showed little regional variability. The authors considered that such consistent values indicated inputs were largely through long range aerial deposition rather than from a local source (Falandysz *et al.* 1997e).

Unlike in plankton, levels of HCB in fish varied in different regions of the Baltic Sea. Cod liver oil from the southern part of the Baltic Sea had the highest concentrations of HCBs (280 ppb fat) with fish from this part of the Baltic containing almost double the concentration found in fish from the west Baltic (160 ppb fat). This compared with cod from the north Atlantic, for example, where concentrations were about half those in the Baltic (110 ppb fat), and were similar to those from the North Sea (130 ppb fat) (see table 3-4) (Falandysz 1994). Even within a much smaller area, the Gulf of Gda sk, there was variability in HCB concentrations in fish. Stickleback sampled from a site impacted by industry in the Gulf of Gda sk had elevated concentrations of HCB (40 ppb fat) compared with 3 other sites in the Gulf where concentrations ranged from 18-28 ppb fat (see table 3-4) (Falandysz *et al.* 1997a). If levels of HCB were indeed consistent throughout the Baltic Sea as a result of aerial deposition, it would seem unlikely that more polluted sites on the south coast should give rise to elevated concentrations of this compound in fish. Clearly there are other routes by which HCB continues to enter the Baltic Sea.

There may be some disagreement about the route by which HCBs are entering the Baltic Sea but there is little doubt that it continues to do so. Although there had been a slow decline during the 1970s and 1980s in HCB contamination of Baltic fish, the declines are no longer consistent and some areas have reached a steady state of HCB contamination, indicating a continuous input of this compound (Falandysz 2000, Falandysz *et al.* 1994b, Falandysz *et al.* 2000b).

Figure 3.3: HCB concentrations in herring from five sites in the Baltic in the years 1994-1996 (from Bernes 1998)



Levels of HCBs in herring (See Fig 3.3) around the Swedish coast follow a similar pattern to a number of other POPs. Highest values (30 ppb fat) are found around the Stockholm archipelago, lower levels in herring from Bothnian Bay (20 ppb fat) and lowest levels of all (about 12 ppb fat) on the Swedish west coat, outside the Baltic (Bernes 1998).

Table 3-4: Concentrations of HCH, Chlordanes, Heptachlor epoxide, dieldrin and HCBs in Baltic Marine Fish

Species	Compound HCH	Chlordanes	Heptachlor Epoxide	Dieldrin	HCB	References
plankton		8.0 ppb fat	3.3 ppb fat	31 ppb fat		Falandysz *et al.* 1998c
plankton					10-16 ppb fat	Falandysz *et al.* 1997e
mussels (*Mytilus edulis*) - harbour	4.9-22.3 ppb dwt				0.8-2.4 ppb dwt	Lee *et al.* 1996b
mussels (*Mytilus edulis*) - beach	1.5-8.7 ppb dwt				<LoD-6.5 ppb dwt	Lee *et al.* 1996b
mussels (*Mytilus edulis*) – open sea	4.7-6.4 ppb dwt				1.0 ppb dwt	Lee *et al.* 1996b
blue mussel (*Mytilus trossulus*)		12 ppb fat		19 ppb fat		Falandysz *et al.* 1999a
three-spined Stickleback (*Gasterosteus aculeatus*) (unpolluted site) (S.Baltic)					18-28 ppb fat	Falandysz *et al.* 1997a
three-spined Stickleback (*Gasterosteus aculeatus*) (polluted site) (S. Baltic)					40 ppb fat	Falandysz *et al.* 1997a
perch (*Perca fluviatilis*)	αHCH 4.7-126 ppb fat γHCH 5.4-63 ppb fat			4.3-17 ppb fat	3.8-15 ppb fat	Olsson *et al.* 1999
perch (*Perca fluviatilis*)					0.85 ppb wwt	Falandysz 2000
herring (*Clupea harengus*) oil	61 ppb fat				41 ppb fat	Öberg *et al.* 1999
herring (*Clupea harengus*)					41 ppb fat (PCBz 8.4 ppb fat)	Falandysz *et al.* 1997b
herring (*Clupea harengus*)		*trans*-chlordane 2.3 ppb fat *cis*-chlordane 14 ppb fat Oxychlordane 6.0 ppb fat *trans*-nonachlor 12 ppb fat *cis*-nonachlor 3.6 ppb fat	3.9 ppb fat			Strandberg *et al.* 1998a
herring (*Clupea harengus*)					3.7 ppb wwt	Falandysz 2000
herring (*Clupea harengus*)					3.7 ppb wwt	Falandysz *et al.* 2000b
cod (*Gadus morhua*) liver					60 ppb wwt	Falandysz 2000
cod (*Gadus morhua*) liver oil					160 ppb fat	Falandysz 2000
cod (*Gadus morhua*)					0.46 ppb wwt	Falandysz *et al.* 2000b
cod (*Gadus morhua*) liver oil	450-580 ppb fat	240-340 ppb fat	12-19 ppb fat	240-270 ppb fat	160-280 ppb fat	Falandysz 1994
cod (*Gadus morhua*)					0.46 ppb wwt	Falandysz 2000
salmon (*Salmo salar*)					1.4 ppb wwt	Falandysz 2000

3.2.2.3 *Hexachlorocyclohexanes (HCHs)*

Total HCHs reported for mussels from the south coast of the Baltic Sea ranged from 1.5-22.3 μg/kg (ppm) dry weight (dwt) (Lee *et al.* 1996b), with the highest levels in those areas expected to be more contaminated. In the Gulf of Gda sk, for example, the least contaminated were those collected from beaches and the most heavily contaminated those collected from harbours (Lee *et al.* 1996b). The data were reported as dry weight concentrations so they are not directly comparable with other studies reviewed.

HCHs in perch (*Perca fluviatilis*) from the Gulf of Riga contained both α- and γ-HCHs at similar levels to those in other areas of the Baltic. The isomers were found at a 1:1 ratio, and this was similar to ratios found in other studies on Baltic fish. Concentrations averaged around 10 ppb (ppb) fat, and the authors considered that current input was predominantly through long-range atmospheric transport and aerial deposition rather than contamination by a local source (Olsson *et al.* 1999). Fish collected from one site, however, appeared to have a much lower α-HCH:γ-HCH ratio, which suggested a recent input (of lindane, or technical HCH) near the Gulf of Riga (Olsson *et al.* 1999). The highest values reported for HCHs in fish were for cod liver oil from the western and southern parts of the Baltic Sea (450-580 ppb fat respectively) – an order of magnitude greater than the concentrations found in perch (Falandysz 1994). The αHCH/γHCH ratios in cod liver oil were approximately 2.0 and 2.2 for western and southern Baltic samples respectively, indicating a far greater proportion of γHCH (and therefore a more recent input of lindane) than was found in North Sea (ratio = 12) and Icelandic Shelf (ratio = 9.3) samples (Falandysz *et al.* 1994b).

3.2.2.4 ***Dieldrin***

For dieldrin available data indicate that aerial deposition from long range transport is again the most probable current route of pollution by this pesticide, as concentrations throughout the Baltic were very uniform in the plankton sampled (mean 31 ppb fat) (see table 3-4) (Falandysz *et al.* 1998c). Dieldrin concentrations do, however, appear to be quite stable in the Baltic environment (Falandysz *et al.* 1999a).

In a Polish study of cod liver oil from the Baltic, North Sea and Atlantic, dieldrin was detected in all the samples analysed at concentrations ranging from 90 to 330 ppb fat and means within the Baltic of 240 – 270 ppb fat (see table 3-4) (Falandysz *et al.* 1994b) were recorded. These values were significantly higher than those found in samples of North Atlantic origin (130-190 ppb fat) (Falandysz *et al.* 1994b).

A Latvian study examined levels of dieldrin in perch (*Perca fluviatilis*). The study was unable to compare levels within the Baltic due to lack of data. However, it was found that Baltic perch contained significantly lower concentrations of dieldrin (about 10 ppb fat – range 4.3-17 ppb fat) (see table 3-4) than were found in a similar Dutch study, where concentrations were up to six times higher. It should be noted, however, that the samples from the Dutch study were taken from waters connected to the River Rhine, which is one of the most polluted waterways in Europe (Olsson *et al.* 1999).

3.2.2.5 ***Chlordanes***

There is a relative paucity of data on chlordane concentrations in Baltic biota when compared with other persistent organohalogen compounds. Nor is it easy to compare values between the limited sources as they are frequently reported for different chlordane congeners or combinations of congeners. Very high residues (240-340 ppb fat) were, however, reported in cod liver oil in the early 1990s (Falandysz 1994) and high levels of *cis*-chlordane continued to be present in marine animals later in that decade (14 ppb fat) (Strandberg *et al.* 1998a). The data for chlordane presented in table 3-4 are illustrative of the way in which each successive step up the food chain exhibits increasing concentrations of this group of compounds. At the bottom of the food chain plankton contains chlordane concentrations of 8.0 ppb fat. Mussels, also near the bottom of the food chain, are reported as having a similar chlordane loading (12 ppb fat). Predatory fish such as cod are, however, reported as having tissue concentrations an order of magnitude greater than these (240-340 ppb fat).

3.2.2.6 ***Toxaphene***

Germany banned the used of toxaphene in 1980 and introduced residue limits in 1997 for fish and fish products, thus acknowledging the environmental damage caused by these compounds and their effect on human health (Witte *et al.* 2000). The polychlorinated bornanes (which form part of the very complex mixture of toxaphene congeners) in particular are major contaminants in fish (Witte *et al.* 2000).

A study of toxaphene content in a number of fish species found that, although levels were comparable between the more polluted western Baltic Sea and areas such as the Kattegat, and the North Sea, congener #26 was more prevalent in western Baltic samples (table 3-5). Concentrations of this congener in fish from the western Baltic up to 21.4 μg/kg (ppm) fat were found in cod liver oil; 23.1 ppm fat in salmon and 13.5-19.5 ppm fat in herring (Fromberg *et al.* 2000).

Table 3-5: Toxaphene in Baltic Marine Fish

Species	Compound Toxaphene	References
herring (*Clupea harengus*)	3 congeners (#26,#50,#62) 8-88 ppb fat	McHugh *et al.* 2000
herring (*Clupea harengus*)	#26 13.5-19.5 ppb fat	Fromberg *et al.* 2000
cod (*Gadus morhua*) liver oil	#26 15.2-21.4 ppb fat	Fromberg *et al.* 2000
salmon (*Salmo salar*)	1.59-8.70 ppb wwt.	Atuma S.S. *et al.* 1998a
salmon (*Salmo salar*)	#26 13.4-23.1 ppb fat	Fromberg *et al.* 2000

3.2.2.7 ***Heptachlor and heptachlor epoxide***

High concentrations of heptachlor and its metabolite heptachlor epoxide were found in populations of plankton (3.3 ppb fat) throughout the Baltic Sea and appeared to be related to pollution from the surrounding land masses rather than from aerial transport and deposition (table 3-4) (Falandysz *et al.* 2000b). Slightly higher values (3.9 ppb fat) were detected in herring from the southern Baltic (Strandberg *et al.* 1998a). The highest concentrations of these compounds in Baltic fish were found in cod liver oil (12-19 ppb fat) (Falandysz 1994). As for chlordane, the limited data for heptachlor epoxide indicate its capacity to biomagnify (see table 3-4). As would be expected, lowest residues were found in plankton (3.3 ppb fat) and the highest (12-19 ppb fat) in cod. The figures cited here do not suggest that biomagnification in these compounds is particularly high, but it should be borne in mind that they can be used only as a guideline as data from different studies are rarely directly comparable.

3.2.3 **Other compounds**

3.2.3.1 **Tris*(4-chlorophenyl)methane and –methanol (TCPM-H & TCPM-OH)***

Recently the compounds *Tris*(2-chlorophenyl)methanol (TCPM-OH) and *tris*(4-chlorophenyl)methane (TCPM-H) have been linked with DDT pollution, after it was discovered that they had occurred as contaminants in older DDT formulations. The compounds biomagnify through the food chain, with the highest concentrations appearing in top predators. Much lower concentrations, sometimes below limits of detection, are found in species lower down the food chain such as plankton and mussels (Falandysz *et al.* 1998d). In a study that examined concentrations of TCPM-H/OH throughout the Baltic food chain, no quantifiable residues were found in blue mussels, but they occurred, in conjunction with DDT (and its metabolites), in all samples of fish analysed (see table 3-6) (Falandysz *et al.* 1998d). Perch (25 ppb fat) and pikeperch (27 ppb fat) contained the highest reported concentrations (Falandysz *et al.* 1998d, Falandysz *et al.* 1999b).

It is of great concern to note that TCPM-H and TCPM-OH appear to be still more persistent in the environment than DDT and its metabolites (Falandysz *et al.* 1998d). The very high biomagnification factors of these compounds (about 70 for TCPM-H and about 20 for TCPM-OH), (Falandysz *et al.* 1998d) and the consequent possible impacts they may make on organisms at the top of the Baltic food web, makes their further study and understanding a priority.

Table 3-6: TCPM-H/-OH in Baltic Marine Fish

Species	Compound TCPM-OH / TCPM-H	References
plankton	TCPM-H/OH 19 ppb fat	Falandysz *et al.* 2000a
three-spined Stickleback	TCPM-H/OH 17 ppb fat	Falandysz *et al.* 1999b
three-spined Stickleback (*Gasterosteus aculeatus*)	TCPM-OH 2.4-11 ppb fat TCPM-H 6.4-18 ppb fat	Falandysz *et al.* 1998d
perch (*Perca fluviatilis*)	TCPM-OH 2.7-7.1 ppb fat	Falandysz *et al.* 1998d
perch (*Perca fluviatilis*)	TCPM-H/OH 25 ppb fat	Falandysz *et al.* 1999b
pikeperch (*Stizestedion luciperca*)	TCPM-H/OH 27 ppb fat	Falandysz *et al.* 1999b
lamprey (*Lampetra fluviatilis*)	TCPM-H 4.0-6.8 ppb fat TCPM-OH 1.2-1.7 ppb fat	Falandysz *et al.* 1998d
flounder (*Platychthis flesus*)	TCPM-H 11-24 ppb fat TCPM-OH 2.8-7.3 ppb fat	Falandysz *et al.* 1998d
flounder	TCPM-H/OH 22 ppb fat	Falandysz *et al.* 1999b
herring (Clupea harengus)	TCPM-H/OH 3.7 ppb fat	Falandysz *et al.* 1999b
cod (*Gadus morhua*)	TCPM-H/OH 16 ppb fat	Falandysz *et al.* 1999b

3.2.3.2 Nitro-musks (musk xylene, musk ketone, musk ambrette, musk moskene & musk tibetene)

These chemicals have been widely used as perfume agents in cosmetics and detergents, but their propensity towards accumulation in biological tissues has only recently been recognised. The nitro-musks are highly lipophilic and persistent in the environment. Their presence in human adipose tissues and in human milk have led researchers to look for these compounds in other organisms, including those in the human food chain such as fish (Rimkus & Wolf 1995).

In samples of fish and crustaceans taken from various freshwater and marine waters throughout Europe, musk xylene and musk ketone were detectable in nearly all the samples analysed. Highest reported concentrations are for fish from the river Stör in Schleswig Holstein, Germany (mean 90, range 20-350 ppb fat musk xylene; mean 120; range 10-380 ppb fat musk ketone in freshwater fish). Musk xylene concentrations were consistently higher than musk ketone concentrations, except in some river fish. Mussels and shrimp from the North Sea and the Baltic Sea contained low concentrations of musk xylene and musk ketone (10-50 ppb fat) (Rimkus & Wolf 1995). The results suggested that the primary sources for these compounds were communal wastewater treatment plants rather than industrial sources (Rimkus & Wolf 1995).

3.2.3.3 Polychlorinated diphenyl ethers (PCDEs) and Polybrominated diphenyl ethers (PBDEs)

PCDEs are a group of compounds, some congeners of which exhibit toxic properties similar to those found in co-planar PCBs. As with PCBs they have spread throughout the environment (Koistinen *et al.* 1995a). Very little data exists on their concentrations in living organisms, although a Finnish study examined salmon from various locations and found that concentrations of congener #47 were 20 times higher in fish taken from the Simojoki river (0.64-2.4 ppb fwt) than at other sites studied in Finland (Koistinen *et al.* 1993). The estimated iTEQ (sum of congeners #77, #105, #114 and #167) were below 0.2 ppt fresh weight (fwt) from all three samples sites, although the authors noted a considerably higher PCDE load in Baltic samples compared with those from the Arctic (Koistinen *et al.* 1993).

Dietary uptake efficiency of PBDEs increases with degree of bromination up to six bromine atoms, such that hexa-brominated congeners are more persistent in the organism than those with fewer bromine atoms. With more highly brominated congeners (e.g. octa- and deca-bromodiphenyl ethers), however, uptake declines with increasing bromination, possibly because the size of the molecules slows transport across membranes. In general, as for the persistent organochlorines, the highest concentrations of these compounds are found in animals at the top of food chains (Burreau *et al.* 2000).

Table 3-7: PCDEs and PBDEs in Baltic Marine Fish

Species	Compound PCDEs	PBDEs	References
perch (*Perca fluviatilis*)		2,2'4,4'-tetrabromodiphenylether 6.4-10 ppb fat	Olsson *et al.* 1999
herring (*Clupea harengus*) - Baltic		0.0-19.1 ppb fat	Burreau *et al.* 2000
herring (*Clupea harengus*) - Atlantic		0.0-4.1 ppb fat	Burreau *et al.* 2000
salmon (*Salmo salar*)	<0.2-250 ppb fat		Koistinen *et al.* 1995a
salmon (*Salmo salar*) – Baltic		0.4-45.8 ppb fat	Burreau *et al.* 2000
salmon (*Salmo salar*) - Atlantic		0.4-7.6 ppb fat	Burreau *et al.* 2000

In a laboratory research experiment conducted on blue mussels (*Mytilus edulis*), bioaccumulation factors were calculated for PBDEs, and congeners #47 and #99 were found to be particularly bioaccumulative. The bioaccumulation factors calculated for these congeners were up to eight times higher than for a number of PCBs also tested, including PCB 153, indicating that these substances could be more difficult to metabolise or excrete even than some of the more bioaccumulative PCBs (Gustafsson *et al.* 1999).

Research has shown that some PBDE congeners are biomagnified in Baltic Sea food chains. In a recent study concentrations of specific PBDE congeners were found to be significantly higher in salmon from the Baltic Sea (eg #51 + #49, 11.7 ppb fat) than in comparable fish from the Atlantic (#51 + #49, 0.9 ppb fat). This tendency was also evident in herring, although to a lesser extent (#51 + #49, 4.3 ppb fat, Baltic; #51 = #49, 1.6 ppb fat, Atlantic) (Burreau *et al.* 2000). The ranges for PBDEs were also significantly greater in Baltic salmon (0.4-45.8 ppb fat) than in Atlantic caught fish (0.4-7.6 ppb fat), and this was true too for herring (0.0-19.1 ppb fat, Baltic; 0.0-4.1 ppb fat, Atlantic) (see table 3-7) although to a lesser extent (Burreau *et al.* 2000).

In a study undertaken on the eastern coast of the Baltic, in the Gulf of Riga, all the samples of perch analysed were contaminated with PBDEs (6.4-10 ppb fat) (see tab;e 3-7). The authors reported that these concentrations were similar in magnitude to those found in herring from the Swedish east coast, although no comparative data were given. The authors considered that the major input of PBDEs to Latvian waters was long-range transport and air deposition (Olsson *et al.* 1999).

3.2.3.4 *Polychlorinated naphthalenes (PCNs)*

Little research has been undertaken to investigate the environmental levels of PCNs in the Baltic Sea. In one study, PCNs were analysed in mussels (*Mytilus trossulus*) from two sites in the Gulf of Gda sk and were found to be at relatively high concentrations when compared with fish higher in the food chain collected at the same time (see table 3-8) (Falandysz *et al.* 1997g). Although food can be the main route of exposure to PCNs for fish, it was considered that for filter feeding mussels, particularly in the case of the lower-chlorinated PCNs, the greatest exposure would be directly from the water rather than through their food (Falandysz *et al.* 1997g).

The levels of PCNs in fish shown in table 3-8 are very similar to those for mussels. Perch and flounder from the southern Baltic sea had concentrations of 19-69 ppb fat and 36-83 ppb fat respectively (Falandysz *et al.* 1997g). Lowest concentrations were found in Lamprey (*Lampetra fluviatilis*) (6.3-8.9 ppb fat) also from the southern

Baltic (Falandysz *et al.* 1997g). No data were found for PCN levels in other areas of the Baltic.

Table 3-8: PCNs in Baltic Marine Fish

Species	Compound PCNs	References
mussel (*Mytilus trossulus*)	80-110 ppb fat	Falandysz *et al.* 1997g
perch (*Perca fluviatilis*)	19-69 ppb fat	Falandysz *et al.* 1997g
lamprey (*Lampetra fluviatilis*)	6.3-8.9 ppb fat	Falandysz *et al.* 1997g
flounder (*Platychthis flesus*)	36-83 ppb fat	Falandysz *et al.* 1997g

3.2.3.5 ***Butyltins***

Tributyltin (TBT) is known to cause increased occurrence of imposex (a condition where females begin to grow penises and eventually become incapable of reproduction) in some species of marine shellfish. In addition TBT is toxic to fish, particularly during early life stages. Recent studies of TBT in fish and other aquatic organisms suggest that it is still present in considerable concentrations in the aquatic environment of the southern Baltic Sea despite restrictions on its use (Senthilkumar *et al.* 1999).

Butyltin compounds were found in herring from the southern Baltic. Total butyltin concentrations up to 4800 ppb wwt in herring liver, and up to 370 ppb wwt in herring eggs, were detected (see table 3-9) (Senthilkumar *et al.* 1999). According to the authors, people eating significant quantities of herring (ie more than 250 g per day) would exceed the Polish Tolerable Daily Intake (TDI) for butyltins (15 μg) (based on a 60 kg person) (Senthilkumar *et al.* 1999). This value, which amounts to 0.25 μg/kg/day is one derived by Penninks (1993) that is generally accepted and referred to in the literature (Belfroid *et al.* 2000).

Levels of butyltins reported by Senthilkumar *et al.* (1999) were very much higher than those found in another study of a number of fish species from the southern Baltic, although the data are not directly comparable as they refer to herring liver and eggs as opposed to whole fish. In the study by Kannan & Falandysz (1997) considerable range existed in the concentrations of total butyltins determined in fish, from 19-455 ppb wwt. Particularly high concentrations were found in pikeperch (455 ppb wwt), flounder (316 ppb wwt) and eel (188 ppb wwt) (see table 3-9). The authors reported that these levels were higher than those found in fish from Asian countries and similar to those in the muscle of tuna from Italian coastal waters (Kannan & Falandysz 1997).

The fish species analysed by Kannan & Falandysz (1997) (table 3-9) were largely those consumed by humans. Herring, cod and flounder constitute 90% of the Baltic fisheries product eaten by Poles for example. The calculated daily intake of these species gave a range of 770-22800 ng per person of butyltins, and the authors commented that the upper level would exceed the Tolerable Daily Intake and consumption advisories for butyltin in fish in Poland (Kannan & Falandysz 1997).

Blue mussels from the Swedish coast of the Baltic have been reported as containing 150-3000 ppb dwt butyltins, though it should be noted that these values are not directly comparable as they are reported as dry, rather than wet, weight (Kannan & Falandysz 1997).

Table 3-9: Butyltins in marine fish

Species	Compound BTs	References
eel (*Anguilla anguilla*)	188 ppb wwt	Kannan & Falandysz 1997
eelpout (*Zoarces viviparus*)	130 ppb wwt	Kannan & Falandysz 1997
pikeperch (*Stizostedion luciperca*)	455 ppb wwt	Kannan & Falandysz 1997
flounder (*Platychthis flesus*)	316 ppb wwt	Kannan & Falandysz 1997
Turbot (*Psetta maxima*)	39 ppb wwt	Kannan & Falandysz 1997
mackerel (*Scomber scombrus*)	27 ppb wwt	Kannan & Falandysz 1997
herring (*Clupea harengus*) eggs	370 ppb wwt	Senthilkumar *et al.* 1999
herring (*Clupea harengus*) liver	4800 ppb wwt	Senthilkumar *et al.* 1999
herring (*Clupea harengus*)	40 ppb wwt	Kannan & Falandysz 1997
cod (*Gadus morhua*)	19 ppb wwt	Kannan & Falandysz 1997
Sea trout (*Salmo trutta*)	51 ppb wwt	Kannan & Falandysz 1997

3.3 **Trends**

3.3.1 **Time trends in fish and marine molluscs**

Temporal (time) trends of organochlorine and organohalogen compounds in the Baltic Sea are clearly illustrated by concentrations analysed in different individual species. In this section various studies on the most commercially important fish species are reviewed.

3.3.1.1 ***Cod and cod liver***

Of the many species of fish caught for human consumption and oil extraction, cod liver, and the oil extracted from it, is probably most highly affected by accumulation of organohalogen compounds. Concentrations of certain compounds in cod-liver oil have in the recent past occasionally exceeded the tolerance limits set by most European countries leading to bans on sale being temporarily imposed in some places (eg Sweden and Poland). High concentrations of POPs have been present in cod livers from the Baltic Sea since the late 1960s, but some have fallen since the imposition of environmental controls (Falandysz *et al.* 1994b, Falandysz *et al.* 1992).

No significant temporal trend has been found in the concentrations of dioxins in cod livers. A slight increased in five congeners that occurred from 1987-1994 has been attributed to long range atmospheric deposition rather than local input (Sinkkonen & Paasivirta 2000).

Researchers have produced conflicting results regarding trends in the levels of PCBs in Baltic cod. It is not clear whether they have declined as suggested by one author, or whether they have been stable since 1971 after an initial decline (Falandysz 1994, Falandysz *et al.* 1994b, Falandysz *et al.* 1992, Sinkkonen & Paasivirta 2000). Researchers seem to agree, however, that rates of decline have slowed considerably. Whether rates of decline have levelled off completely in cod may become clearer as more data is gathered. If PCB levels in cod are no longer declining this suggests that there may be continuing input of these compounds from surrounding countries, possibly from Russia and countries formerly part of the USSR where environmental controls are weaker (Falandysz *et al.* 1994b). A study undertaken in the early 1990s indicated that average levels of PCBs in canned cod livers from Poland exceeded the 2.0 ppm regulatory limits specified for total PCBs in foodstuffs set by a number of authorities including the US Food & Drug Administration, and that there continued to be a human health hazard associated with eating them (Falandysz *et al.* 1992, Jacobs *et al.* 1997).

Most authors agree that levels of DDT and its metabolites have generally been falling in Baltic cod. DDD fell significantly (from 50-20 ppb fat) during the period 1987-1998. DDE, however, does not appear to have exhibited any significant downward trend. For DDTs, however, there has been an average annual decrease of 9-13% (Falandysz *et al.* 1994b, Roos *et al.* 1998, Sinkkonen & Paasivirta 2000). As with PCBs the dramatic decreases in concentrations of DDTs that occurred in species such as herring have now largely levelled off.

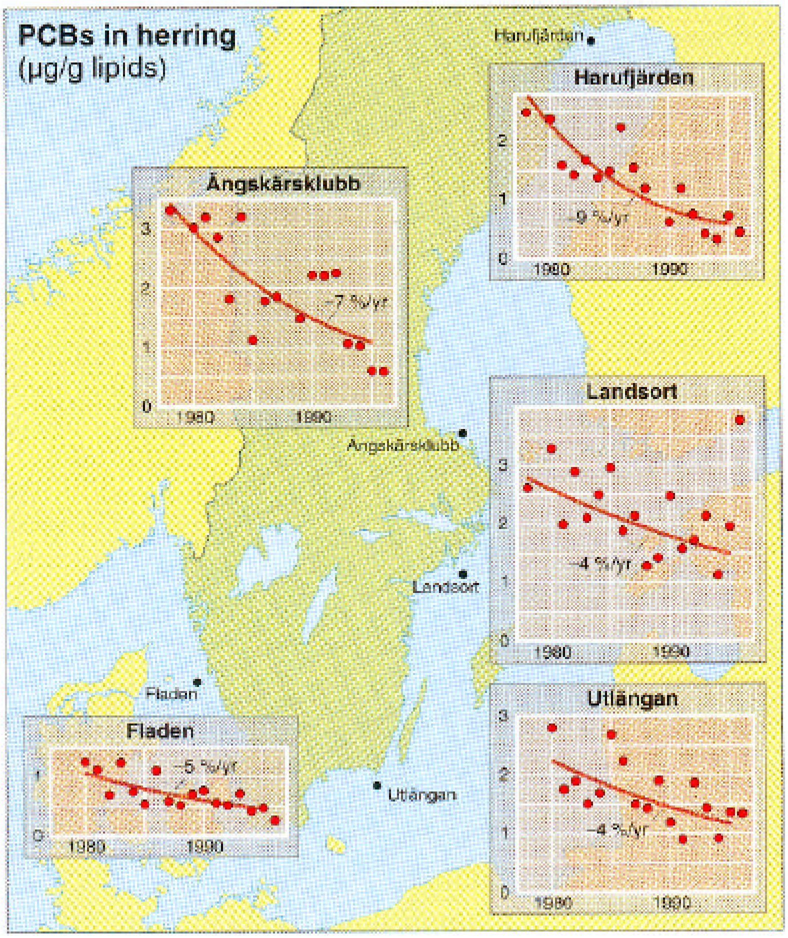


Figure 3.4: Levels of PCBs in herring from the Baltic Sea over the past 20 years (from Bernes 1998)

Trends in HCB concentrations in cod are not clear. Researchers are suggesting that, at best, concentrations are declining very slowly, and that, at worst, a steady state concentration has been reached (Falandysz *et al.* 1994b, Sinkkonen & Paasivirta 2000).

Although HCH concentrations began to fall in the 1960s there is some dispute about concentration trends in these substances thereafter. γ-HCH appears to have reached a steady state probably as far back as 1971, and only α-HCH continued to decline very slowly after that. γ-HCH (lindane) was widely used in Poland until the late 1980s and this could go some way to explaining its continued presence in cod livers (Falandysz *et al.* 1994b, Sinkkonen & Paasivirta 2000).

Dieldrin concentrations in cod appear to be declining very slowly (Falandysz *et al.* 1994b).

The literature does not confirm any clear trend in levels of chlordane in cod. At best concentrations are declining very slowly, but it is possible that they also have reached a steady state (Falandysz *et al.* 1994b, Sinkkonen & Paasivirta 2000).

Chloronaphthlalenes and PCDEs exhibited no significant trend, although the sum of 45 PCDE congeners reached a peak in 1994 of 20.96 ppb fat decreasing again quickly by 1998 (Sinkkonen & Paasivirta 2000).

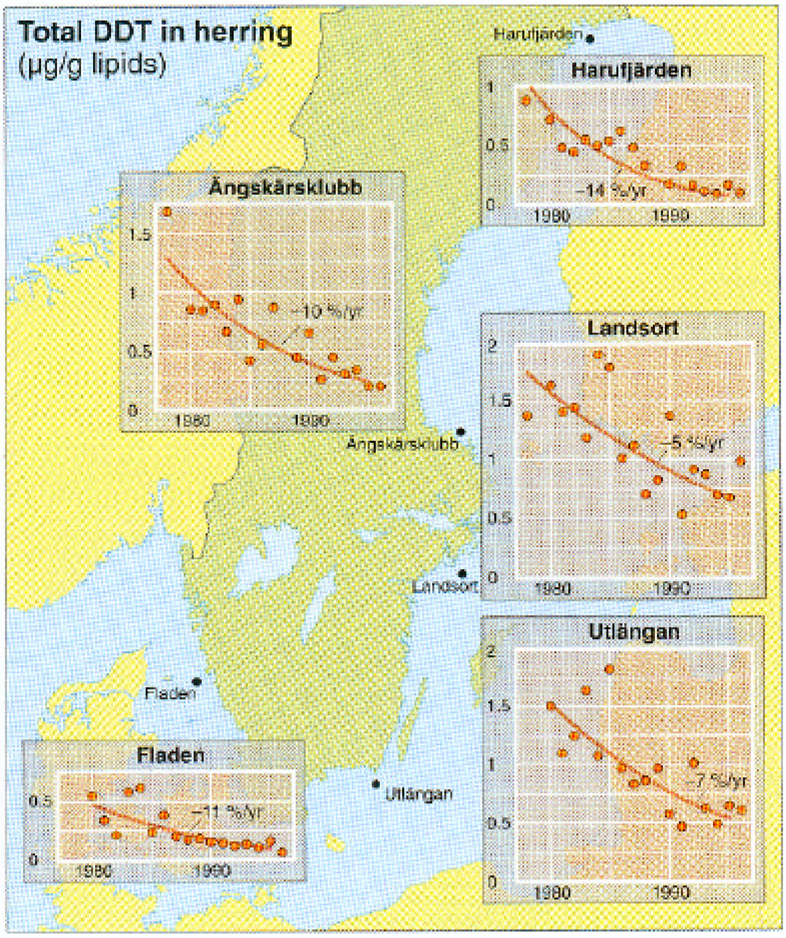


Figure 3.5: Time trends in DDT concentrations in herring over the last 20 years (from Bernes 1998)

3.3.1.2 *Herring*

Concentrations of PCBs and DDT (and its degradation products DDD and DDE) have been measured in herring (*Clupea harengus*) since the early 1970s, and have shown a relatively steady decline since that time, to approximately 15% and 5% respectively of the levels recorded in the late 1960s and early 1970s. Concentrations of PCDD/Fs have shown a similar decline (Bernes 1998, Jonsson *et al.* 1996, Korhonen & Vartiainen 1997, Olsson *et al.* 1999).

Time trend studies of PCBs and DDTs in herring (See Figs 3.4 & 3.5) indicated that there was an average annual decrease in PCBs of 9-10% between 1965 and 1995, and of 9-13% for DDTs (Roos *et al.* 1998). In 1986 DDT concentrations in herring muscle in fish from the Gulf of Finland ranged from 0.3 ppm fat in Bothnian Bay, to 0.7 ppm fat in the Gulf of Finland (Roots 1995). Time trend studies in herring also indicated a general decline in the concentrations of PCDD/Fs over the last 30 years in fish taken from the northern Baltic (Korhonen & Vartiainen 1997).

3.3.1.3 *Other fish species*

PCB concentrations in Finnish pike muscle have decreased considerably both in fish from coastal and inland areas between 1971 and 1994. In 1971 pike from coastal waters had average concentrations around 10 µg/g (ppm) fat and in 1994 this had decreased to 2.1 ppm fat. Pike from inland waters in 1971 exhibited average PCB concentrations >7 ppm fat and this had fallen to 1.7 ppm fat by 1994 (Korhonen *et al.* 1995). PCDD/F concentrations in pike from the northern Baltic have shown a general decline over the past 30 years (Korhonen & Vartiainen 1997).

3.4 Effects of POPs on marine fish and molluscs

3.4.1 Effects on reproduction

A number of fish species in the Baltic Sea have shown signs of reproductive disorders in recent decades, in particular Atlantic salmon (*Salmo salar*), sea trout (*Salmo trutta*), cod (*Gadus morhua*) and burbot (*Lota lota*). A recent study indicated that reproductive success in cod early life stages was severely affected in animals taken from the Baltic when compared with those from an unpolluted area. The unaffected cod larvae showed between 75% and 85% viability, compared with only 3% - 30% in the larvae from the Baltic Sea. A number of different deformations were found, including embryos with cord curvatures, or with disrupted yolk sacs. Abnormalities could occur in up to 80% of embryos. The researchers concluded that environmental pollution was the most probable underlying cause for the increase in incidence of deformity and decrease in cod reproductive success in the Baltic Sea. It was also suggested that over-fishing and a change in spawning time towards the summer when oxygen levels in water are lower could also be factors contributing to low recruitment (Åkerman *et al.* 1996, Vallin *et al.* 1999).

The pulp and paper industry have been responsible for releasing large amounts of chlorinated organic compounds into the environment. Bleached kraft pulp mill effluents (BKME) are produced by the use of chlorine bleaching technology. The effluents, which contain a vast number of chlorinated organic chemicals, are known to be particularly toxic with respect to reproduction in fish. In the mid-1980s physiological disturbances were found in fish caught near Swedish pulp mills. These disturbances included reduced plasma sex hormone levels, reduced gonad growth and delayed sexual maturity. Chlorine bleaching is now much less common, and fish reproductive success has increased, but there remain concerns over pulp mill effluents as they continue to exert a weakly estrogenic effect in male fish. Pulp and paper mill effluents contain oestrogen- and dioxin-like compounds including plant sterols and PCDFs (Vos *et al.* 2000). Females exposed to these compounds become highly masculinised leading to impaired reproductive function, or even chemical neutering. Unfortunately the complex mix of chemicals in BKME makes it extremely difficult to identify the causative agent(s) (Vos *et al.* 2000). The concentrations of chlorophenolics found in e.g. bile from perch (*Perca fluviatilis*) tends to decrease with increasing distance from the effluent source (Söderström *et al.* 1994).

Exposure to other POPs, like the chemicals found in such polluted habitats as industrialised estuaries and coastal waters, can have a deleterious effect on reproductive success in fish. It was reported that elevated tissue concentrations of PCBs appeared to be connected to reduced egg weight and number. Other chlorinated organics (HCB, DDT and PCBs) were implicated in precocious (or early) maturation of fish (Vos *et al.* 2000). Several fish species have exhibited symptoms such as larval deformities and the increased mortality of pelagic or demersal eggs that are thought to be caused by exposure to chlorinated hydrocarbons and other highly persistent bio-accumulative compounds. There have also been reports of embryonic deformities (Vos *et al.* 2000).

Populations of Atlantic salmon from the Baltic Sea have experienced continuous reproductive failure in hatcheries. Hatcheries use fish ascending rivers to spawn as breeding stock. Although the authors were, of course, unable to demonstrate a causal link, the presence of a large number of chlorinated and mixed brominated and chlorinated phenol-type compounds in salmon plasma is clearly cause for concern (Asplund *et al.* 1999).

3.4.1.1 ***M74 syndrome***

Over the last ten years there has been a significant increase in mortality of farmed salmon fry, characterised by oedema, haemorrhage and ataxia. Together these symptoms are known as M74 syndrome (Larsson *et al.* 1996). A possible contributing factor for this syndrome is the eutrophication of the Baltic Sea that has occurred since the 1970s. This has encouraged a proportional growth in herring populations compared with other species. Herring, a favoured prey species of salmon, are quite fatty in comparison with other prey and accumulate significant amounts of lipophilic pollutants. The pollutant content of prey is one of the factors governing uptake in predators and, by eating more herring, the salmon may have been exposed to much higher pollution loads. This in turn is suspected of leading to a decrease in reproductive capability (Larsson *et al.* 1996).

The incidence of M74 syndrome has been variable through the years but has been steadily increasing since the late 1980s. It has not yet been possible to determine conclusive relationships between M74 and known pollutants or environmental factors (Bengtsson *et al.* 1999, Jonsson *et al.* 1996). However, the occurrence of M74 syndrome has been found in connection with elevated levels of DDTs, PCDD/Fs and PCBs in the environment. This syndrome is now also affecting the few remaining wild populations of Atlantic salmon in the Baltic, and may yet cause them to become extinct in the region (Karlström 1999, Vos *et al.* 2000).

3.4.1.2 ***Non-reproductive effects***

Immunosuppression in fish has also been related to such contaminants as PCBs, PCDDs and TBT, resulting in increased disease susceptibility in wild fish stocks. Epizootic skin and liver diseases, including cancer, have been linked to polluted habitats in Europe (Vos *et al.* 2000).

In a laboratory experiment, PCNs fed to juvenile Baltic salmon (*Salmo salar*) caused adverse effects on liver function and changes in liver enzymes in the fish when compared with a control sample (table 3-10). Gonadal morphology was also altered with exposed fish showing delayed development of the gonads (Åkerblom *et al.* 2000).

Experiments indicate that BKME (bleached kraft pulp mill effluents) cause changes in immunological and physiological function in exposed fish. The effluents lower the ability of fish to fight cancers and cause changes in white blood cell counts (Luebke *et al.* 1997). Malformations of the spine have also been associated with dioxins and other chlorinated compounds (table 3-10).

Table 3-10: Environmental Impacts on fish and possible causative agents (adapted from Swedish EPA, 1996)

Observation/Impact	Sensitive Species	Substance	Association/Causation
Reproduction (M74)	salmon	chlorinated substances	2
Induction of metabolising enzymes	perch	chlorinated/unchlorinated organic mixture/PCDD/F	3-4
Spine malformations	four-horned sculpin	Chlorinated/unchlorinated organic mixture/ PCDD/F	3-4
Larval damage	sea mussel	chlorinated/unchlorinated organic mixture	3

Association/causation is assessed on the scale: 1 = no observed association, 2 = suspected association, 3 = weak association, 4 = clear association, 5 = significant association

4 POPS IN BALTIC MARINE BIRDS

4.1 Introduction

There are a host of different water birds and birds of prey that also consume fish as the main constituent of their diets. As a consequence many of these species become exposed to contaminants accumulated in their prey. Removal of these pollutants from their tissues relies on metabolic degradation of compounds and excretion through urine and faeces. Birds may also sequester some contaminants such as butyltins in their feathers (Falandysz *et al.* 2000a, Kautsky & Kautsky 2000, Senthilkumar *et al.* 1999). Removal of persistent organic pollutants from avian tissues can be very slow, often slower than the rate of uptake, leading to a gradual increase in tissue residues of toxic compounds through life. As a consequence older animals can have very high concentrations of these substances in their organs and tissues (Falandysz *et al.* 2000a, Kautsky & Kautsky 2000). One of the difficulties associated with reporting on levels of POPs in bird species, particularly the raptors, is that data cannot be obtained systematically. The species most threatened are, of course, protected so samples can only be obtained when dead, or sometimes dying, birds are found. With sea birds it is possible, in certain circumstances, to collect a very few eggs for analysis without affecting the population as a whole.

Birds that inhabit the Baltic marine area can be divided largely into two categories; seabirds, such as gulls, terns, guillemots and cormorants, that live in large colonies, and raptor species, such as fish eagles, that live singly or, at most, as breeding pairs. Only a few bird species reside in the Baltic all year round, although there are a number of important Baltic wintering sites on the German-Polish coast (Vorpommern and Szczecin lagoons), Pomeranian Bay between Denmark, Germany and Poland, and further north in the Gulf of Riga (Falandysz *et al.* 2000a).

This review focuses on three fish eating bird species that are high in the Baltic food chain, the black cormorant (*Phalacrocorax carbo*), the black guillemot (*Uria aalge*) and the white-tailed sea eagle (*Haliaeetus albicilla*). Predatory birds such as these three species provide a useful monitor of environmental contamination by organochlorine compounds. Populations of all these species are resident in the Baltic, although they may migrate from north to south during the winter and researchers cannot always state categorically (other than for ringed birds) that every bird they examine is a Baltic resident. In addition to local birds, white-tailed sea eagles from NE Russia and Swedish and Finnish Lapland often spend their winters within the Baltic area (Koistinen *et al.* 1995a).

These predatory bird species, in particular the white-tailed sea eagle, have been under severe threat due to such pollutants as DDT and concentrations of a number of pollutants remain at very high levels in their tissues. The white-tailed sea eagle is a generalist feeder predominantly eating fish (of a variety of species) but also taking other birds, small mammals and carrion (Koistinen *et al.* 1995a).

The black guillemot is a strict specialist and feeds almost entirely on eelpout (*Zoarces viviparus*), sprat (*Sprattus sprattus*) and herring (*Clupea harengus*) (Falandysz *et al.* 1997c, Koistinen *et al.* 1995a). The cormorant (or black cormorant), *Phalacrocorax carbo*, inhabits coastal waters, particularly rocky shorelines, but will also fly to inland water bodies. It eats only fish, but of many different species depending on availability, and so is less specialised than the guillemot.

4.2 Levels

4.2.1 Dioxins and PCBs

4.2.1.1 Dioxins (PCDDs) and Furans (PCDFs)

Predatory birds may biomagnify dioxins to levels ten times as high as in their food (Bernes 1998). A study of guillemots (eggs) and white-tailed sea eagles (breast muscle) found that the concentrations of the most toxic 2,3,7,8-substituted PCDDs and PCDFs ranged from <0.04-1.3 ppb fat in guillemot eggs and from <0.03-80 ppb fat in eagles. 2,3,7,8-TeCDD, the most toxic congener, was detected at concentrations between 0.4 and 13 ppb fat in eagles and exhibited an average concentration of 0.56 ppb fat in guillemots eggs (Koistinen *et al.* 1995a). In both guillemot eggs and white-tailed sea eagles, however, the contribution towards toxicity made by PCDD/Fs was very much lower than that made by the dioxin-like PCBs (See Section 4.2.1.2). The TEQ of PCDD/Fs was, on average 1.6 ppb fat in guillemot eggs and varied from 0.8-66 ppb fat in eagles (Koistinen *et al.* 1995a). Although the figures for eggs and breast muscle are not directly comparable, guillemot eggs had consistently lower levels of dioxins than did the breast muscle of eagles.

Both PCDDs and PCDFs were found in the tissues of white-tailed sea eagles from coastal areas (230 ppb fat and 240 ppb fat respectively) whereas residues in birds from inland areas were below detection limits (Falandysz *et al.* 2000a). This pattern is similar to that found in other compounds such as DDTs for which eagles from coastal areas contained consistently higher tissue residues than those from inland areas (See Section 4.2.2.1).

Table 4-1: PCDD/Fs in marine birds

Species	Compound PCDD/Fs	References
black cormorant (*Phalacrocorax carbo*) liver	420 ppt fat	Falandysz *et al.* 1997d
black cormorant (*Phalacrocorax carbo*)	PCDD 50 ppb fat iTEQ PCDD/Fs 58 ppb fat iTEQ	Falandysz *et al.* 2000a
black cormorant (*Phalacrocorax carbo*) breast muscle	110 ppt fat	Falandysz *et al.* 1997d
white-tailed sea eagle (*Haliaeetus albicilla)* breast muscle adult	470 ppt iTEQ fat	Falandysz *et al.* 1997d
white-tailed sea eagle (*Haliaeetus albicilla)* liver adult	870 ppt iTEQ fat	Falandysz *et al.* 1997d
white-tailed sea eagle (*Haliaeetus albicilla*) egg	50-57 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*) muscle	28-240 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*)	PCDD 230 ppb fat iTEQ (coastal) PCDFs 240 ppb fat iTEQ (coastal)	Falandysz *et al.* 2000a

4.2.1.2 Polychlorinated biphenyls (PCBs)

High levels of PCBs continue to be found in such sensitive species as the white-tailed sea eagle. In a study that summed the effects of dioxins and dioxin-like PCBs found in the eggs of white-tailed sea eagles, dioxin-like PCBs contributed much more to the total toxicity (approximately 75%) than did dioxins themselves. The dioxin-like PCB concentrations were at levels 50 times those expected to cause no adverse effect to the population (Koistinen *et al.* 1997a). The authors pointed out that even if concentrations of PCBs were reduced 50 times, adverse effects would not necessarily be prevented because other similar compounds, including dioxins, add to the total toxicity. Populations of white-tailed sea eagles are increasing on Baltic coasts, but there are not yet enough breeding pairs for the population to be considered stable.

The authors concluded that dioxin-like PCBs were now probably the primary contributor to lowered success in reproduction in this species (Koistinen *et al.* 1997b).

There was wide variation in PCB concentrations in white-tailed sea eagles found dead or dying in both inland and coastal areas of Poland, with the highest residues found in the breast muscle of birds from the Baltic coast (PCBs 66-480 ppm fwt). Residues in breast muscle from birds from inland areas were very much lower (4.6-32 ppm fwt) (see table 4-2). In two birds that were particularly highly contaminated, the concentration of total PCBs in the lipid fraction of the breast muscles reached 16000 ppm (1.6%), a very similar concentrations to those found in dead eagles from the Stockholm archipelago in the mid 1960s when concentrations ranging between 150-240 ppm wwt in breast muscle (which translated to 8,900-17,000 ppm, or 0.89%-1.7%, fat) were recorded (Falandysz *et al.* 1994d). The concentrations of co-planar PCBs in adults were the highest ever reported in wildlife, although total PCBs were similar to those reported in dead eagles from the breeding sites of the coastal area of the northern Baltic in Sweden and Finland in the 1960s and 1970s (Falandysz *et al.* 1994d). Similar findings were recorded by another study that compared eagles from coastal and inland areas. In this study white-tailed sea eagles from coastal areas of the Baltic had PCB concentrations in their tissues that were 73 times greater than those from inland areas (1100 ppm fat compared with 15 ppm fat) (Falandysz *et al.* 2000a).

A study of guillemots and white-tailed sea eagles found that concentrations of the main PCB congeners varied between <0.03 and 7.4 ppm fat in guillemot eggs, and between <0.2 and 1,300 ppm in white-tailed sea eagles. When calculated as TEQs for coplanar PCBs these amounted to an average of 5 ppb TEQ fat in guillemots and ranged from 9 to 340 ppb TEQ fat in eagles. The highest TEQ for total PCBs was found in one of the eagles examined and amounted to 1000 ppb fat (Koistinen *et al.* 1995a).

Table 4-2: Concentrations of PCBs in Baltic seabirds and fish-eating raptors

Species	Compound PCBs	References
black cormorant (*Phalacrocorax carbo*)	34 ppm fat	Falandysz *et al.* 2000a
guillemot (*Uria aalge*) eggs	180-260 ppm	Bignert *et al.* 1995
white-tailed sea eagle (*Haliaeetus albicilla*)	66-480 ppm fwt (coast)	Falandysz *et al.* 1994d
white-tailed sea eagle (*Haliaeetus albicilla*)	4.6-32 ppm fwt (inland)	Falandysz *et al.* 1994d
white-tailed sea eagle (*Haliaeetus albicilla*)	0.69-72 ppb iTEQ	Falandysz *et al.* 1994d
white-tailed sea eagle (*Haliaeetus albicilla*)	300-2020 ppm fat	Helander *et al.* 1998
white-tailed sea eagle (*Haliaeetus albicilla*) egg	76-340 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*) muscle	77-1100 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*)	1100 ppm fat (coastal)	Falandysz *et al.* 2000a
white-tailed sea eagle (*Haliaeetus albicilla*)	15 ppm fat (inland)	Falandysz *et al.* 2000a

4.2.2 Organochlorine Pesticides

4.2.2.1 DDTs

The high levels of DDT (and its metabolites) concentrating in birds at or near the top of food chains and the consequent problems, particularly with eggshell thinning, were discovered as long as ago as the 1960s (Ratcliffe 1967). Severe pollution of the Baltic particularly by DDT (and PCBs) engendered a monitoring programme for which guillemot (*Uria aalge*) eggs were used. As some of the fish they eat are themselves migratory throughout the Baltic, black guillemots provide a means of looking at generalised pollution throughout the Baltic (Bernes 1998).

Since DDT use was reduced, and eventually banned, concentrations of this contaminant have declined in guillemot eggs, predominantly during the 1970s. Concentrations in the 1990s were in the range 240-320 ppm fat. Eggshell thickness has also increased over this period, although eggshells remain considerably thinner than in pre-1946 samples, indicating the ongoing influence of this pollutant (Bignert *et al.* 1995).

Concentrations of DDTs have fallen considerably since the mid-1970s when environmental levels were at their highest. One white-tailed sea eagle from the archipelago of Stockholm collected in the mid-1960s was found to have a concentration of DDTs in its body fat of 3.6% (Falandysz *et al.* 1994d). White-tailed sea eagles from coastal areas of the Baltic Sea continue to have significantly elevated concentrations of DDTs in their tissues when compared with birds of the same species from inland areas (780 ppm fat compared with 130 ppm fat). Similarly in another study, values were between 2-30 times greater in eagles from coastal areas than from inland areas of Poland (Falandysz *et al.* 2000a, Strandberg *et al.* 1998c). Fig x.1 shows how concentrations of the DDT metabolite DDE in white-tailed sea eagle eggs that failed to hatch is strongly negatively correlated with the number of successful hatchings at the same nest. In other words the higher the concentration of DDE in unhatched eggs, the fewer successful hatchings occur (Bernes 1998).

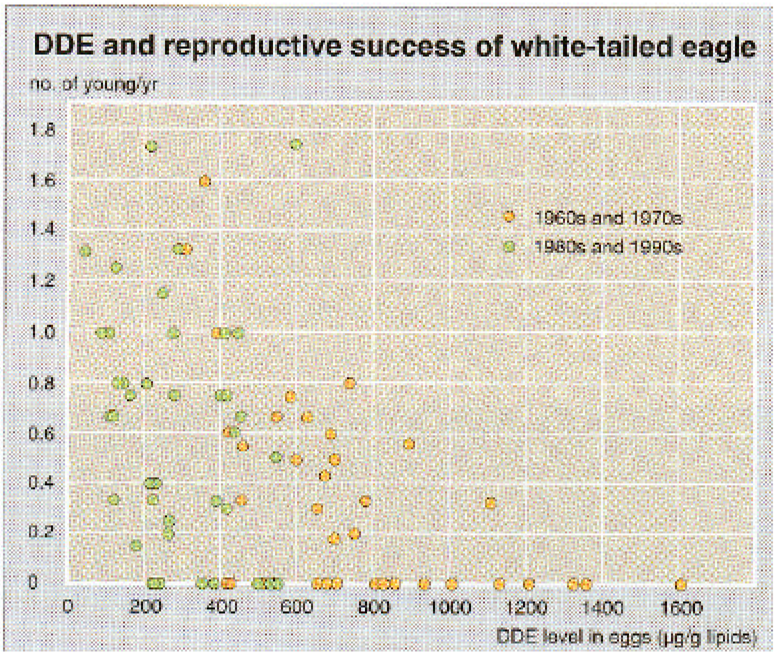


Figure 4.1: Correlation of DDE in eggs of white-tailed sea eagle against number of eggs laid (from Bernes 1998)

Table4-3: DDTs in Baltic Marine Birds

Species	Compound DDTs	References
black cormorant (*Phalacrocorax carbo*) liver	8.3 ppm fat	Falandysz *et al.* 1999b
black cormorant (*Phalacrocorax carbo*) breast muscle	9.5 ppm fat	Falandysz *et al.* 1999b
black cormorant (*Phalacrocorax carbo*)	9.5 ppm fat	Falandysz *et al.* 2000a
black Guillemot (*Uria aalge*) eggs	240-320 ppm fat	Bignert *et al.* 1995
white-tailed sea eagle (*Haliaeetus albicilla*)	<0.1-4400 ppm fat	Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*)	38-4700 ppm fat	Strandberg *et al.* 1998c
white-tailed sea eagle (*Haliaeetus albicilla*)	130-1600 ppm fat	Helander *et al.* 1998
white-tailed sea eagle (*Haliaeetus albicilla*) egg	p,p'-DDD 60-175 ppt fwt p,p'-DDE 5210-9000 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*) muscle	p,p'-DDD 48-721 ppt fwt p,p'-DDE 3518-27500 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*)	780 ppm fat (coastal) 130 ppm fat (inland)	Falandysz *et al.* 2000a
white-tailed sea eagle (*Haliaeetus albicilla*) breast muscle	780 ppm fat	Falandysz *et al.* 1999b
white-tailed sea eagle (*Haliaeetus albicilla*) liver	4500 ppm fat	Falandysz *et al.* 1999b
white-tailed sea eagle (*Haliaeetus albicilla*) egg	270 ppm fat	Falandysz *et al.* 1999b

4.2.2.2 HCB

An average of 470 ppb fat HCB has recently been reported for cormorant tissues (see table 4-4) (Falandysz *et al.* 2000a). This is significantly lower than the residues found in white-tailed sea eagles, for which a maximum concentration of 9200 ppb fat was recorded for one individual from the Baltic south coast (Strandberg *et al.* 1998c). As with many other compounds analysed, eagles from coastal areas had very much higher residues of HCB in their tissues (3300 ppb fat) than did birds from inland areas (190 ppb fat) (see table 4-4), again highlighting the increased exposure of birds living in a marine environment to many different POPs (Falandysz *et al.* 2000a).

4.2.2.3 HCHs

Black cormorants (breast muscle) contained concentrations of 430 ppb fat HCHs (Falandysz *et al.* 2000a). Concentrations of HCHs were similar in the breast muscle of white-tailed sea eagles from both coastal and inland areas (42 and 57 ppb fat respectively) (see table 4-4) (Falandysz *et al.* 2000a). These figures are, of course, taken from different studies and are not directly comparable. However, it is interesting that, of all the compounds reviewed, only HCH's show considerably higher residues (x10) in cormorants than in white-tailed sea eagles.

4.2.2.4 Dieldrin

There are limited data available on dieldrin concentrations in predatory birds in the Baltic area. A concentration of 190 ppb fat was detected in the tissues of a single cormorant from the south coast of the Baltic (Falandysz *et al.* 2000a). White-tailed sea eagles may exhibit far higher tissues residues; one bird analysed had a concentration of 19000 ppb fat dieldrin in its breast muscle. The same study noted, however, that there was a wide variation in residues among eagles from different breeding sites, as well as within sampling areas. In general birds from the Baltic sea coast exhibited the highest concentrations when compared with birds from other areas in Poland (Strandberg *et al.* 1998c). The findings of another study were similar; dieldrin concentrations of 3300 ppb fat (coastal) as compared with 190 (inland) ppb fat were detected in white-tailed sea eagles (see table 4-4) (Falandysz *et al.* 2000a).

4.2.2.5 Chlordanes

Guillemots eggs appear to contain relatively low levels of chlordanes (<30-190 ppb fat). These values are similar to those found in the breast muscle of a cormorant (100 ppb fat) from the southern Baltic coast, although the figures for different tissues are not directly comparable (Falandysz *et al.* 2000a, Koistinen *et al.* 1995a, Strandberg *et* *al.* 1998c). White-tailed sea eagles were highly contaminated with chlordanes. The breast muscle of one bird contained 20000 ppb α-chlordane (Koistinen *et al.* 1995a). White-tailed sea eagles from coastal areas had significantly more chlordanes in their tissues when compared with birds from inland areas (6100 ppb fat and 210 ppb fat respectively) (see table 4-4) (Falandysz *et al.* 2000a).

Table 4-4: Chlordanes, Dieldrin, HCBs and HCHs in Baltic Marine Birds

Species	Compound				References
	HCB	HCH	Dieldrin	Chlordanes	
black cormorant (*Uria aalge*)	470 ppb fat	430 ppb fat	190 ppb fat	100 ppb fat	Falandysz *et al.* 2000a
guillemot (*Uria aalge*) eggs				<3-190 ppb fat	Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*)				110-20000 ppb fat	Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*) breast muscle	77-9200 ppb fat	16-110 ppb fat	70-19000 ppb fat		Strandberg *et al.* 1998c
white-tailed sea eagle (*Haliaeetus albicilla*) egg	51-166-596 ppt fwt			63-313 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*) muscle	38-596 ppt fwt			76-1500 ppt fwt	Koistinen *et al.* 1997b
white-tailed sea eagle (*Haliaeetus albicilla*)	2200 ppb fat (coastal) 530 ppb fat (inland)	42 ppb fat (coastal) 57 ppb fat (inland)	3300 ppb fat (coastal) 190 ppb fat (inland)	6100 ppb fat (coastal) 210 ppb fat (inland)	Falandysz *et al.* 2000a

4.2.2.6 ***Toxaphene***

Few data appear to have been published on levels of toxaphene in Baltic sea birds. One study reported declining concentrations of toxaphene in guillemot (*Uria aalge*) eggs over the period 1976-1989. Concentrations fell from 14 ppm fat in 1976 to 5 ppm fat in 1987, rising very slightly to around 6 ppm fat in 1989. Concentrations in breast muscle, however, did not decline over the same period. This was not, in itself, considered evidence of continuing toxaphene input, but rather that the very much lower concentrations in muscle tissue did not provide so clear a picture of trends in toxaphene residues (Wideqvist *et al.* 1993).

4.2.3 **Other compounds**

4.2.3.1 ***PCDEs in Baltic seabirds and fish eating raptors***

PCDEs are a group of compounds, some congeners of which exhibit toxic properties similar to those found in co-planar PCBs. Similarly too they are ubiquitous in the environment. Individual PCDE congeners ranged in concentration from 3-79 ppb fat in guillemot eggs and from <5-13,000 ppb fat in the breast muscles of eagles. The TEQs for these compounds were also calculated and ranged from 0.1-1.4 ppb fat in eagles, and averaged 0.02 ppb fat in guillemot eggs (see table 4-5) (Koistinen *et al.* 1995a).

Table 4-5: Concentrations of PCDEs and PCNs in Baltic seabirds and fish-eating raptors

Species	Compound		References
	PCDEs	PCNs	
black cormorant (*Phalacrocorax carbo*) breast muscle		75-160 ppb fat	Falandysz *et al.* 1997f
black cormorant (*Phalacrocorax carbo*) liver		160-240 ppb fat	Falandysz *et al.* 1997f
black cormorant (*Phalacrocorax carbo*)		210 ppb fat	Falandysz *et al.* 2000a
guillemot (*Uria aalge*) eggs	0.02 ppb TEQ fat		Koistinen *et al.* 1995a
guillemot (*Uria aalge*) eggs	3-79 ppb fat		Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*)	<5-13000 ppb fat		Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*)	0.095-1.375 ppb TEQ		Koistinen *et al.* 1995a
white-tailed sea eagle (*Haliaeetus albicilla*)		960 ppb fat (coastal) 73 ppb fat (inland)	Falandysz *et al.* 2000a

4.2.3.2 ***Polychlorinated naphthalenes***

In a study conducted on three dead black cormorants (*Phalacrocorax carbo*) found on the Polish coast PCNs were present at between 75-160 ppb fat in breast muscle and between 160-240 ppb fat in liver (see table 4-5). The study concluded that the most likely source of these compounds in the birds was the incineration of municipal solid waste (MSW) and that the PCNs were ingested through their main food source (Falandysz *et al.* 1997f).

White-tailed sea eagles from coastal areas exhibited significantly elevated concentrations of PCNs when compared with birds from inland areas (960 ppb fat as compared with 73 ppb fat respectively) (see table 4-5) (Falandysz *et al.* 2000a).

4.2.3.3 **Tris*(4-chlorophenyl)methane and –methanol (TCPM-H & TCPM-OH)***

It was mentioned in the marine fish section of this report that these compounds are particularly persistent and bioaccumulative. No data were found on TCPM-H/OH in guillemots, but average values of 56 (TCPM-H) and 170 (TCPM-OH) ppb fat were detected in the breast muscle of black cormorants. Still higher values were found in the liver (75 and 550 ppb fat respectively) (Falandysz *et al.* 1998e). As with DDTs, concentrations of TCPM-H and TCPM-OH were significantly elevated in white-tailed sea eagles. Those from coastal areas were particularly contaminated when compared with birds from inland areas (16000 as against 460 ppb fat) (see table 4-6) (Falandysz *et al.* 2000a).

Table 4-6: TCPM-H/OHs in Baltic Marine Birds

Species	Compound TCPM-H/OH	References
black cormorant (*Phalacrocorax carbo*) breast muscle	TCPM-H 56 (25-77) ppb fat TCPM-OH 170 (93-190) ppb fat TCPM-H/OH 230 (120-300) ppb fat	Falandysz *et al.* 1998e
black cormorant (*Phalacrocorax carbo*) liver	TCPM-H 75 (60-82) ppb fat TCPM-OH 550 (530-580) ppb fat TCPM-H/OH 620 (620-630) ppb fat	Falandysz *et al.* 1998e
black cormorant (*Phalacrocorax carbo*)	TCPM-H/OH 230 ppb fat	Falandysz *et al.* 2000a
white-tailed sea eagle (*Haliaeetus albicilla*) breast muscle	TCPM-H mean 5300 (9.4-33000) ppb fat TCPM-OH mean 6700 (1-54000) ppb fat TCPM-H/OH 12000 (13-73000) ppb fat	Falandysz *et al.* 1998e
white-tailed sea eagle (*Haliaeetus albicilla*) liver	TCPM-H 20000 (<1-110000) ppb fat TCPM-OH 5300 (<1-22000) ppb fat TCPM-H/OH 25000 (<1-120000) ppb fat	Falandysz *et al.* 1998e
white-tailed sea eagle (*Haliaeetus albicilla*) adipose fat	TCPM-H 4.5 (2-7) ppb fat TCPMM-OH 10 (9-10) ppb fat TCPM-H/OH 15 (12-16) ppb fat	Falandysz *et al.* 1998e
white-tailed sea eagle (*Haliaeetus albicilla*) egg	TCPM-H 1200 ppb fat TCPM-OH 1300 ppb fat TCPM-H/OH 2500 ppb fat	Falandysz *et al.* 1998e
white-tailed sea eagle (*Haliaeetus albicilla*)	TCPM-H/OH 16000 ppb fat (coastal) TCPM-H/OH 460 ppb fat (inland)	Falandysz *et al.* 2000a

4.2.3.4 ***Butyltins***

The findings of a Polish study indicated very high levels of butyltins in the liver of a long-tailed duck (*Clangula hyemalis*) (4600 ppb wwt) from the Polish coast. Concentrations if butyltins in the livers of other birds collected in the same area ranged between 35-870 ppb wwt (table 4-7). The generally higher concentrations found in these birds when compared with their prey species suggests biomagnification of this compound. Birds are capable of excreting considerable amounts of butyltins by deposition in their feathers. The concentrations found in the birds examined, however, suggested recent exposure to these compounds (Kannan & Falandysz 1997).

Table 4-7: Butyltins in marine birds

Species	Compound BTs	References
red-throated diver (*Gavia stellata*) liver	610 ppb wwt	Kannan & Falandysz 1997
razorbill (*Alca torda*) liver	330 ppb wwt	Kannan & Falandysz 1997
great crested Grebe (*Podiceps cristatus*) liver	540 ppb wwt	Kannan & Falandysz 1997
black cormorant (*Phalacrocorax carbo*) liver	870 ppb wwt	Kannan & Falandysz 1997
long-tailed duck (*Clangula hyemalis*) liver	280 ppb wwt	Kannan & Falandysz 1997
long-tailed duck (*Clangula hyemalis*) liver	4600 ppb wwt	Kannan & Falandysz 1997
guillemot (*Uria aalge*) liver	500 ppb wwt	Kannan & Falandysz 1997
white-tailed sea eagle (*Haliaeetus albicilla*) liver	35 ppb wwt	Kannan & Falandysz 1997

4.3 Trends

Populations of sea eagles crashed during the 1960s and 1970s as a result of pollution by such substances as DDT but these are now slowly recovering (See Figure 4.2) (Bernes 1998, Jansson & Dahlberg 1999). Efforts to limit environmental concentrations of this substance, and more direct measures such as the provision of artificial nests and winter feeding, allowed the Finnish population of white-tailed sea eagles to increase markedly during the 1980s and 1990s from about 40 to about 100 breeding pairs (Jansson & Dahlberg 1999). In neither Finland nor Sweden, however, have numbers reached pre-1950s levels. There is, however, considerable improvement from the period of worst contamination in the 1970s. For example, current concentrations of DDE are approximately 30 times lower than they were in Baltic white-tailed sea eagle in the late 1970s. The range of DDE concentrations in eggs from this species collected in the Baltic between 1978-82 ranged from 245-1900 ppm fresh weight (fw) with a mean of 825 ppm fw. Total concentrations of PCBs have also declined significantly from a mean of 1110 ppm fw in 1980 (and individual concentrations as high as 2200 ppm fw). Concentrations of individual PCB congeners in the mid-1990s were down by between 80 and 97% compared with 1982 (Koistinen *et al.* 1997a).

Guillemot eggs have been used in the Swedish Contaminant Monitoring system because they provide measure of toxic pollutant levels in the Baltic sea as a whole. The guillemots sampled obtain their food from numerous different populations of herring and other fish and the levels of contaminants in the eggs provide, therefore, a useful integration of those occurring in the Baltic fish populations. In the eggs of guillemots (*Uria aalge*), concentrations of p,p'-DDT and PCBs have decreased to about 5% and 15% respectively of those that existed in the late 1960s and early 1970s. That PCB concentrations have not declined as fast as those of DDT in most species surveyed is thought to be because PCBs continue to enter the system (EEA 1998, Bernes 1998, Valters *et al.* 1999).

4.3.1 Reproductive and non-reproductive effects of POPs in marine birds

High concentrations of POPs are known to be lethal to various bird species. This has been confirmed both by experiment and field observation. Female birds pass on large amounts of PCBs to their eggs, and contamination by PCB of eggs has been associated with eggshell thinning, much as with DDTs. White- tailed sea eagles failed altogether to reproduce in the southwestern Baltic in the 1960s and increased their reproductivity only to 0.17 in the 1970s (compared with normal levels of 0.80-1.60). Observations made on the breeding success of white-tailed sea eagle pairs have been carried by the Swedish authorities since populations plummeted in the 1960s (See Fig x.2). It is clear that some improvements have occurred as successful breeding has risen considerably, though not to the levels that existed before the introduction of DDT (Bernes 1998, Helander *et al.* 1998). The concentrations of toxic PCBs found continued to be high enough to affect breeding success in this species (Falandysz *et al.* 1994d).

Table 4-8: Environmental impacts on marine birds and possible causative agents (Adapted from Swedish EPA, 1996)

Observation/Impact	Sensitive Species	Substance	Association/Causation
Eggshell thinning	guillemot, eagle, osprey, peregrine falcon	DDT	5
Reproductive disturbances	osprey	DDT, PCB	4-5
Reproductive disturbances	eagle	DDT, PCB	2-3

Association/causation is assessed on the scale: 1 = no observed association, 2 = suspected association, 3 = weak association, 4 = clear association, 5 = significant association

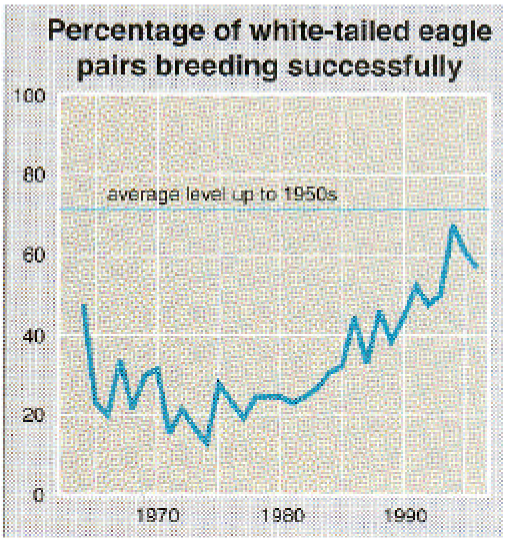


Figure 4.2: Trends in breeding success of white-tailed sea eagles in Sweden (from Bernes 1998)

PCBs were identified as environmental pollutants in a white-tailed sea eagle found dead in the Stockholm archipelago in the late 1960s. The reproductive success of white-tailed sea eagles from the Baltic region was subsequently shown to be negatively correlated to levels of persistent organochlorines in their eggs (Vos *et al.* 2000). Many lipophilic chemicals such as PCBs and PCDD/Fs are readily taken up and excreted in the egg yolk in laying birds, thus exposing the embryo to high levels of toxic chemicals during the earliest stages of development. There are several examples of pollutant-induced toxicity in wild bird populations following exposure to PCBs and PCDFs, but it is difficult in field situations to determine the mechanism of action of these chemicals or indeed which ones are primarily responsible when complex mixtures are involved, as is generally the case in the wild (Ratcliffe 1967, Vos *et al.* 2000).

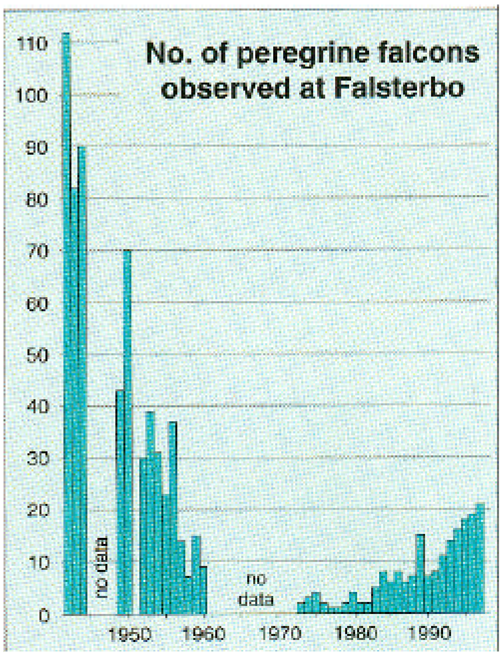


Figure 4.3: Trends in numbers of peregrine falcons in an area of Sweden (from Bernes, 1998)

The problems associated with high levels of endocrine disruptors such as DDT (and its metabolites DDD and DDE) for birds of prey was first pinpointed in the United Kingdom. Significant thinning of avian eggshells from 1946 onwards was revealed by Ratcliffe (1967) (Ratcliffe 1967) and was later attributed to DDT. Indeed, it was the expansion of DDT use after the second world war that was eventually found to be the cause of falling bird of prey populations throughout Europe and North America (Ratcliffe 1967, Vos *et al.* 2000). Populations of certain species such as the

Peregrine falcon (*Falco peregrinus*), the osprey (*Pandion haliaetus*) and the white-tailed sea eagle had also crashed in the Baltic region (See Fig 4.3) (Bernes 1998). The greatest threat to birds of prey such as the osprey and the white-tailed sea eagle is not direct poisoning by pesticides and other POPs. Much more seriously, there occurs a dramatic increase in reproductive failure, even in seemingly healthy adult pairs with well below lethal levels of toxicants in their tissues. They continue to lay the same number of eggs, but fewer of them hatch, primarily because their shells become so thin that they are broken during incubation. In some species, particularly ospreys (table 4-8), DDE also appears to have resulted in delayed ovulation, which in turn delays hatching. This effect contributed to the poor breeding success of ospreys, particularly during the 1970s (Bernes 1998). A further effect of eggshell thinning appears to have been an abnormal rate of dehydration in the eggs, an effect that persists in females born during the 1960s-1970s, despite otherwise normal POPs residues in their tissues and eggs, that is thought to be a result of their previous exposure to PCBs and DDTs (Helander *et al.* 1998).

5 POPS IN BALTIC MARINE MAMMALS

Marine mammals such as seals, whales and dolphins accumulate relatively high body burdens of POPs. The accumulation of POPs in marine mammals, in particular organochlorines, has given rise for concern about the long-term impact of these chemicals on these animals (see Reijnders 1996).

Marine mammals are susceptible to the accumulation of POPs in their tissue due to their position in the food chain, their high quantities of fatty blubber tissue and the passage of POPs from females to their young. POPs accumulate in their thick layer of fatty blubber and, once retained in such fat-rich tissue, they are not easily eliminated. POPs may be released from fat, however, both during periods of starvation or weight loss and during lactation. The milk of marine mammals has a high fat content and a high proportion of POPs stored in the body of a female may be passed to her offspring during lactation (see e.g. Tanabe *et al.* 1994). Yet another reason for the relatively high levels of POPs in marine mammals is that some marine mammals are more susceptible to bioaccumulation of organochlorines due to their limited capacity to break down contaminants such as PCBs (Tanabe *et al.* 1994, Tanabe *et al.* 1988).

Levels of contaminants in marine mammals have been reported for decades throughout the world's ecosystems. The most frequently measured POPs in marine mammals are organochlorines such as DDT and PCBs (Norstrom & Muir 1994). Other POPs, such as organotins and polybrominated flame retardants have been less frequently monitored in marine mammals.

Marine mammal species which inhabit the Baltic Sea include the grey seal (*Halichoreus grypeus*), ringed seal (*Pusa hispida*), harbour or common seal (*Phoca vitulina*) and the Baltic porpoise (*Phoceona phoceona*). The grey seal is the most commonly occurring species in the Baltic Sea. The ringed seal is rare and the harbour seal occurs mainly in the Kattegat and Skagerrak. The Baltic porpoise occurs mainly in the west (Falandysz *et al.* 2000a).

5.1 Tissue Levels of Pops in Marine Mammals

5.1.1 Seals

Several persistent organochlorines and other POPs have been detected in seals from the Baltic Sea. Levels are comparatively high in Baltic seals, reflecting the proximity to industrial and agricultural sources and the generally high degree of contamination of the Baltic sea. DDT and PCB levels are particularly elevated in Baltic seals; for example, levels are 15-20 times higher than in seals from Scotland, Norway and Canada (Jenssen 1996).

5.1.1.1 ***PCDD/Fs***

Analyses of seal blubber from the Baltic Sea and from the Swedish west coast during the 1980s revealed that levels of PCDD/Fs were similar in both locations see Bergek *et al.* (1992). Another study conducted on seals collected from the east and west coast of Sweden between 1979 and 1990 also revealed the same result (Bergek *et al.* 1992). This is surprising because fish that are prey for seals of the Baltic Sea are much more highly contaminated with PCDD/Fs than fish on the Swedish west coast. Moreover, the higher contamination of fish in the Baltic Sea with other organochlorines such as PCBs and DDT is reflected in relatively high tissue levels of these compounds in Baltic seals compared to those from other regions. Further investigations will be required, therefore, in order to determine the underlying reasons for the relatively constant dioxin levels.

A study on ringed seals collected from Bothnian Bay in 1997 reported that levels of PCDD/Fs in blubber of Baltic ringed seals were significantly higher (by 35-times) (e.g.164 ppt WHO-TEQ fat in males) than ringed seals from the Arctic Ocean collected from Spitsbergen (4.6 ppt WHO-TEQ fat in males). The differences in the levels of PCDD/Fs, and in the pattern of dioxin congeners found in the seals at the two sites implied different sources of these chemicals in the two regions. There are numerous sources of dioxins in the Baltic Sea compared to the Arctic environment, which likely receives dioxins mostly through atmospheric deposition (Koistinen *et al.* 1999).

Research conducted by Bergek *et al.* (1992) showed that blubber from Baltic ringed seals contained noticeably higher levels of PCDD/Fs compared with blubber from grey seals and harbour seals. For example, the total concentration of PCDD/Fs in blubber from adult male Baltic seals was 39 ppt Nordic-TEQ fat in grey seals and 455 ppt Nordic-TEQ fat in ringed seals. Similarly, levels in ringed seals collected from the Gulf of Finland in 1991-2 were higher (range 45 to 150 ppt I-TEQ fat) than grey seals from the same area (range 12 to 61 ppt I-TEQ fat) (Koistinen *et al.* 1997c). The cause of the difference in tissue levels of PCDD/Fs between ringed seals and grey seals may be due to differences in feeding habits between the two species or to differences in the rate of metabolism/elimination of PCDD/Fs.

Despite variations between species, there do not appear to be any strong spatial trends in levels of PCDD/Fs in seals within the Baltic region. For instance, a study on seals from the Gulf of Finland in 1991-2 reported levels of PCCD/Fs in grey seals ranged from 12 to 61 ppt I-TEQ and ringed seals from 50 to 150 ppt I-TEQ. This range of PCDD/F concentrations was similar to levels found in seals from other areas in the Baltic Sea and in Lake Saimaa (Koistinen *et al.* 1997c).

With regard to time trends of PCDD/Fs in Baltic seals, the available data suggests no decline in levels. For example, Koistinen *et al.* (1999) noted that levels of PCDD/Fs in Baltic seals in 1997 did not differ significantly from levels found in the 1980s.

5.1.1.2 ***PCBs***

Comparatively high levels of PCBs have been reported in Baltic seals. Blomkvist *et al.* (1992) made a study of grey, ringed and harbour seals from Swedish waters, reporting that levels of PCBs in the blubber of harbour seals from the Baltic were twice as high as levels in their counterparts on the Swedish west coast.

Blomkvist *et al.* (1992) also found significant differences in the levels of PCBs between the three Baltic seal species. For example, the mean PCB levels in blubber from juvenile seals were 77 mg/kg (ppm) fat for grey seals, 36 ppm fat for harbour seals and 17 ppm fat for ringed seals. The authors noted that differences in the levels of PCBs in the species could be explained by their different feeding habits.

Levels of PCBs and certain other persistent organochlorines in seals also vary with age and sex. Tissue concentrations increase with age and body size in males. For instance, a level of 210 ppm fat was measured in Baltic adult male ringed seals compared to 17 ppm fat for juveniles (Blomkvist *et al.* 1992). In females, concentrations are reduced after they have given birth and suckled offspring (Aguilar 1988, Cockcroft *et al.* 1989).

Comparison of PCB levels between studies is difficult because there are often differences in the PCB congeners (compounds) which are analysed in different studies. However, it has been noted that, in general, similar concentrations of PCBs have been reported for seals from different regions of the Baltic Sea. For instance, a study in the Gulf of Finland noted that levels of PCBs in ringed seals and grey seals were similar to levels reported for seals from Lake Saimaa in Finland, and were not high compared to other studies on Baltic seals (Koistinen *et al.* 1997c). A study undertaken on Baltic seals collected from coastal areas of Estonia in 1994 also revealed that the blubber of grey seals from the Vilsandi National Park contained similar concentrations of PCBs to the blubber of seals from other areas of the Baltic (Roots 1998, Roots & Talvari 1999). In the Vainameri Sea off the coast of Estonia, however, levels of PCBs were comparatively lower in grey seals, possibly due to the availability of different prey.

At lake Ladoga, the largest lake in Europe located in Russia, near to the Baltic Sea, research has been conducted on a subspecies of ringed seals (*Phoca hispida ladogensis*) (Kostamo *et al.* 2000). Levels of PCBs in the seals were found to be lower than in seals from Lake Saimaa and the Baltic Sea. This was not surprising given that levels in fish from the Lake are notably lower than in fish from the Baltic.

Blomkvist *et al.* (1992) compared results of their study on levels of PCBs in juvenile Baltic seals with previous studies to determine time trends of PCBs contamination. The comparison indicated that PCB levels in ringed seals from the Gulf of Bothnia had decreased significantly since the early 1970s. However, no decline was evident for grey seals over this same period. This result was unexpected because levels of PCBs in fat of some Baltic fish had decreased by about 50% since the early 1970s. Further research by Roos *et al.* (1998) suggested a possible explanation for the absence of a decline in PCBs in the Baltic grey seal, with the observation that female seals with concentrations of PCBs above a certain threshold level in their tissues could not breed. This and other effects of high PCB contamination are discussed further in section 5.2.1.

Roos *et al.* (1998) investigated levels of PCBs in juvenile grey seals from the Swedish Baltic coast between 1989 and 1997. Comparison of the results with previous studies indicated that between 1969 and 1997, levels of PCBs in grey seals overall had declined only at a slow rate of 2% per year in pups and 4% per year in juvenile grey seals. By contrast, the rate of decrease monitored during a similar time period in guillemot eggs and in herring from the Baltic was 9% per year.

A study on seals from Lake Ladoga, Russia, showed no decline in PCB levels (Kostamo *et al.* 2000), with concentrations similar to those recorded 15 years previously.

5.1.1.3 ***Other PCB compounds***

PCB methyl sulfones are metabolites (breakdown products) of PCBs that are highly persistent. These compounds have been identified in blubber from harbour, ringed and grey seals from Swedish waters of the Baltic Sea (Haraguchi *et al.* 1992). The concentrations of PCB methyl sulfones were found to be only slightly lower than those of PCBs themselves, ranging from 1 to over 110 ppm fat.

Haglund *et al.* (1995) reported the presence of brominated PCBs in blubber of a ringed seals collected from the Swedish coastline of the Baltic Sea between 1980-7. These compounds had not previously been reported in biological samples. The study reported an average concentration of 26 ng/g (ppb) of brominated PCBs, about 1000-fold lower than total PCBs in ringed seal blubber. Due to analytical factors which might have led to significant underestimation of concentrations, this level was thought to represent a minimum level in the seal blubber. There are a number of possible sources of brominated PCBs in the environment, including trace contamination of PCB formulations or formation as by-products from municipal and chemical waste incineration. Based on their structural similarity to PCBs, it is very plausible that brominated PCBs have similar toxicity to PCBs.

5.1.1.4 ***DDT***

Comparatively high levels of DDT compounds have been reported for Baltic seals compared to seals from other areas. For instance, a study on seals from Swedish waters reported that levels of DDTs were 4-times greater in the blubber of juvenile Baltic harbour seals, (mean 27 ppm fat), than in juvenile harbour seals from the Swedish west coast (4.1 ppm fat at Skagerrak and 6.9 ppm fat at Kattegat) (Blomkvist *et al.* 1992). Levels in ringed seals (13 ppm fat) were about 3-times higher and levels in grey seals (35 ppm fat) were the highest of all.

Like PCBs, the concentration of DDTs varies with age and sex in seals such that adult seals accrue higher levels than juveniles and females have reduced levels following pregnancy and lactation. In a study on Baltic grey seals Blomkvist *et al.* (1992) reported that the highest levels of DDT (mean 560 ppm fat) were found in adult female seals that were of poor nutritional status. These seals only have a thin layer of blubber and/or a low amount of extractable fat in blubber. The high DDT levels most likely occurred in these starved female seals because DDT cannot be broken down in their bodies at the same rate as the fat stores are metabolised and so this contaminant becomes more concentrated in the blubber.

A study on Lake Ladoga in Russia found that levels of DDT in blubber of the Ladoga seal were lower than levels in seals from Lake Saimaa (Finland) and the Baltic Sea (Kostamo *et al.* 2000). Levels in seal blubber ranged from 8.4 to 16.7 ppm fat. This was not unexpected because Lake Ladoga is not as highly contaminated with these chemicals as the Baltic Sea.

With regard to time trends, Blomkvist *et al.* (1992) reported a significant decrease in levels from the early 1970s to the late 1980s in Baltic grey seals and ringed seals. Another study by Roos *et al.* (1998) reported a decline in DDT levels in juvenile grey seals from the Swedish Baltic between 1989 and 1997. Between 1969 and 1997 the rate of decline in DDT levels in pups and juveniles was respectively 11% and 12% per

year. In contrast to this data from the Baltic, no downward trend in DDT levels was found during the past fifteen years in seals from Lake Ladoga in Russia (Kostamo *et al.* 2000).
`

5.1.1.5 ***Organochlorine Pesticides***

Aside from DDT, other organochlorine pesticides have been detected in Baltic seals. Andersson & Wartanian (1992) found chlordane compounds and toxaphene in grey, ringed and harbour seals collected from the Swedish Baltic coast during the 1980s. Previous studies had reported toxaphene and chlordane levels in the range of 1 to 10 μg/g (ppm) fat in Baltic seal blubber and similar levels were reported by Andersson and Wartanian (1992). Levels of both chlordanes and toxaphene were higher in Baltic seals than in seals from the west coast of Sweden. For example, the levels of chlordanes and toxaphene in juvenile harbour seals from the Baltic were 3.6 and 1.8 ppm fat respectively compared with 0.47 and 0.5 ppm fat in the Skagerrak.

Among the Baltic seals, levels did not generally vary greatly between seals of different ages and sex. However, ringed seals had generally higher levels of chlordane and toxaphene that harbour and grey seals. It was suggested that this could be due to higher environmental pollutant levels in the northern part of the Baltic which is inhabited by the ringed seals or alternatively to a different ability between the species to breakdown or eliminate the organochlorines.

Levels of α-HCH and γ-HCH (lindane) were found to be significantly higher (by at least 2-times) in blubber from Baltic harbour and grey seals than the same species of seals from the North Sea and Iceland (Hummert *et al.* 1995). Kostamo *et al.* (2000) reported that HCB (below detection limit to 0.04 ppm fat) and lindane (0.02 to 0.04 ppm fat) were detected in seals from Lake Ladoga in Russia.

5.1.1.6 ***PBDEs and PCDEs***

PBDE's have been detected in seal blubber from the Baltic Sea. Andersson & Wartanian (1992) reported levels of 0.23, 0.28, and 0.32 ppm fat respectively in juvenile harbour, grey and ringed seals collected from the Swedish Baltic coast throughout the 1980s. The levels were a little higher in the Baltic seals than levels reported in juvenile harbour seals from the Swedish west coast. Within the Baltic species, adult and juvenile seals had similar levels of PBDEs in blubber. However, levels 2 to 3-times higher were found in diseased adult female seals which had reproductive impairments (occluded uteri) (see further on reproductive impairments in section 5.2.1.1).

Another study reported similar levels of PBDE's in ringed seal (0.38 ppm fat) and grey seal (0.468 ppm fat) blubber collected from the Swedish Baltic coast between 1981 and 1988 (Haglund *et al.* 1997) to those reported by Andersson & Wartanian (1992). In addition, this study reported the presence of methoxy-PBDEs in the seal blubber. Levels of 0.22 and 0.12 ppm fat methoxy-PBDEs were reported in grey seal and ringed seal blubber respectively. The source of methoxy-PBDEs is currently unknown but multiple sources are possible, including breakdown products (metabolites) of PBDEs, microbial degradation of PBDEs and a variety of industrial sources.

In addition to PBDEs, polychlorinated diphenyl ethers (PCDEs) have also been detected in seal blubber from the Baltic Sea. These compounds were detected at ppb (fat) levels in blubber of ringed seals and grey seals collected from the Gulf of Finland in 1991-2 (Koistinen *et al.* 1997c). The level of these compounds was higher than levels of PCDD/Fs in seal blubber and of some, but not all, PCB congeners. Another study (Haglund *et al.* 1997) confirmed the presence of PCDEs as well as methoxy-PCDEs compounds in liver tissue of ringed seal that was collected from the Swedish Baltic coast in the 1980s.

5.1.1.7 *Other POPs*

Tris(4-chlorophenyl) methane and *tris*(4-chlorophenyl) methanol, are persistent environmental contaminants that have recently been identified in humans and several animal species throughout the world. The main source of these compounds may be technical grade DDT but their persistence appears to be much greater than DDT and DDE (Falandysz *et al.* 1999). *Tris*(4-chlorophenyl) methanol was identified in blubber of grey seal from the Baltic Sea (Haraguchi *et al.* 1992). Both *tris*(4-chlorophenyl) methane and *tris*(4-chlorophenyl) methanol were also detected in ringed seal tissues from the Baltic Sea in ppb to ppm levels in a study by Zook *et al.* (1992).

Another compound, bis(4-chlorophenyl) sulfone (BCPS) was found in grey seal blubber from the Swedish Baltic at concentrations of 53 - 88 ppb fat (Olsson & Bergman 1995). The study noted that it was the first time that BCPS has been reported as a contaminant in wildlife. This environmentally persistent contaminant had, up to the time of the study, been previously overlooked. BPCS is used in the production of high temperature polymers which are used in a variety of products from components in electronics to pots for microwave oven cooking and aircraft fixtures. The production of high temperature polymers may be an important source of BCPS in the environment. It may also still be used as a component in reactive dyes for the textile industry. BPCS is structurally similar to certain DDT compounds, namely p,p'-DDT, p,p'-DDE and p,p'-DDD and its insecticidal activity is similar to that of DDT, although there are few data on toxicology of the compound (Olsson & Bergman 1995).

Since the mid-1990s, an organochlorine designated Q1, of unknown structure but with the elemental composition $C_9H_3Cl_7N_2$, has been detected in biota from several regions of the world though, in Baltic grey seals, levels were comparatively low (Vetter 1999). Research has indicated that Q1 represents a unique case of an environmental contaminant organochlorine as it is more abundant in the remote area of the Antarctic than in the otherwise highly polluted waters of middle and northern Europe.

5.1.2 **Baltic Harbour Porpoise**

Harbour porpoises feed at high levels in the food chain and have a small body size. These factors contribute to accumulation of high levels of POPs in their body tissues.

In Swedish waters the harbour porpoise is listed as a vulnerable species and was included on the national list of protected species by the Swedish Environmental Protection Agency in 1973 (see Börjesson & Berggren 1997). In the Baltic Sea, the harbour porpoise has some small biological differences from harbour porpoise in neighbouring seas and so are considered to belong to a separate population. At the

beginning of the century, the Baltic porpoise was widely distributed throughout the Baltic Sea (see Siebert *et al.* 2001). However, the Baltic harbour porpoise population was reported to undergo a drastic decline after the 1940s. Possible reasons mentioned for this decline include hunting (Börjesson & Berggren 1997), and high tissue levels of persistent organochlorines (see Strandberg *et al.* 1998a). Presently, the status of Baltic harbour porpoise is potentially under threat from numerous pressures, including bycatches in fishermen's nets (Börjesson & Berggren 1997), low population numbers, high level of persistent organochlorine contamination, food depletion and human disturbance (Berggrena *et al.* 1999). Indeed, with regard to organochlorine contaminants (specifically PCDD/Fs, PCBs and DDT), a recent study has noted,
" *that the contaminant levels recorded in the Baltic sea are a serious cause for concern and could have management implications for the already threatened status of the harbour porpoise in this area*" (Berggrena *et al.* 1999).

As in many other cases with marine mammals, researchers have studied levels of POPs in harbour porpoises using tissues from specimens that have either been found stranded on the coast or killed accidentally in fishing nets. As in seals, tissue levels of some POPs in porpoises increase with age but decrease in females after they become pregnant as a result of both placental transfer to the foetus and passage in milk to the new born.

5.1.2.1 ***PCDD/Fs***

Falandysz *et al.* (1997d) reported a mean level of 5.6 pg/g (ppt) fat in the blubber of 4 harbour porpoises from the Polish coast in 1991-1993. The age and sex of the animals was not recorded in this study and consequently comparison of these results with other studies is difficult. Another study investigated PCDD/F levels in 47 male harbour porpoises from the Swedish Baltic Sea between 1985-93, and from the Kattegat-Skagerrak Seas and west coast of Norway between 1978-1993 (Berggrena *et al.* 1999). This study reported that somewhat higher levels of PCDD/Fs were found in the Baltic harbour porpoise compared to porpoises from other regions, especially in the case of mature animals. For example, mean levels in immature (age 1-3 years) porpoises from the Baltic were 13 ppt fat compared with 9.2 ppt fat in the Kattegat-Skagerrak. In mature porpoises (age 4 or older), a mean level of 36 ppt fat was recorded for Baltic animals compared to 16 ppt fat in the Kattegat-Skagerrak region and 12 ppt fat in the west coast of Norway. Unpublished data cited by Berggrena *et al.* (1999) has also indicated that higher levels of PCDD/Fs occur in male porpoises from the Baltic Sea (16-18 ppt fat) compared to their counterparts from the west coast of Sweden (3-9 ppt fat).

A lack of older data on Baltic harbour porpoises precludes the determination of trends in PCDD/F levels. However, Berggrena *et al.* (1999) noted that levels of PCDD/Fs, PCBs and DDT in Baltic porpoises are still as high as they were a decade ago in porpoise from the Kattegat-Skagerrak. It was suggested that levels of PCDD/Fs plus dioxin-like PCBs may be sufficient to cause adverse effects on the immune system of Baltic harbour porpoises. For example, the maximum concentration of PCDD/Fs and dioxin-like PCBs expressed using the Nordic TEQ system to estimate toxicity was 208 ppt Nordic-TEQ fat. This can be compared to a threshold TEQ of 209 ppt fat in Baltic harbour seals which has been demonstrated to cause suppression of the immune system under experimental conditions.

5.1.2.2 ***PCBs***

Berggrena *et al.* (1999) investigated PCB levels in 47 male harbour porpoises from the Swedish Baltic Sea, between 1985-93, and from the Kattegat-Skagerrak and west coast of Norway, between 1978-1993. This study reported that higher levels of PCBs were found in the Baltic harbour porpoises compared to those from other seas. For instance, total PCBs in the blubber of mature Baltic porpoises were 3-times higher than levels in porpoises from Kattegat-Skagerrak and Norweigian west coast. The mean total PCB concentration in Baltic porpoises was 46 μg/g (ppm) fat compared to 13 ppm fat in their counterparts from Kattegat-Skagerrak and 15 ppm fat from Norwegian west coast. The authors commented that the higher PCB levels in harbour porpoises from the Baltic was not surprising given that the Baltic aquatic environment contained higher levels of PCBs than the Kattegat-Skagerrak. Another study (Falandysz *et al.* 1994a) reported the presence of PCBs in Baltic harbour porpoises collected in 1989-90 from the inner Gulf of Gda sk, Poland.

According to Berggrena *et al.* (1999), it is not possible to establish whether the levels of PCBs have decreased in Baltic harbour porpoises over time due to a lack of data from pervious years. However, as noted for PCDD/Fs (see above) and DDT in mature Baltic porpoises, the study did indicate that levels in porpoises from the Kattegat-Skagerrak Sea are still as high as they were a decade ago. The authors stressed that the ranges of concentrations of total PCBs in Baltic harbour porpoises (46 ± 29 ppm fat) spanned the concentration (>50 ppm fat) which has been proposed as a health risk to these animals. Furthermore, total PCB levels greatly exceeded levels which have been estimated to cause adverse impacts on the nervous system (neurobehaviour).

5.1.2.3 ***Organochlorine Pesticides***

DDT compounds have been identified in blubber of harbour porpoises collected from the Swedish Baltic in 1985-93 (Berggrena *et al.* 1999) and in the southern part of the Baltic Sea in 1991-3 (Falandysz *et al.* 1997d). Berggrena *et al.* (1999) reported that levels of total DDTs in mature Baltic harbour porpoises were higher than their counterparts in the Kattegat-Skagerrak waters and the west coast of Norway. For example, the mean total DDTs in blubber of Baltic porpoises was 116 ppm fat compared to 25 ppm fat in Kattegat-Skagerrak and 9.1 ppm fat in the west coast of Norway. The authors noted that this result was not unexpected given the higher contamination of the Baltic Sea with DDT than the Kattegat-Skagerrak.

A study on harbour porpoises collected in 1986-88 from Danish waters, including the Baltic and North Sea, noted that levels of total DDTs appeared to have declined when compared to data from the mid-1970s and early 1980s (Granby & Kinze 1991).

Chlordane compounds, HCB, HCHs, dieldrin and mirex were all detected in the blubber of 4 harbour porpoises collected in the southern part of the Baltic in 1991-3 (Strandberg *et al.* 1998a). It was noted that levels of HCHs, HCB and dieldrin and some of the chlordane compounds were higher in the Baltic porpoises compared with levels reported in a previous study on harbour porpoises from Danish and Norwegian waters in 1987-90. Of all the organochlorines analysed in Baltic porpoises in this study, PCBs and DDTs were found at the highest levels.

5.1.2.4 *Other POPs*

The organotin compound tributyltin (TBT) accumulates in the fat of animals as well as in organs such as the liver and kidney. Kannan & Falandysz (1997) analysed TBT in liver tissue taken from two neonatal porpoises from the Polish coast in 1991 that had been accidentally killed in fishing nets. Levels of TBT in the porpoise liver tissues were 18 and 27 ng/g (ppb) wet weight. These levels were substantially lower than levels found in porpoises from Japan (1120-10200 ppb wet weight) and dolphins from the Mediterranean (1200-2200 ppb wet weight). However, direct comparison in this case is not very relevant as the Baltic porpoises were analysed at a much younger stage of life. Phenyltins were also detected in the Baltic harbour porpoises.

Tris(4-chlorophenyl) methanol and *Tris*(4-chlorophenyl) have also recently been identified in the blubber of harbour porpoises collected from the southern Baltic Sea (Falandysz *et al.* 1999b).

5.2 Biological Effects of POPs in Marine Mammals

It is difficult to assess the impact of contaminants on wildlife populations, not least because individual animals are exposed to complex mixtures of chemical contaminants. Nevertheless, in some instances, immunological and reproductive disorders in marine mammals have been linked to elevated levels of organochlorine chemicals. These effects have undoubtedly contributed to the dramatic declines in some populations of marine mammals from northern temperate latitudes, including the Baltic Sea.

It is of particular concern that POPs are transferred from females to their young because the early stages of life are commonly the most vulnerable to such chemicals (e.g. Colborn *et al.* 1993, vom Saal *et al.* 1992). POPs are passed *via* the placenta to the developing foetus in the womb and *via* milk to the suckling young. Exposure to POPs during the developmental stages of life may lead to adverse effects on health that are irreversible (Colborn *et al.* 1993).

Reproductive and immune disorders for which PCBs, DDT and their respective metabolites have been implicated have been identified in wild populations of seals around the world, including in the Baltic. Disruption of the endocrine system may be an important underlying mechanism resulting in effects on reproductive and immune development and function (Reijnders 1994).

5.2.1 **Seals**

Baltic seal populations have been severely impacted by reproductive impairment and has also suffered from a devastating disease-complex in recent decades. Effects on reproductive success are suspected to be caused by persistent organochlorines. These same chemical contaminants may also have played a part in exacerbating thousands of deaths of seals caused by a virus in the Baltic and other regions in 1988 by impairing the immune system of seals.

5.2.1.1 *Reproductive Failure and Disease-Complex*

Seal populations in the Baltic Sea have declined dramatically during the last century. While much of the early decline was caused by over-hunting, the main reason for the decline in the past three to four decades has been reproductive failure in female seals (Bergman 1999, Harding & Harkonen 1999). More recently, reproductive failure may be slowing the recovery from earlier population declines. This effect, as well as other adverse effects on health of Baltic seals, has been associated with pollution with PCBs, DDT and their respective metabolites.

Reproductive failure in female Baltic seals was first noted in the 1970s. During this time there was a sharp decline in the population of both ringed and grey seals. Only 27% of females were pregnant compared to a usual figure of 80-90% (Helle *et al.* 1976a). The seals were found to have pathological changes in their uteri, including occlusions (closure) and stenoses (narrowing) (Helle *et al.* 1976b). These obstructions of the uterus most likely develop after interrupted pregnancy and may be associated with hormonal imbalance and suppression of the immune system, probably due to impacts of organochlorines (Bergman 1999).

Further investigations showed that other adverse effects were also commonly occurring in Baltic seals. Symptoms included changes in skull shape (Zakharov & Yablokov 1990), loss of bone in the skull (osteoporosis), loss of teeth, lesions in the claws, ulceration of the intestines (often fatal), adverse effects on the kidney and arteries, uterine muscle cell tumours (leimyomas) and enlargement of the adrenal gland (adrenocortical hyperplasia). Studies revealed that all these effects, along with the reproductive effects observed in females, were part of a disease-complex called hyperadrenocorticism (Bergman 1999, Reijnders & Brasseur 1992). Research suggested that organochlorines, in particular PCBs, can interfere with the hormonal system to cause these effects (see Reijnders & Brasseur 1992).

At the beginning of the 1980s, seal numbers in the Baltic Sea were very low. The number of grey seals was estimated to be 1000-1500, ringed seals 10,000 and harbour seals 200. Recent research shows that the number of grey seals has increased although numbers are still low compared with historical estimations of over 88,000 to 100,000 animals at the beginning of the century (Bergman 1999, Harding & Harkonen 1999). In 1995, numbers of grey seals were estimated as 5300, ringed seals 5500 and harbour seals 575. Concomitant with the increase in grey seal numbers, a recent study has reported improvement in reproductive health and some other problems related to the disease complex in Baltic grey seals. At the same time, however, there has been an increased prevalence of colonic ulcers (Bergman 1999). The improvement in effects of the disease complex in grey seals may be explained in part by the decrease in levels of PCBs and DDTs in Baltic biota from the very high levels which occurred in the early 1970s.

Bergman (1999) investigated the health of Baltic grey seals using postmortem examinations. 159 grey seals were examined over the period 1977-1996 and trends in disease complex symptoms were reported. Overall, the research showed that a high prevalence of lesions belonging to the disease complex still exists in Baltic seals. The prevalence of uterine occlusions and stenoses in females has declined over time and the rate of pregnancy had increased from 9% in the period of 1977-86 to 60% during 1987-96. In animals born after 1980, compared to those born prior to this date, the incidence of uterine tumours had decreased slowly from 53% to 43%. This means, however, that incidence of the tumour remains very high. There was a decline in the prevalence of claw lesions and adrenocortical hyperplasia in the seals over time, though neither decline was statistically significant. A decrease in the prevalence and degree of skull bone lesions was also identified in seals after 1980.

Colonic ulcers, becoming more frequent in grey seals in recent years, particularly in juveniles aged 1-3 years, accounted for the second greatest cause of death (7%) among the seals examined in this study after drowning in nets (72%) (Bergman *et al.* 1999). Although these ulcers are probably initiated by a parasite, this parasite does not normally cause gross intestinal changes such as those observed in Baltic seals. Bergman *et al.* (1999) commented that the high prevalence of colonic ulcers found in Baltic grey seals indicates an increased negative impact on their immune system. This increase in ulcers has occurred despite a reduction in some persistent organochlorines in the Baltic, and might indicate that the diet of the seals now contains "new" or increased amounts of hitherto unidentified toxic contaminants that adversely affect the immune systems. Bergman *et al.* (1999) noted that PBDEs and bis (4-chlorophenyl) are contaminants that were unknown until more recently in the Baltic, but that the toxic effects of these on the ecosystem is currently limited.

5.2.1.2 ***Effects on the Immune System***

In 1988, an estimated 18 000 harbour seals and several hundred grey seals died around the coasts of Denmark, Sweden, The Netherlands, Norway, United Kingdom and Ireland. In the Baltic, 60% of the harbour seal population died. The primary cause of this mass mortality, or epizootic, was a moribillivirus called phocine distemper virus (PDV), similar to that which causes distemper in dogs (see Kennedy 1996). There is some evidence that the concentration of organochlorine contaminants in the seal's tissues may have influenced the ability of the animals to mount an effective immune response to this disease (Simmonds & Mayer 1997). For instance, some research showed that levels of organochlorines in the blubber of the animals that died were higher than those in animals that survived (Hall *et al.* 1992). Around Britain, seals living in less contaminated areas had lower mortality rates than those in more polluted waters (Simmonds *et al.* 1993). Other studies of organochlorine contaminants found in seals from UK waters (Law *et al.* 1989, Mitchell & Kennedy 1992), have also lead to the conclusion that organochlorines may have exacerbated the effects of the virus that caused the epizootic, and the incidence of secondary infections which were often the final cause of death in affected animals (Heide-Jorgensen *et al.* 1992, Munro *et al.* 1992).

It should be noted that the role of organochlorine contaminants in the mass mortalities remains unproven . For instance, research in the Baltic did not find higher levels of organochlorines in seals infected with the virus compared to unaffected seals (Olsson *et al.* 1994). At the same time, however, experimental studies on seals have shown that pollution may have played a role (de Swart *et al.* 1994, de Swart *et al.* 1996, Ross *et al.* 1995). These studies also demonstrated that marine mammals that inhabit many more polluted coastal environments of Europe may have an increased susceptibility to infections. The studies involved feeding herring from the highly polluted Baltic Sea or from the relatively unpolluted Atlantic Ocean to two groups of harbour seals (*Phoca vitulina*) for two and a half years. Results showed that certain cells of the immune system were reduced in animals fed the more contaminated fish. This indicated that impaired immunological function could result from long-term exposure to pollutants in fish. In particular, the part of the immune system that deals with defence against viral infection was affected.

Experimental studies on harbour seals have also shown that levels of retinol (vitamin A) and thyroid hormones in blood, were reduced in seals that were fed the more highly PCB-contaminated fish from the Baltic Sea as opposed to fish from the North Atlantic (Brouwer *et al.* 1989). Vitamin A deficiency can lead to alterations in immunity, and thyroid hormones are important in development and growth, including neurological development (Jenssen 1996). Effects of reduced levels of thyroid hormones and retinol in seals could lead to an increased susceptibility to microbial infections, reproductive disorders and other pathological alterations. Reduced retinol and thyroid hormone levels have been implicated in reproductive disorders and viral infections of seals and other marine mammals in the Baltic, North Sea and Wadden Sea (Brouwer *et al.* 1989).

5.2.2 **Harbour Porpoise**

As discussed above (section 5.1.2), levels of some persistent organochlorines in harbour porpoises from the Baltic Sea are in the same range as levels at which adverse impacts on immune and nervous systems have been estimated to occur in seals. However, it appears that research on the health status of Baltic porpoises is quite limited.

Siebert *et al.* (2001) performed post mortem examinations on 445 porpoises collected from the Baltic and the North Seas between 1991 and 1996, along with more detailed medical tests on 133 of these animals. The study noted that most lesions found in the animals were caused by parasites. A high proportion (46%) of the porpoises that had been found stranded in this study had died from pneumonia. Investigations showed that the main cause of pneumonia was secondary bacterial infection and not parasitic infection alone. Tests on the immune system, however, did not find any indication of a bacterial or viral epidemic among the porpoises in the period 1991 to 1996. The study included tests for moribillivirus infection, a type of virus which has caused mass mortalities of seals, possibly exacerbated by high tissue levels of organochlorines, and which can also affect dolphins and porpoises (see Section 5.2). Similarly, another study showed that moribillivirus infection was not the cause of death in 74 porpoises collected from the Baltic and North Seas between 1991 and 1997 (Müller *et al.* 2000). Both studies, however, found moribillivirus antibodies in many of the porpoises, indicating the spread of the virus through the Baltic and North Seas populations. Persistent organochlorines have previously been linked to adverse impacts on the immune system of marine mammals. The above study on Baltic porpoises by Siebert *et al.* (2001) suggested that high tissue concentrations of organochlorines and heavy metals in Baltic porpoises could have played some role in the high prevalence of severe parasitic and bacterial infection identified in these animals, although the mechanisms remain to be elucidated.

6 TISSUE LEVELS OF POPS IN THE HUMAN POPULATION OF BALTIC COUNTRIES

As a consequence of the persistent, lipophilic, bioaccumulative properties of many POPS, and tendency of some to biomagnify within food chains, long-term exposure to relatively small concentrations of these compounds leads to the accumulation of considerable deposits in animal and human tissues. Levels of POPs in human tissue have been most commonly assessed in blood, adipose tissue or breast milk. Persistent organochlorines, namely PCDD/Fs, PCBs and several organochlorine pesticides have been found in human tissues on a global scale. In addition to these chemicals, other POPs, such as brominated flame retardants (e.g. PBDE's) and nitro musks have also been found to contaminate human tissues. The contamination of human tissues with POPs may, therefore, be related not only to local uses but also to long distance transport of POPs and contamination of the food chain.

6.1 PCDD/Fs

Many studies have shown that levels of PCDD/Fs in human tissues are generally higher in more industrialised countries and lower in less industrialised countries. This is due to the release of PCDD/Fs as by-products of many industrial processes, notably those involving the production, use or disposal of organochlorines. Regional differences within countries may also arise for the same reason so that, for example, tissue levels may be higher in urban areas than rural areas. Differences between regions may also occur due to dietary habits. For instance, a high intake of fatty fish from the Baltic Sea among Swedish east coast fishermen has resulted in high levels of PCDD/Fs in their tissues due to contamination of the fish (see section 7.1).

To date, the most comprehensive set of studies on the levels of PCDD/Fs and PCBs in human tissue in a number of different countries was performed by the World Health Organisation (WHO) on breast milk samples in the late 1980s and early 1990s (WHO 1996). The study included several Baltic countries namely Denmark, Finland, Germany, Lithuania and the Russian Federation. Results of PCDD/F levels reported by this study are presented in Table 6-1. It should be noted that the results were generated in specific areas of each country which may not necessarily be representative of areas adjacent to the Baltic Sea.

Table 6-1: Mean Levels of PCDD/Fs and dioxin-like PCBs (pg TEQ/g fat, or ppt) in human milk for Baltic countries in 1987/88 and 1992/3, published by WHO (1996)

Country	Area	n (1987/88)	ppt TEQ [PCDD/F] 1987/88	n (1992/93)	ppt TEQ [PCDD/F] 1992/93	ppt TEQ [dioxin-like PCBs] 1992/93
Denmark	7 different cities	42	17.8	48	15.2	4.5
Finland	Helsinki	38	18.0	10	21.5	4.6
	Kuopio	31	15.5	24	12.0	2.4
Germany	Berlin	40	32.0	10	16.5	11.7
Lithuania	Palanga (coastal)			12	16.6	20.4
Russian Federation	Arkhankeisk			1	15.2	8.6

Table 6-2: Mean Levels of PCDD/Fs (pg TEQ/g fat, or ppt) in Breast Milk in Various Baltic Countries

Country	Area	Year samples taken / number (n) of samples	ppt TEQ [PCDD/Fs]	Reference
Estonia	Tarto and Tallin	1991 n = 12	17.5 (Nordic)	Mussalo-Rauhamaa & Lindstrom 1995
Sweden	Sundsvall, Umea, Goteborg, and Borlange	1987 n = 40	22.4 (Nordic)	Mussalo-Rauhamaa & Lindstrom 1995
Russia	Murmansk	1993 n = 30	15.8	Polder *et al.* 1996

The WHO (1996) study recorded human milk levels of PCDD/Fs in 17 different countries. The highest PCDD/F levels were in the range of 20 to 30 pg TEQ/g fat (ppt). These levels were apparent in a few industrialised countries, including Finland (Helsinki). In most countries, including Denmark, Germany and Lithuania, levels were within the range of 10 to 20 ppt TEQ. Lower levels were (4-10 ppt TEQ) were found in a few countries including less industrialised regions of the Russian Federation.

Studies other that those undertaken by WHO (1996) have been published on levels of PCDD/Fs in breast milk of women from Baltic countries (see table 6-2). Levels of PCDD/Fs in breast milk from Estonia and Sweden are reported in Nordic TEQ which is similar but not directly comparable to the I-TEQ system used in the WHO (1996) study. In Murmansk, located on the Kola Peninsula in northern Russia, levels of PCDD/Fs in human milk were within the range of 10-20 ppt TEQ as reported for the majority of countries by WHO (1996) (Polder *et al.* 1996). This study noted that levels of PCDD/Fs in Murmansk were higher than levels found in rural regions of Russia but are similar to levels found in Lithuania.

In addition to PCDD/Fs polybrominated dibenzo-*p*-dioxins and dibenzofurans have also been detected in human milk. Wiberg *et al.* (1992) reported low concentrations (< 20 ppt fat) of PBDD/Fs in the breast milk of Swedish women. These compounds are of particular concern because they add to the body burden of dioxin-like compounds.

6.1.1 **Time Trends**

Laboratory methods for analysing PCDD/Fs and PCBs in breast milk have improved considerably during the 1990s. Results of the second (1992/3) round of the WHO study were consequently more accurate than those of the first round (1987/8) due to this analytical improvement, as well as due to an improved study design. In the first round of the study, the method used dictated that only general comparisons could be made between PCDD/F and PCB concentrations in different countries. Although there were differences in the accuracy of data between the first and second round of the WHO study, results from both rounds have been compared by WHO (1996) in an attempt to identify any trends in the levels of PCDD/Fs that may be occurring over time.

For the 11 countries studied in both the first and second round, which included some Scandinavian and European countries and Canada, it was concluded that levels of PCDD/Fs were not increasing with time. In some countries, the levels had decreased between 1987/88 and 1992/3, and in a few cases dramatic decreases of up to 50%

were reported. For example, results for Germany (see table 6-1) showed that average levels decreased from 32 to 16.5 ppt TEQ. However, in some countries decreases were smaller or not evident. For instance, the average decrease in combined rural (Kuopio) and urban (Helsinki) regions in Finland was 28% (Kiviranta *et al.* 1998), while in Denmark no significant decline was observed.

Other studies have also shown a decline in PCDD/F levels in breast milk in Germany in recent years and in Sweden. In Germany, Päpke (1999) reported a decrease of 50% to 70% in levels of PCDD/Fs in human milk between the late 1980s and 1999. Similarly, Vieth *et al.* (2000) reported a decrease in average breast milk levels of 60% between the late 1980s and 1998. In Sweden, a downward trend of about 50% in the levels of PCDD/Fs was observed between 1986 and 1994 for breast milk samples taken in Uppsala and Sundsvall. The authors noted, however, that only a low number of samples were analysed in this study (10 milk samples) and consequently no far reaching conclusions can be drawn.

In summary, substantial declines in levels if PCDD/Fs in breast milk have been reported in a number of countries including Germany and Sweden, and to a lesser extent in Finland. However, a downward trend has not been found in all European countries studied to date (e.g. WHO 1996). It is therefore not possible to predict trends in Baltic countries for which insufficient data are available. The generally low rate of decline of PCDD/Fs and PCBs (see section 6.2) indicates that a prolonged exposure to these compounds may also be of concern in the coming decades (Norén *et al.* 1999).

Moreover, even in countries where decreased levels of PCDD/Fs in breast milk have been reported, researchers have stressed that the exposure of babies during breast-feeding is still a matter for concern (Fürst 2000, Päpke 1999). The PCDD/Fs intake via human milk for a fully breast fed infant in most industrialised countries exceeds the intake of adults by 1 to 2 orders of magnitude (10 – 100 times). This high exposure of babies during the breast feeding period is of concern given the results of several studies which show that subtle adverse health impacts in babies are associated with prenatal exposure to PCDD/Fs. Both Fürst (2000) and Päpke (1999) commented that the concern regarding exposure of babies via breast milk justifies taking further measures to reduce dioxin emissions into the environment.

6.2 **PCBs**

In addition to monitoring PCDD/Fs, the WHO study undertaken in 1992-3 (WHO 1996) also measured dioxin-like PCBs (non-ortho and mono-ortho PCBs) and several indicator PCBs. The study showed that levels of these compounds did not correspond to the ranking of high to low levels of PCDD/Fs which had been found for the different countries. Thus, countries which had displayed high levels PCDD/Fs in human breast milk did not necessarily also have comparably high levels of PCBs.

Most countries and regions were found to have similar levels of PCBs, and only a few had significantly higher or lower levels. The majority of samples had levels of dioxin-like PCBs below 15 ppt TEQ. Table 6-1 shows levels (as TEQ) of dioxin-like PCBs reported for Baltic countries by the WHO (1996) study. Within the Baltic region, Denmark, Finland, Germany and the Russian Federation had levels less than 15 ppt

TEQ, similar to results from the majority of countries. An exception is Lithuania for which higher levels of dioxin-like PCBs were found (mean 20.4 ppt TEQ).

PCBs may contribute substantially to the total dioxin TEQ, that is the sum of PCDD/Fs and dioxin-like PCBs. The WHO study found that the contribution made by PCBs to total dioxin TEQ varies considerably from country to country. For example, in Belgium, the contribution from PCBs is very low in comparison with PCDD/Fs, whereas in Tromso (Norway) the TEQ for PCBs is twice that of the PCDD/Fs. For the Baltic countries, the study showed the contribution of PCBs to the total dioxin TEQ was particularly high for Lithuania (see table 6-1). Another study in Sweden on persistent organochlorine levels in breast milk of women from Uppsala and Sundsvall in 1994, reported that PCBs contributed over 70% of the total TEQ (Atuma *et al.* 1998b).

6.2.1 **Time Trends**

Research has shown a downward trend in tissue levels of PCBs in several countries over the past two to three decades although the decline is slow and, in some instances, PCB levels appear to have stabilised over prolonged time periods. The decline of PCB levels has been attributed to the cessation of use of PCBs. However, the slowness of the decline, and the periodic stabilisation of levels, is indicative of the persistent nature of these compounds, coupled with their continued leakage into the environment such as from refuse sites. PCBs may now make a higher contribution to the total levels of persistent organochlorines in human tissues in the Baltic than previously due to their slower rate of decline in comparison to DDT compounds. For example, a study of human milk levels in Sweden revealed that in 1972 p,p'-DDE was the predominant compound, constituting 57% of the total organohalogen compounds per gram of milk fat, whereas in 1997, it constituted 27% of the total and PCBs comprised the major part (67%) (Norén & Meironyté 2000).

A study of Swedish blood samples which had been stored frozen for several years reported that PCB levels declined between 1972 and 1980, but did not decline between 1985 and 1989 (Norén 1993). A more recent study by the same author summarised results of investigations undertaken at different times between 1967 and 1997 on breast milk organochlorine levels of women residing in the Stockholm region (Norén & Meironyté 2000). This study showed that the overall decline in the level of total PCBs between 1972 and 1997 was 70%. In the last 10 years or so the data showed a clear decline illustrated in Figure 6.1. Levels fell from 510 ppb fat in 1990 to 324 ppm fat in 1997. Another study reported a decrease in the level of dioxin-like PCBs in breast milk from Uppsala and Sundsvall between 1986 and 1994, although the number of samples in the study was low and prevents firm conclusions from being drawn (Atuma *et al.* 1998b). Similarly, an investigation undertaken as

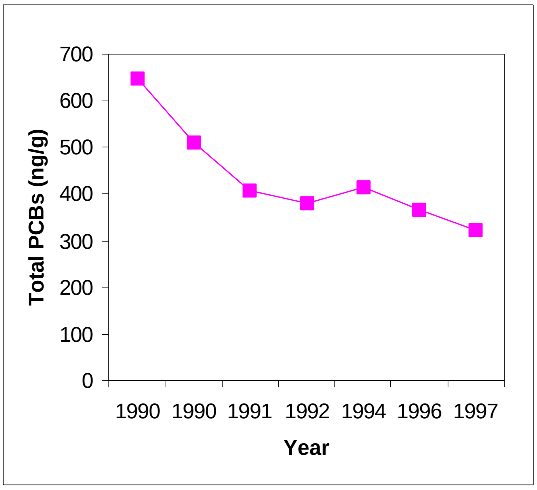


Figure 7.1: PCB levels in the breast milk of Stockholm women (data from Norén & Merionyté, 2000)

part of the WHO study on organochlorine levels in breast milk reported a 53% decrease in dioxin-like PCBs in Finland between 1987 and 1994 (Kiviranta *et al.* 1998).

A study on breast milk in Germany reported that for PCB congeners 138, 153 and 180, levels appeared to remain constant from 1984 to 1989, although in the following two years results indicated a slight decline (Fürst *et al.* 1994). Another study undertaken in northern Germany analysed more than 3500 breast milk samples from women between 1986 and 1997 (Schade & Heinzow 1998). Levels were variable between 1986 and 1991 but a continuous decline was observed each year between 1991 and 1997. Overall, the median level of total PCBs fell from 1280 ppb fat in 1986 to 470 ppb fat in 1997, representing a 60% decrease. Research undertaken at Middle Hesse in Germany investigated levels of PCBs in breast milk in 1984/5, 1990/1 and 1995 (Failing *et al.* 1999). The authors reported significant decreases in high-chlorinated PCB congeners (no. 101, 138, 153 and 180), by about 50% between 1984/5 and 1995. On the other hand, for the low-chlorinated congeners, an increase of PCB number 28 and no change in the levels of congener numbers 31, 49 and 52 were reported. The study concluded that contamination of human milk by PCBs had been substantially reduced in recent years reflecting restrictive measures taken in Germany.

A study undertaken in Poland reported very similar PCB levels in human adipose tissue from Skierniewice in 1979 (1200 ± 400 ppb fat for total PCBs), and from Gda sk, on the southern coast of the Baltic, in 1990 (1500 ± 1300 ppb fat of total PCBs) (Falandysz *et al.* 1994c).

6.3 **Organochlorine Pesticides**

DDT has been banned in most countries for agricultural use although available evidence indicates that it is still used for vector control programs in some areas of the world. Tissue levels of DDT and its metabolites have decreased in countries where it has been banned. Nevertheless its breakdown product DDE is still a widespread contaminant of human tissues. For instance, studies have reported that DDE contaminates nearly 100% of breast milk samples tested from many countries (see Allsopp *et al.* 1998). In Baltic countries, levels of DDT compounds in human milk were in a similar range for Stockholm in Sweden (Norén & Meironyté 2000), Denmark (Hilbert *et al.* 1996) and northern Germany (Schade & Heinzow 1998), but were notably higher for Murmansk in north-west Russia (Polder *et al.* 1996). For instance, levels of p,p-DDE were 129 ng/g (ppb) fat in Sweden in 1997, about 200 ppb fat in Denmark in 1993, but were 1269 ppb fat in Murmansk, Russia in 1993.

HCB was previously used as a fungicide to treat seed grain, but is currently still produced as an unwanted by-product or impurity in the manufacture of some chlorinated compounds and as a by-product from municipal waste incineration. Research has shown that HCB is a widespread contaminant of human milk, being present in most samples of milk that have been tested from many countries (see Allsopp *et al.* 1998). High levels of HCB in breast milk have been reported in east European countries namely the Czech Republic (639 ppb fat) (Schoula *et al.* 1996) and Slovakia (829 ppb fat) (Kocan *et al.* 1995). In the Baltic, relatively low levels have been reported for Sweden (37 ppb fat) (Vaz *et al.* 1993) and Denmark (<40 ppb fat). In north-west Russia, moderate levels of HCB were reported in human milk from the Archangels district (<200 ppb fat) (Polder *et al.* 1998) and in Murmansk (129 ppb fat) (Polder *et al.* 1996).

Technical grade HCH is a mixture of different isomers including α-HCH, β-HCH and γ-HCH. Its use is banned or severely restricted in most countries. However, the pesticide lindane (γ-HCH) is still in use in some countries, although recently (July 2000) a unanimous vote was taken by the EC standing committee on plant health to ban its use for agricultural purposes in Europe by excluding it from Annex 1 of the Council Directive 91/414/EE. The ban will be implemented once the decision is formally approved by the European Commission.

In human tissues, α-HCH and γ-HCH are cleared more rapidly from the body than β-HCH. As a consequence of this, and because the β-isomer is the most persistent, β-HCH is the more widespread isomer in human tissues and is usually present at the highest levels. High levels of β-HCH (222 ppb fat) have been reported in human milk from Nikel, northern Russia, at concentrations about 25 times greater than those found in Arctic Norway (8.1 ppb fat) and Sweden (9.2 ppb fat) (Gilman *et al.* 1997). This suggested either significant uses of HCH in north-west Russia or significant amounts in the food supply. Polder *et al.* (1996) also reported high total HCH levels for Murmansk in north-west Russia (858 ppb fat), about 99% of which was the β-HCH isomer. Comparatively low levels of β-HCH have been reported in Germany (40 ppb fat) (Schade & Heinzow 1998), Sweden (20 ppb fat) (Vaz *et al.* 1993) and Denmark (< 40 ppb fat) (Hilbert *et al.* 1996).

Dieldrin, aldrin and endrin are organochlorine pesticides which have been banned or severely restricted in most countries. Despite restrictions, these chemicals may still be found in human tissues due to their persistent nature. Levels of these pesticides in human tissues are less commonly reported than those of DDT, HCB and HCHs and consequently data are limited.

Table 6-3: Mean Levels of dieldrin in human milk in various Baltic countries

Country	Year	Mean Level (ppb)	Reference
Sweden	1984/5	10	Norén & Meironyté 2000
Denmark	1993	<20	Hilbert *et al.* 1996
Germany	1990/1	9	Alder *et al.* 1994
Russia (5 different areas)	1988/9	2-3	Schecter *et al.* 1990

Table 6-3 shows mean levels of dieldrin in human milk from various Baltic countries. These levels are at the lower end of the global range (Allsopp *et al.* 1998). With regard to aldrin, a study of maternal blood levels of POPs in Arctic countries did not report elevated levels of this chemical in Sweden or Russia (Gilman *et al.* 1997).

Chlordane was previously mainly used as a crop insecticide and for the control of termites. Comparison of chlordane levels in human tissues between countries is difficult because there are several chlordane isomers, a varying number of which are reported by different studies. Chlordane was detectable in human milk from Murmansk in north-west Russia in 1993 (Polder *et al.* 1996) and a study in the Archangels district of northern Russia reported moderate levels of chlordane that were comparable to levels in Norway (Polder *et al.* 1998). A study of maternal blood levels of POPs in circumpolar regions reported that levels of some chlordane metabolites in Canada and Greenland were greater than levels in Arctic Sweden and Norway (Gilman *et al.* 1997).

6.3.1 Time Trends

A downward trend in levels of organochlorine pesticides has been reported for many countries following bans and restrictions on their use. Levels of DDT, DDE, HCB, β-HCH, and dieldrin have all significantly declined in human milk in several Baltic countries. An exception is chlordane for which levels appear more stable.

A decline in the levels if DDT and its metabolites has been recorded since the 1970s for some countries. In Sweden, research in the Stockholm region has recorded decreasing levels of DDT and DDE in human milk from 1972 through until 1997 (Norén & Meironyté 2000). For instance, DDE levels fell from 2420 ppb fat in 1972 to 129 ppb fat in 1997. The authors noted that of all the organochlorine pesticides considered in the study, the most consistent decline was observed for DDT and DDE. The level of DDT in 1997 was only 1% of the level in 1967 and the level of DDE in 1997 was 5% of the level in 1972. In Denmark, levels of DDE in human milk decreased from around 1000 ppb fat in 1982 to around 200 ppb fat in 1993 (Hilbert *et al.* 1996). Similarly, in northern Germany, levels of total DDT (DDT + DDE) dropped from 920 ppb fat in 1986 to 230 ppb fat in 1997 (Schade & Heinzow 1998).

A downward trend in the level of HCB in human milk has been reported to have occurred in Germany, Sweden and Denmark over recent years. For example, levels in northern Germany decreased from 510 ppb fat in 1986 to 60 ppb fat in 1997. In the Stockholm area of Sweden, levels were reported to remain relatively stable between 1972 and 1980 (at around 120 ppb fat, but then gradually declined throughout the 1980s and 1990s to reach a levels of 12 ppb fat in 1997 (Norén & Meironyté 2000). The average level of HCB in 1997 was 5% of that in 1974. In Denmark, levels fell from about 130 ppb fat in 1982 to between 30 and 40 ppb fat in 1993 (Hilbert *et al.* 1996).

Decreases in the levels of β-HCH have also been reported to occur in Germany, Sweden and Denmark. In northern Germany, a study showed that β-HCH decreased very slowly but continuously between 1986 (190 ppb fat) and 1997 (30 ppb fat) (Schade & Heinzow 1998). In Sweden, levels were fairly steady between 1974 and 1980 (at around 100 to 120 ppb fat) but declined to 72 ppb fat by 1984/5 (Norén & Meironyté 2000). Vaz *et al.* (1993) also reported a decline human milk levels of β-HCH in Sweden between 1981, 1986 and 1990. In Denmark, levels in 1982 were around 130 ppb fat and decreased to reach a level of about 35 ppb fat by 1993 (Hilbert *et al.* 1996).

Few data are available on the time trends of other persistent organochlorine pesticides in Baltic countries. In Sweden, a gradual decline in dieldrin levels was recorded in the Stockholm region between 1972 and (49 ppb fat) and 1984/5 (10 ppb fat) (Norén & Meironyté 2000). The average concentration of dieldrin in 1985 was 13% of that in 1967. However, levels of chlordane isomers (oxychlordane and trans-nonachlor) appeared to remain relatively stable between 1976 and 1988/9. Similarly, a review of chlordane levels in adipose tissue in the US indicated that there was no decline in levels between 1974 and 1978 (Dearth & Hites 1991). However, insufficient data are available to determine trends in these compounds in more recent years.

6.4 PBDEs and PCDEs

Recent studies in Germany and Sweden are of particular concern since they show that human tissue levels of PBDEs are increasing over time. Norén & Meironyté (2000) reported that the concentration of PBDE's in human milk from the Stockholm region has increased exponentially over the period 1972 to 1997, showing a doubling of levels every 5 years. Levels in 1972 were 0.07 ng/g lipid or ppb (sum PCDEs) and increased to 4.01 ng/g lipid (ppb) in 1997. The amount of PBDEs accounted for about 1% of the total of organohalogen contaminants present in breast milk. The authors noted that PBDEs are not produced in Sweden but are imported for flame retardant applications and in goods containing flame retardants. It was concluded that the exponential decrease of PBDEs in breast milk is alarming and calls for measures to stop exposure to PBDEs.

Polychlorinated diphenyl ethers (PCDEs) are chemicals that are generated from combustion sources and from chlorophenol preparations. PCDEs have been detected recently in Finnish adipose tissue (Koistinen *et al.* 1995b), German breast milk and Swedish blood samples (Wehler *et al.* 1997).

6.5 Other POPs

Research in Germany by Schröter-Kermani *et al.* (2000) also reported increasing concentrations of PBDEs between 1985 and 1999 in samples of whole blood from Germany. Levels increased from a median of 3.075 to 4.687 ng/g lipid (ppb). The study noted that levels were generally higher than levels previously determined in Sweden.

An organochlorine contaminant, octachlorostyrene was found in the blood of Germans for the first time in a study on individuals residing near to the river Elb (Lommel *et al.* 1992). Previously, this organochlorine had only been reported in individuals working at magnesium and PVC producing plants in Norway. Its presence in the blood of individuals from the general population in Germany was associated with fish consumption from the river Elb, and thus corresponded to high octachlorostyrene levels in the fish. The widespread nature of octachlorostyrene as an environmental contaminant was recently demonstrated after it was detected at low concentrations in polar bear tissue from the Arctic (Sandau *et al.* 2000).

The nitro musk compounds musk xylene and musk ketone have been detected in human milk (Failing *et al.* 1999). A study in Middle Hesse in Germany reported that these compounds were present in most samples of breast milk samples that were tested in 1995 confirming the widespread nature of these environmental pollutants. Musk xylene was measured at a mean concentration of 41 μg/kg fat (ppb) and musk ketone at 10 μg/kg fat (ppb).

6.6 Fish Consumption and Exposure to POPs

The contamination of the Baltic Sea has led to higher levels of persistent organochlorines in fish from this region. These chemicals still persist in fish at higher levels than in other waters. For instance, the pollution load of several persistent organochlorines in herring from the Baltic Sea is over two-times higher than levels found in herring from the Swedish west coast (Atuma *et al.* 1996). Research has shown that consumption of contaminated Baltic Sea fish is associated with an increase of persistent organochlorines in human tissues.

Research in Sweden has shown that intake of fish from the Baltic Sea among the general population is a major determinant of blood (plasma) levels of some persistent organochlorines (Asplund *et al.* 1994). This study investigated levels of PCBs, DDT, DDE and PCCD/Fs in 37 individuals from south-east Sweden who were designated to have either a high, moderate or no intake of Baltic Sea fish. The research showed that levels of several PCB congeners (non- and mono-*ortho*), DDT and DDE in high fish consumers were significantly greater than levels in non-consumers and also greater than moderate consumers. This is illustrated by table 6-1 in which blood levels of DDT and DDE in high, moderate and non-fish consumers are given.

Table 6-4 shows levels of dioxin-like PCBs and PCDD/Fs expressed as TEQs. Again, for both PCDD/Fs and dioxin-like PCBs, the blood levels of high fish consumers were clearly higher than levels in non-consumers. The PCB contribution to the total TEQ was substantial, almost 80%, whereas that from PCDD/Fs was only 20%. The main conclusion from the study was that fish from the Baltic Sea is a major source of exposure to these compounds in the Swedish general population. A study undertaken in Finland which investigated dietary intake of Baltic Sea fish and blood levels of PCDD/Fs, also noted that in Finland, up to 80% of exposure to these chemicals in adults is via fish and fish products (Kiviranta *et al.* 2000).

Table 6-4: Mean blood levels of p,p'- DDT and p,p'- DDE in Three Groups of Men with Different Fish Consumption

Fish Consumption	None (n = 8)	Moderate (n = 7)	High (n = 11)
p,p'- DDE (ng/g lipid)	290 - 1,100	300 - 1900	1300 - 14,000
p,p'- DDT (ng/g lipid)	9 - 48	19 - 63	76 - 300

Source: Asplund *et al.* 1994.

Table 6-5: Mean Blood Levels of PCDD/Fs and 10 PCB Congeners Expressed as TCDD Equivalents (TEQ) in Two Groups of Men with Different Fish Consumption

Fish Consumption	No Fish intake (n = 9)	High Fish Intake (n = 9)
PCB (N-TEQ)	77 (35 – 136)	234 (96-356)
PCDD/F (N-TEQ)	18 (11 – 33)	77 (35 – 136)

Notes: N-TEQ = TEQ, according to Nordic Expert groups. Source: Asplund *et al.* 1994.

7 TISSUE LEVELS OF POPS IN HUMANS AND HEALTH IMPACTS IN HIGHLY EXPOSED GROUPS.

A study by Asplund *et al.* (1994) (see above section 6.6) indicated that fish from the Baltic Sea is a major source of exposure to PCBs, DDT and DDE in the Swedish population. For instance, a high dietary intake of fatty fish from the Baltic Sea led to elevated blood levels of these persistent organochlorines in Swedes. Subsequent research on fishermen and their families from the southeast coast of Sweden, and on fishermen from the Gulf of Finland and Latvia also showed that high fish consumption in these people results in elevated levels of persistent organochlorines in their body tissues (see section 7.1 below). Concern regarding the health of the Swedish fishermen and their families from an increased exposure to persistent organochlorines has led to research into potential health impacts (see section 7.2 below).

7.1 Fish Consumption and Tissue levels

Research has linked high Baltic Sea fish consumption with increased body tissue levels of persistent organochlorines (Sjödin *et al.* 2000, Svensson *et al.* 1995b). Svensson *et al.* (1995b) studied fish consumption and blood levels of organochlorines in groups of 250 fishermen from the Swedish east coast, 250 fishermen from the west coast and 250 individuals from the general population. Results showed that, in general, fishermen ate almost twice as much fish as individuals from the general population in these regions. Among the fishermen, the groups from the east coast and west coast ate the same amount of fish, but fishermen from the east coast ate proportionately more fatty fish. In theory, this would lead to a higher intake of organochlorines by east coast fishermen. This is because fatty fish tends to be more highly contaminated with persistent organochlorines than lean fish. In addition, fish from the Baltic eaten by the east coast fishermen is more highly contaminated with persistent organochlorines than fish from the west coast of Sweden.

Table 7-1: Comparisons of PCDD/F and PCB blood concentrations between fishermen from the east and west coasts of Sweden

	Contaminant	
Location	**PCDD/Fs (ppt TEQ fat - Nordic)**	**PCBs (ppb fat)**
East coast (Sea of Bothnia)	154	2200
East coast (Baltic Proper)	80	1696
East coast (Baltic South)	131	3067
West coast	42	1336

The study reported that blood levels of persistent organochlorines were consistent with east coast fishermen eating more highly contaminated fish. This is illustrated in table 7-1. For instance, the east coast fishermen had higher blood (plasma) levels of PCDD/Fs and PCBs, expressed as TEQ, than the west coast fishermen. The latter had blood levels of these substances of the same magnitude as the general population from these regions. Levels of PCDD/Fs in a pooled blood samples from west coast fishermen averaged 42 ppt TEQ (Nordic) fat, whereas from the east coast fishermen averaged 154 ppt TEQ (Nordic) fat in the Sea of Bothnia region, 80 ppt TEQ (Nordic) fat in the Baltic Proper region and 131 ppt TEQ (Nordic) fat in the Baltic South. All the east coast fishermen had two-times or greater blood levels of PCDD/Fs compared to the general population in the region. Levels of the most toxic PCDD/F congeners (TCDD and 2,3,4,7,8-pentachlorodibenzo-furan) were two to six times higher in pooled blood samples from the east coast fishermen compared to the local general population.

Levels of different PCB congeners were also higher in east coast fishermen than west coast fishermen (see table 7-1). For total PCBs, the west coast fishermen had an average blood level of 1336 ng/g (ppb) fat whereas the east coast fishermen had levels of 2200 ppb fat in the Sea of Bothnia region, 1696 ppb fat in the Baltic Proper and 3067 ppb fat in the Baltic South. Considering both dioxin-like PCBs and PCDD/Fs together, the total TEQ for the east coast fishermen was about two-times higher than the west coast fishermen and the general population from the regions. The main conclusions from the study were that the dietary intake of fatty fish from the Baltic Sea was almost two-times higher among east coast fishermen and that this was reflected in corresponding increases in blood levels of persistent organochlorines.

A study by Sjödin *et al.* (2000) investigated the impact of fish consumption from the Baltic Sea on blood (plasma) levels of Latvian and Swedish men. A groups of 43 men from south-east Sweden and 67 Latvian men were selected for study. Within each group of men the amount of fish they ate was categorised as a high, moderate or low fish consumption. The high fish consumers were mainly professional fishermen from both countries, those in Latvia coming from villages around the Gulf of Riga. Results showed that the blood concentrations of PCBs, DDE, DDT and HCB were all statistically correlated to estimated fish consumption in the men, such that the highest levels were evident in high fish consumers with progressively lower levels in moderate and low fish consumers. For example, analysis showed that the total PCB level increased by about 7% with each additional fish meal per month. The study also showed that levels of persistent organochlorines in high fish consumers increased with increasing age, but this relationship was not true for lower fish consumers. Thus ageing in conjunction with high fish consumption led to increasing blood levels of persistent organochlorines.

The study also investigated blood levels of a more recent environmental contaminant, the polybrominated diphenyl ether (PBDE) BDE-47. Again, blood levels were significantly correlated with fish consumption. The concentration of BDE-47 in the blood of all individuals analysed ranged from <0.1 to 11 ng/g (ppb) fat. This is low in comparison to PCBs, for instance PCB-153 (22-2,300 ppb fat). Nevertheless, BDE-47 showed the most pronounced relationship to fish consumption out of all the POPs studied. For instance, the blood level of BDE-47 increased by an estimated 13% with each additional fish meal per month. This indicated that consumption of fish is a major route of exposure among high fish consumers. The authors noted that the strong link between fish consumption of levels of BDE-47 in blood indicated a need for further research on dietary exposure to PBDE's in the population of the Baltic Sea region.

Blood levels of pentachlorophenol (PCP) were investigated in the Latvian and Swedish men. Results showed that unlike other persistent organochlorines blood levels of PCP were not related to fish consumption, suggesting that fish is not a major source of exposure to PCP. Levels of PCP were, however, strongly related to country of residence, such that levels were much lower in Latvian men than Swedish men. It was suggested that there must be other sources of exposure to PCP in the environment which remain to be identified. A likely source of exposure is the use of PCP as a wood preservative, a practice that occurred widely in Sweden before 1975.

A study on 47 fishermen from the Gulf of Finland demonstrated a link between high fish consumption from the Baltic Sea and elevated blood levels of PCDD/Fs (Kiviranta *et al.* 2000). The fishermen were compared with a control group of age-matched individuals from southern Finland who had a relatively low fish consumption of less than one fish meal per week. The control group had an average blood level of PCDD/Fs of 33 ppt TEQ fat. This can be considered as a background concentration for men in Finland aged 40-70. By comparison, 26 of the fishermen who were defined as having a high fish intake, consumption of fish at least twice a week, had a much higher average blood level of 180 ppt TEQ fat. The remaining fishermen, who ate fish less than twice a week, also had increased blood levels of PCDD/Fs (110 ppt TEQ fat). The study noted that levels of PCDD/Fs in the blood of fishermen from the Gulf of Finland were elevated to levels that were comparable with those seen in inhabitants of Seveso, Italy, after the accidental release of dioxin (TCDD) in 1976.

Only one study has reported no relationship between fatty fish consumption from the Baltic Sea and tissue levels of persistent organochlorines (Hagmar *et al.* 1998). This research investigated levels of PCBs in women from the Åland and Turku archipelago in Finland. 30 women were selected for the study who were due to give birth at a Finnish hospital in Åland, 20 of whom ate around 5 meals per month of fatty fish from the Baltic Sea and 10 who never ate such fish. The study investigated the levels of PCBs both in whole blood from the women and in blood from the umbilical cord at delivery. No differences were found in the concentrations of PCBs in whole blood or cord blood between the fish and non-fish consumers.

The authors proposed several explanations as to why higher fish consumption did not result in higher blood levels in the women. Firstly, the study had limited statistical power to find a relationship because the number of women in the study was low. Secondly, fish consumption among women in the study was lower than expected. In addition, levels of PCBs decreased in herring and salmon from the Baltic Sea in the 1970s and 1980s, and, as a consequence of this, it is possible that young women in this study had taken in lower amounts of PCBs during their lifetime through fish consumption than older individuals included in the study of the Swedish general population by (Asplund *et al.* 1994).

7.2 Health Impacts of POPS in Highly Exposed Groups.

Persistent organochlorines have been associated with a multitude of adverse health impacts in laboratory animals and in many species of wildlife. Furthermore, in humans, accidental exposure to high concentrations of persistent organochlorines, occupational exposure and exposure of the general population to current background concentrations in several countries, has been associated with a broad range of undesirable health effects. These effects include impacts on the nervous system, immune system and reproductive system and cancer.

Swedish east-coast fishermen and their families consume at least twice as much fatty Baltic Sea fish than the general Swedish population and consequently they have higher levels of persistent organochlorines in their body tissues. There is, therefore, particular concern regarding the possible health impacts of elevated levels of persistent organochlorines in Swedish east-coast fishermen and their families. During the past few years, a series of studies have been undertaken on this group to investigate whether a higher intake of fatty Baltic Sea fish increases the risk of certain health conditions.

Researchers have established groups of individuals (cohorts) of professional fishermen, their wives and their sisters, for the purpose of study. Cohorts of fishermen and members of their families from the Swedish west coast have also been established for comparative purposes. This group of individuals has a similar lifestyle to the east-coast fishermen but consumes fish that is much less contaminated by persistent organochlorines. These studies are discussed in detail below.

7.2.1 Reproduction and Development

7.2.1.1 Growth Retardation

In humans, growth retardation of the foetus in the womb, a condition known as intrauterine growth retardation (IUGR), can manifest as reduced weight and size at birth. Previously, several studies have reported an association between increased exposure to PCBs, or other persistent organochlorines, in women and lowered birth weight in their newborns. For instance, exposure of pregnant women to high levels of dioxins and PCBs during food contamination incidents in Japan (Yusho) and Taiwan (Yucheng) resulted in lowered birth weight in their offspring (see Hsu *et al.* 1994, Masuda 1994). Exposure to lower levels of organochlorines has also been associated with low birth weight in some but not all studies (see Rylander *et al.* 2000). For example, a study of women who had consumed a moderate amount of fish from Lake Michigan, a region where fish was contaminated with high levels of PCBs, gave birth to babies with lower birth weights than women who had not eaten Lake Michigan fish (Fein *et al.* 1984). Moreover, elevated levels of PCBs in umbilical cord blood was linked to lower birth weights as well as smaller head circumference.

In Sweden, studies have also reported an association between exposure to dietary PCBs through fatty fish consumption from the Baltic Sea and reduced birth weight of infants (Rylander *et al.* 1995, Rylander *et al.* 2000). In summary, the studies reported that birth weight was lower among infants born to women who consumed large quantities of fish on the east coast compared to similar women from the west coast, as well as compared to lower fish consumers from the general population. In addition, an increased incidence of low birth weight *per se*, defined as a weight of less than 2500g in one of the studies or less than 3000g in the other, was reported for high fish consumers on the east coast.

Rylander *et al.* (1995) investigated birth weight of 1501 infants born to fishermen's wives on the Swedish east coast between 1973 and 1991, and 3553 infants born to fishermen's wives on the west coast. The study revealed that fishermen's wives from the east coast gave birth to babies with significantly lower birth weights than the general population, whereas the opposite was true for west coast fishermen's wives. Furthermore, results showed that there was a greater number of infants born who had low birth weight (defined as <3000 g) to fishermen's wives from the east coast, compared with fishermen's wives from the west coast. The study concluded that the results supported, but did not prove, an association between a high consumption of contaminated fish from the Baltic Sea and an increased risk for low birth weight.

Further studies by the same researchers gave similar results (see Rylander *et al.* 2000). For instance, an increased risk for low birth weight was found among east coast mothers who had relatively high concentrations of PCB-153 (used as a marker for total PCBs) in their blood. Another study found an increased risk among mothers who had grown up in a fishing village, a parameter that can be interpreted as an indirect measure of a mother's accumulated consumption of fish from the Baltic Sea.

Finally, Rylander *et al.* (2000) conducted a study among fishermen's sisters from the Swedish east coast and west coast. The study assessed birth weight of 1719 infants from the east coast and 2682 infants from the west coast. Results showed that infants born to east coast fishermen's sisters were significantly lower in weight (median birth weight 3500 g) compared to infants born to west coast women (median birth weight 3560 g). After adjustment of the results there was mean birth weight difference of 72 g between the two groups. Moreover, east coast women had a 1.6-fold higher risk of having a baby with low birth weight (defined as < 2500 g). In addition, the head circumference of infants from the east coast were significantly smaller compared to west coast infants. The study concluded that both this study and the previous studies indicate that a high intake of persistent organochlorine contaminated fish from the Baltic Sea may cause intra-uterine growth retardation.

7.2.1.2 ***Fertility***

Recent studies have investigated whether a high dietary intake of fish contaminated with persistent organochlorines can affect human fertility. In rats, exposure to PCBs has been found to reduce the number of successful pregnancies (see Axmon *et al.* 2000a). Studies on humans, both in the Baltic and at Lake Ontario have investigated the length of time it takes to become pregnant. Both found an association between high consumption of fish contaminated with persistent organochlorines and an increase in time to pregnancy (see below).

For example, a study on anglers at Lake Ontario found that increased consumption of fish by women was associated with a greater time to pregnancy (Buck *et al.* 2000). The time to pregnancy was greater for women who had a history of higher fish consumption for 3 to 6 years or who had recent fish consumption of one or more meals per month. The study noted that these results may only be considered as preliminary, but concluded that maternal consumption of contaminated fish may increase time to pregnancy among couples attempting pregnancy (although it was not certain that persistent organochlorines were the cause of the increase).

In the Baltic, a study on fishermen's wives from the Swedish east and west coast assessed their time to first planned pregnancy (Axmon *et al.* 2000b). Results indicated there was a significant increase in time to pregnancy for the east coast women compared with the west coast women, although this was restricted only to women who were heavy smokers. It is notable that smoking itself is known to prolong time to pregnancy and, in this study, it is possible that smoking acted synergistically (in a greater than additive manner) with fish consumption. Although the study found that time to pregnancy was increased in east coast fishermen's wives compared to west coast women, it did not find a direct link with exposure to persistent organochlorines. Exposure was assessed as current fish consumption or as "growing up in a fishing village". However, it is possible that these exposure assessments may have been unsatisfactory. A more precise assessment of exposure would be measurements of blood levels of organochlorines, for example, PCB-153 (a marker for long-term PCB exposure), on an individual basis.

The study on the fishermen's wives by Axmon *et al.* (2000b) also collected information on subfertility and infertility. Women were defined as subfertile if they had not become pregnant, while trying, during a 12 month period (excluding times that eventually led to pregnancy) or, if it had taken them longer to get pregnant than 12 months for their first 5 pregnancies. If subfertile women had no children they were defined as infertile. Results of the study showed there was a slight increased risk of subfertility among east coast women compared to west coast women. The risk was greater for heavy smokers. There was also a statistically significant increased risk (2.49-fold) of infertility among east coast women. The number of infertile women in the east coast (3% of the group) was higher than among west coast women (1% of the group).

The overall conclusions from the study were that the data on time to pregnancy, subfertility and infertility give some support for a negative association between exposure to persistent organochlorines and fertility among heavy smokers. However, more research which entails use of better individual exposure assessments to persistent organochlorines was recommended before firm conclusions can be drawn.

7.2.1.3 ***Miscarriage and stillbirths***

In laboratory animals, exposure to PCBs has been found to increase the risk of miscarriage. In humans a possible link was also reported by a study which found that women who were hospitalised for miscarriages had higher PCB levels than a control group of full-term pregnancy women (see Axmon *et al.* 2000a). However, studies on women who ate contaminated fish from the Great Lakes did not find any relationship between high fish consumption and risk of miscarriage (Axmon *et al.* 2000b).

An investigation on whether the dietary intake fish from the Baltic Sea contaminated with persistent organochlorines had any effect on miscarriage was published recently by Axmon *et al.* (2000b). The study included 795 fishermen's wives from the Swedish east coast and 1,851 fishermen's wives from the west coast. Exposure to persistent organochlorines was assessed for all women using their current consumption of fatty fish, and by concentration of a PCB congener, (CB-153), in blood (plasma) for a small proportion (103) of the women. Results of the study showed there was no increase in miscarriages for east coast women compared to west coast women. In addition, no link was found between the number of miscarriages and exposure to persistent organochlorines as measured by current consumption of Baltic Sea fish or level of CB-153 in blood. The study concluded that the data provided no evidence that dietary exposure to persistent organochlorines from Baltic Sea fish increases miscarriage. The authors (Axmon *et al.* 2000b) (see also Axmon *et al.* 2000a) noted that the results on stillbirth and previous results on lowered birth weight are in line with animal studies which show that a much higher dose of PCBs was needed to induce stillbirths in rhesus monkeys than the level which resulted in low birth weight.

7.2.1.4 ***Congenital Abnormalities***

Exposure of pregnant laboratory animals to PCDD/Fs and PCBs has been shown to cause defects (such as cleft palate) in their offspring at doses that are not toxic to the mother. In humans, pregnant women who were accidentally exposed to high levels of PCDFs and PCBs at food contamination incidents in Japan and Taiwan had babies with structural malformations and organ dysfunction (see Hsu *et al.* 1994, Masuda 1994). In Sweden, a study was undertaken to assess whether a high dietary intake of fatty fish from the Baltic Sea, contaminated with persistent organochlorines, might increase the risk for congenital malformations (Rylander & Hagmar 1999).

The study included 1501 infants born to fishermen's wives from the east coast between 1973-1991 and 3553 infants born to fishermen's wives from the west coast. Results showed that east coast women who had a relatively high consumption of fatty fish from the Baltic Sea, did not have an increased risk of having a malformed infant. For instance, 3.3% of infants born to east coast fishermen's wives had some malformation diagnosis as compared to 5.0% of the west coast infants. A comparison of results with the general Swedish population indicated there were, in total, fewer cases of congenital malformations in the east coast group than expected. In addition, no increase in certain specific malformations was detected in the east coast group.

The authors of the study noted that, on the basis of animal data, it is probable that higher levels of persistent organochlorines are needed before structural malformations to the foetus would occur, which could explain why no abnormalities were detected. In addition, the study had some limitations. The assumption that all women in the east coast group had a high consumption of contaminated Baltic Sea fish, might lead to a misclassification of exposure, and reduce the chance of detecting an association with birth defects. In addition, specific birth defects are rare events and the statistical power of the study may not be great enough (i.e. not enough infants studied) to detect small or moderate increase in incidence of birth defects.

In conclusion, the authors noted, therefore, that the present results reveal no association with consumption of contaminated Baltic Sea fish and the total rate of congenital malformation, though they do not exclude the possibility of slight associations with some specific type of malformation (Rylander & Hagmar 1999).

7.2.2 **Psychometric and Medical Parameters**

Lake Michigan in the USA has been polluted by PCBs and other persistent organochlorines in recent decades and consequently fish from the lake are also contaminated. Research carried out on children born to women who had eaten a moderate amount of PCB-contaminated fish from Lake Michigan indicated that exposure of their children to PCBs while in the womb was associated with subtle adverse effects on intellectual function in the young children. The effects have persisted in children up to the age of 11 (Jacobson & Jacobson 1996).

In Sweden, fishermen's wives and sisters from the southeast coast have been shown to have an average of about 30% higher PCB levels in their blood than women from the general population (see above). This suggested that children who were born to these women had been exposed to higher than average levels of PCBs both prenatally (in the womb) and postnatally (by the transfer via breast milk). Rylander & Hagmar (2000) investigated whether boys born to fishermen's wives and fishermen's sisters on the east coast of Sweden have psychometric impairments by the age of 18.

The study used the results of psychometric tests performed when boys are called up for compulsory military service in Sweden. The psychometric tests were designed to measure general intelligence and the ability to act under pressure. Results of psychometric tests for boys from east coast fishing families were compared to boys of the same age at conscription who were born to fishermen's wives and fishermen's sisters from the Swedish west coast during the same years (1973-1975). The results were also compared with expected values of the psychometric tests based on conscript examination data for the general population in the same geographic areas.

The study showed that there was no significant difference in the psychometric test results between east and west coast boys. However, both groups had a decreased fraction of boys with high intelligence, attributed to possible differences in the level of education of parents or other socio-economic factors. The overall conclusions of the study were that the results do not support any harmful long-term impact of pre- and post-natal exposure to persistent organochlorine compounds from mother's fish consumption on psychometric functions of boys in their conscript examinations.

In addition to an investigation of intelligence, the study by Rylander & Hagmar (2000) also studied the hearing and visual ability in the boys which were tested by medical examination at conscription. Exposure to PCBs during development has caused hearing loss in rats, but this effect has not been reported in humans. In the study on the Swedish boys from fishing families, no difference in hearing ability was found between the east coast group and the west coast group or the general population. With regard to vision, boys on the east coast had lower scores for visual accuracy than west coast boys. However, no difference was apparent between the east coast group and the general population and so any difference with the west coast group was considered to be of little consequence with respect to organochlorine exposure.

7.2.3 Cancer

Some persistent organochlorines, including DDT, PCBs and certain PCDD/Fs, are well known animal carcinogens. Some, such as dioxin (TCDD), are classified as human carcinogens. Consequently, there is a concern regarding possible carcinogenic effects of elevated exposure to these chemicals through a high intake of contaminated fish. Research on fishermen from Sweden has been conducted to investigate the pattern of cancer mortality (death from cancer) and cancer incidence among east and west coast fishermen (Hagmar *et al.* 1992, Svensson *et al.* 1995b). The studies concluded that a high consumption of fatty fish contaminated with persistent organochlorines from the Baltic Sea was associated with a decreased overall mortality from cancer, but an increased incidence of stomach cancer and skin cancer.

Hagmar *et al.* (1992) investigated a group of 1371 east coast fishermen who had been members of a fishing organisation for 12 months or more during 1944 to 1987. Compared with statistical data for the general population of Sweden, the fishermen had a decreased overall mortality from cancer. However, mortality from myeloma was significantly increased and stomach cancer was also increased although not significantly. Incidence of cancer, rather than death from cancer, was also investigated. Unlike cancer mortality, cancer incidence was not lowered among the fishermen. This was mainly due to significant increases in the incidence of squamous cell cancer of the skin and lips, and of stomach cancer.

In a subsequent study, Svensson *et al.* (1995a) extended the observation period of the group of east coast fishermen and included a greater number (2907) of fishermen. A comparison was made with a group of 8493 fishermen from the west coast. The results were compatible with the initial study by Hagmar *et al.* (1992) discussed above. For instance, the study found there was a significantly lowered overall mortality from cancer in both east and west coast fishermen. Results showed a significantly increased mortality from myelomas in east coast fishermen, and significantly increased incidence both of squamous cell cancer of the skin and stomach cancer in east coast fishermen compared to west coast fishermen and to the general population.

In discussion of these results it was observed that both the east coast and west coast fishermen had a lowered rate of cancer compared with the general population. It was noted that other research has suggested that populations consuming large amounts of marine oils have low cancer incidence rates (Hagmar *et al.* 1992). With regard to death from specific cancers, Svensson *et al.* (1995a) found an excess of cancers from multiple myelomas among east coast fishermen, but not west coast fishermen. The authors noted that previous research has suggested increase in multiple myelomas in a dioxin (TCDD)-exposed population in the Seveso area of Italy.

Incidence of lip cancer was higher in groups of both east and west coast fishermen relative to the rest of the population. There was also a higher incidence of squamous cell cancer of the skin in the east coast cohort (SIR 2.28 95% CI 1.45-3.50) relative both to the general population and to the west coast group. A possible cause for the increase in skin cancer among east coast fishermen is occupational exposure to oil and tar, since there is reason to believe that the east coast fishermen had a higher exposure to these compounds than their west coast counterparts. It is also possible that some increase may be due to the 10-15% higher exposure to UV light in the southern part of the Baltic relative to the west coast. However, this difference is too small to explain the observed difference in incidence of skin cancer. A further possibility is that exposure to dioxin (TCDD) and similar compounds through consumption of Baltic Sea fish, could be a contributor to the elevated number of skin cancers among east coast fishermen because these compounds are potent tumour promoters. The blood levels of dioxin-like organochlorines were two-fold greater among east coast compared to west coast fishermen. In many species, epithelial cells (such as the surface of the skin) are the primary targets for the action of TCDD.

The incidence of stomach cancer was greater in east coast fishermen (SIR 1.59 CI 1.03-2.39) but not in west coast fishermen when compared with the general population. Svensson *et al.* (1995a) noted that previous studies have shown that dietary habits, like high salt and smoked food intake, have been associated with an increase in stomach cancer. Fishermen from Canada and Britain have been found to have an elevated risk of mortality from the disease. Svensson *et al.* (1995a) commented that the east coast fishermen consumed about twice as much smoked fish than the west coast fishermen. No comment was made in relation to increased exposure to persistent organochlorines and stomach cancer, thought the possible significance of such exposure as a contributory factor cannot be ruled out.

7.2.3.1 ***Cancer in Women***

Epidemiology research has suggested an association between breast cancer and exposure to various persistent organochlorines such as PCBs and DDTs (Dewailly *et al.* 1993, Savitz 1994, Wolff *et al.* 1993). However, the data are not conclusive because other studies have reported conflicting results which indicate there is no association (Hunter *et al.* 1997, van't Veer *et al.* 1997).

In Sweden, a study has been conducted on cancer in east coast fishermen’s wives who were high consumers of fish from the Baltic Sea (Rylander & Hagmar 1995). The study by found an increase in both mortality and in cancer incidence from breast cancer among these women. It was concluded that the results support, but do not prove, the hypothesis of an association between exposure to a mixture of persistent organochlorine compounds through fish consumption and an increased risk for breast cancer.

Results of the study showed that, for the period 1968-89 for the east coast group, and 1965-89 for the west coast group, the overall mortality did not differ from expected. However, an increased mortality for malignant cancers (SIR 1.23, 95% CI 0.98-1.52) was found for the east coast group compared to the general population. This increase was largely due to elevated mortality from breast cancer (SIR 1.74, 95% CI 1.08-2.72). No such increase was evident in the west coast group.

With regard to cancer incidence, the overall cancer incidence in the east coast group was elevated whereas it was decreased in the west coast group. Similar to cancer mortality, the breast cancer incidence was also higher in the east coast group (1.29, 95% CI 0.96-1.71), but was lower in the west coast group than in the general population. An association was not found between breast cancer and exposure to persistent organochlorines as estimated by a dietary survey among some of the women. Nevertheless, the authors noted that previous research had indicated that blood levels of dioxin-like organochlorines compounds, a more direct indicator of exposure, were at least twice as high for the east coast group compared to the west coast group and to the general population. Rylander & Hagmar (1995) also reported that cervical cancer was increased in the east coast group to a similar extent as breast cancer.

7.2.4 **Immune System**

Dioxin (TCDD) and PCBs have been found to cause toxicity to the immune system in laboratory animals. In humans, accidental exposure to TCDD has been associated with changes in immune system cells such as an increase in numbers of natural killer (NK) cells (see Hagmar *et al.* 1995). In addition, a study on healthy women and their babies from the general population in the Netherlands found that exposure to PCBs in the womb and via breast milk was related to changes in cells of the immune system (Weisglas-Kuperus 1998, Weisglas-Kuperus *et al.* 1995).

In Sweden, research has investigated the impacts of a high intake of fatty fish contaminated with persistent organochlorines from the Baltic Sea on the immune system of adults (Hagmar *et al.* 1995, Svensson *et al.* 1994). One study reported that a high consumption of Baltic Sea fish and increased levels of persistent organochlorines in blood were linked with decreased levels of NK cells (Svensson *et al.* 1994). The study investigated levels of NK cells in 23 men from southeast Sweden who had a high intake of Baltic Sea fish and a control group of 20 men who had virtually no fish intake. Results showed that the weekly intake of fish correlated inversely with levels of NK cells in individuals such that higher fish consumption was associated with lower levels of NK cells. The study also showed that blood levels of certain PCB congeners (126 and 118) and of p,p'-DDT correlated with decreased levels of NK cells. The study concluded that the accumulation of persistent organochlorines in high consumers of fatty Baltic Sea fish may adversely affect NK cell levels.

Another study was undertaken on 68 Latvian individuals who had a high, medium or low fish consumption from the Gulf of Riga (Hagmar *et al.* 1995). The study found that a high intake of fish predicted a change in immune system cells including an increase in B cells and an increase in the ratio of certain immune cells (CD4+/CD8+ ratio). However, previous data from human studies do not support these associations. In addition, the study did not find an effect on NK cells as reported in the previous study on Swedish high fish consumers. The study concluded that there are compounds within fish itself that may effect cells of the immune system in different ways, as well as a variety of different pollutants such as persistent organochlorines that can also impact on immune system cells. It is therefore not possible to separate out the possible different effects of compounds in the fish itself and the different pollutants in order to pinpoint the cause of the observed associations with immune system cells found in this study.

REFERENCES

Addo W, van Pul J, Bidleman TF, Brorstrom-Lunden E, Builtjes JH, Dutchak S, Duyzer JH, Gryning SE, Jones KC, van Dijk HFG & van Jaarsveld JA. 1999. Atmospheric transport and deposition of pesticides: An assessment of current knowledge. Water, Air and Soil Pollution, 115: 245-256.

Aguilar ABA. 1988. Age and sex-related changes in organochlorine compound levels in Fin Whales (Balaenoptea physalus) from the eastern north Atlantic. Mar. Environ. Res., 25: 195-211.

Åkerblom N, Olsson K, Berg AH, Andersson PL, Tysklind M, Förlin L & Norrgren L. 2000. Impact of polychlorinated naphthalenes (PCNs) in juvenile Baltic Salmon *Salmo salar*: Evaluation of estrogenic effects, development and CYP1A induction. Arch Environ Contam Toxicol, 38: 225-233.

Åkerman G, Tjärnlund U, Broman D, Näf C, Westin L & Balk L. 1996. Comparison of reproductive success of cod, *Gadus morhua*, from the Barents Sea and Baltic Sea. Marine Environmental Research, 42:(1-4) 139-144.

Alder L, Beck H, Mathar W & Palavinskas R. 1994. PCDDs, PCDFs, PCBs and other organichlorine compounds in human milk levels and their dynamics in Germany. Organohalogen Compounds, 21: 39-44.

Allsopp M, Stringer R & Johnston P. 1998. Unseen poisons: levels of organochlorine chemicals in human tissues. Exeter, UK. Greenpeace International, pp. 97.

Allsopp M, Costner P & Johnston P. 2001. Incineration and human health. Amsterdam: ISBN: 90-73361-69-9. Greenpeace International, pp. 86.

Andersson O & Wartanian A. 1992. Levels of polychlorinated camphenes (toxaphene), chlordane compounds and polybrominated diphenyl ethers in seals from Swedish waters. Ambio, 21:(8) 550-552.

Asplund L, Svensson B-G, Nilsson A, Eriksson U, Jansson B, Jensen S, Wideqvist U & Skerfving S. 1994. Polychlorinated biphenyls, 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (p,p'-DDT) and 1,1-dichloro-2,2-bis(p-chlorophenyl)-ethylene (p,p'-DDE) in human plasma related to fish consumption. Archives of Environmental Health, 49:(6) 477-486.

Asplund L, Athanasiadou M, Sjödin A, Bergman Å & Börjeson H. 1999. Organohalogen substances in muscle, egg and blood from healthy Baltic Salmon (*Salmo salar*) and Baltic Salmon that produced offspring with the M74 syndrome. Ambio, 28:(1) 67-76.

ATSDR. 1997. Agency for Toxic Substances and Disease Registry, US Public Health Service. Toxicological Profiles. CRC Press Inc.

Atuma SS, Linder C-E, Wickland-Glynn A, Andersson O & Larsson L. 1996. Survey of consumption fish from Swedish waters for chlorinated pesticides and polychlorinated biphenyls. Chemosphere, 33:(5) 791-799.

Atuma SS, Aune M, Bergh A, Wicklund-Glynn A, Darnerud PO, Larsson L, Olsson M & Sandström O. 1998a. Polychlorinated biphenyls in Salmon (*Salmo salar*) from the Swedish east coast. Organohalogen Compounds, 39: 153-156.

Atuma SS, Hansson L, Johnsson H, Slorach S, de Wit CA & Lindstroms G. 1998b. Organochlorine pesticides, polychlorinated biphenyls and dioxins in human milk from Swedish mothers. Food Additives and Contaminants, 15:(2) 142-150.

Axmon A, Rylander L, Strömberg U & Hagmar L. 2000a. Miscarriages and stillbirths in women with a high intake of fish contaminated with persistent organochlorine compounds. Int Arch Occup Environ Health, 73: 204-208.

Axmon A, Rylander L, Strömberg U & Hagmar L. 2000b. Time to pregnancy and infertility among women with a high intake of fish contaminated with persistent organochlorine compounds. Scand J Work Environ Health, 26:(3) 199-206.

Barrie LA, Hartog G, den Bottenheim JW & Landsberger S. 1989. Anthropogenic aerosols and gases in th elower troposphere at Alert Canada in April 1986. J. Atmos. Chem., 9: 101-127.

Becher G, Jensen AJ, Zubchenko A, Haug LS, Hvidsten NA, Johnsen BO & Kashin E. 1998. Dioxins and non-ortho PCBs in Atlantic salmon, *Salmo salar*, from major Norwegian and Russian salmon rivers. Organohalogen Compounds, 39: 427-430.

Belfroid AC, Purperhart M & Ariese F. 2000. Organotin levels in seafood. Marine Pollution Bulletin, 40:(3) 226-232.

Bengtsson B-E, Hill C, Bergman Å, Brandt I, Johansson N, Magnhagen C, Södergren A & Thulin J. 1999. Reproductive disturbances in Baltic fish: a synopsis of the FiRe project. Ambio, 28:(1) 2-8.

Bergek S, Berrgqvist P-A, Hjelt M, Olsson M, Rappe C, Roos A & Zook D. 1992. Concentrations of PCDDs and PCDFs in seals from Swedish waters. Ambio, 21:(8) 553-556.

Berggrena P, Ishaq R, Zebühr Y, Näf C, Bandh C & Broman D. 1999. Patterns and levels of organochlorines (DDTs, PCBs, non-*ortho* PCBs and PCDD/Fs) in male harbour porpoises (*Phocoena phocoena*) from the Baltic Sea, the Kattegat-Skagerrak Seas and the west coast of Norway. Marine Pollut. Bull., 38:(12) 1070-1084.

Bergman A. 1999. Health condition of Baltic grey seal (Halichoerus grypus) during two decades. Apmis, 107: 270-282.

Bergman A, Athanasiadou M, Wehler EK & Sjödin A. 1999. Polybrominated environmental pollutants: human and wildlife exposures. Organohalogen Compounds, 43: 89-92.

Bernes C. 1998. Persistent Organic Pollutants: A Swedish view of an International Problem. Stockholm: Swedish Environmental Protection Agency.

Bidleman TF, Walla MD, Roura R, Carr E & Schmidt S. 1993. Organochlorine pesticides in the atmosphere of the Southern Ocean and Antarctica, January-March, 1990. Mar. Poll. Bull., 26:(5) 258-262.

Bignert A, Litzén K, Odsjö T, Persson W & Reutergårdh L. 1995. Time-related factors influence the concentrations of sDDT, PCBs and shell parameters in eggs of Baltic Guillemot (*Uria aalge*), 1861-1989. Environmental Pollution, 89: 27-36.

Bignert A, Olsson M, Persson W, Jensen S, Zakrisson S, Litzen K, Eriksson U, Haggberg L & Alsberg T. 1998. Temporal trends of organochlorines in Northern Europe, 1967-1995. Relation to global fractionation, leakage from sediments and international measures. Environmental Pollution, 99: 177-198.

Biselli S, Bester K, Huhnerfuss H & Fent K. 2000. Concentrations of the antifouling compound Irgarol 1051 and of organotins in water and sediments of the German North and Baltic Sea marinas. Mar. Poll. Bull., 40:(3) 233-243.

Blomkvist G, Roos A, Jensen S, Bignert A & Olsson M. 1992. Concentrations of sDDT and PCB in seals from Swedish and Scottish waters. Ambio, 21:(8) 539-545.

Börjesson P & Berggren P. 1997. Morphometric comparisons of skulls of harbour porpoises (*Phocoena phocoena*) from the Baltic, Kattegat, and Skagerrak seas. Can J Zool, 75: 280-287.

Brouwer A, Reijnders PJH & Koeman JH. 1989. Polychlorinated biphenyl (PCB)-contaminated fish induces vitamin A and thyroid hormone deficiency in the common seal (Phoca vitulina). Aquatic Toxicol., 15: 99-106.

Buck GM, Vena JE, Schisterman EF, Dmochowski J, Mendola P, Sever LE, Fitzgerald E, Kostyniak P, Griezerstein H & Olson J. 2000. Parental consumption of contaminated sport fish from Lake Ontario and predicted fecundability. Epidemiology, 11:(4) 388-393.
Burreau S, Zebühr Y, Ishaq R & Broman D. 2000. Comparison of biomagnification of PBDEs in food chains from the Baltic Sea and the northern Atlantic Sea. Organohalogen Compounds, 47: 253-255.
Campbell I & McConnell G. 1980. Chlorinated paraffins and the environment. 1. Environmental occurrence. Environ. Sci. Technol., 14:(10) 1210-1214.
Chiu A, Chiu N, Beaubier NT, Beaubier J, Nalesnik R, Singh D, Hill WR, Lau C & Riebow J. 2000. Effects and mechanisms of PCB exotoxicity in food chains. Environ Carcino & Ecotox Revs, C18:(2) 127-152.
Cockcroft V, de Kock A, Lord D & GJB R. 1989. Organochlorines in bottlenose dolphins *Tursiops truncatus* from the east coast of South Africa. S. Afr. J. Mar. Sci., 8:(207-217).
Colborn T, vom Saal FS & Soto AM. 1993. Developmental effects of endocrine-disrupting chemicals in wildlife and humans. Environmental health perspectives, 101:(5) 378-384.
Dannenberger D. 1996. Chlorinated microcontaminants in surface sediments of the Baltic Sea. Investigations in the Belt Sea, the Arkona Sea and the Pomeranian Bight. Mar. Poll. Bull., 32:(11) 772-781.
Dannenberger D, Andersson A & Rappe C. 1997. Levels and patterns of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and bipenyls in surface sediments from the western Baltic Sea (Arkona Basin) and the Oder River estuarine system. Mar. Poll. Bull., 34:(12) 1016-1024.
Dannenberger D & Lerz A. 1996. Polychlorinated biphenyls (PCB) and organochlorine pesticides in sediments of the Baltic and coastal waters of Mecklenburg-Vorpommern. German Journal of Hydrography, 48:(1) 5-26.
de Swart RL, Ross PS, Vedder LJ, Timmerman HH, Heisterkamp S, van Loveren H, Vos JG, Reijnders PJH & E. OADM. 1994. Impairment of immune function in harbor seals (Phoca vitulina) feeding on fish from polluted waters. Ambio, 23:(2) 155-159.
de Swart RL, Ross PS, Vos JG & E. OADM. 1996. Impaired immunity in Harbour Seals (*Phoca vitulina*) exposed to bioaccumulated environmental contaminants: A review of long-term feeding study. Environ. Health Perspect., 104:(4) 823-829.
Dearth MA & Hites RA. 1991. Complete analysis of technical chlordane using negative ionization mass spectrometry. Environ. Sci. Technol., 25:(2) 245-255.
Dewailly E, Dodin S, Verreault R, Ayotte P, Sauve L & Morin J. 1993. High organochlorine body burden in breast cancer women with estogen receptors. Organohalogen Compounds, 13: 385-388.
EEA. 1998. Europe's Environment: The Second Assessment. European Environment Agency, Copenhagen. pp. 115-217.
Failing MOK, Lang U, Gent H-J, Georgii S & Brunn H. 1999. Contamination of human milk in Middle Hesse, Germany - a cross-sectional study on the changing levels of chlorinated pesticides, PCB congeners and recent levels of nitro musks. Chemoshere, 38:(1) 13-32.
Falandysz J. 1994. Polychlorinated biphenyl concentrations in cod-liver oil: evidence of a steady-state condition of these compounds in the Baltic area oils and levels noted in Atlantic oils. Arch Environ Contam Toxicol, 27: 266-271.
Falandysz J. 2000. Residues of hexachlorobenzene in Baltic fish and estimation of daily intake of this compound and pentachlorobenzene with fish and fishery products in Poland. Polish Journal of Environmental Studies, 9:(5) 377-383.
Falandysz J, Yamashita N, Tanabe S & Tatsukawa R. 1992. Isomer-specific analysis of PCBs including toxic coplanar isomers in canned cod livers commercially processed in Poland. Z Lebensm Unters Forsch, 194: 120-123.
Falandysz J, Amashita N, Tanabe S, Tatsukawa R, Rucinska L & Skora K. 1994a. Congener-specific data on Polychlorinated biphenyls in tissues of comon porpoise from Puck Bay, Baltic Sea. Arch. Environ. Contam. Toxicol., 26: 267-272.
Falandysz J, Kannan K, Tanabe S & Tatsukawa R. 1994b. Organochlorine pesticides and polychlorinated biphenyls in cod-liver oils: North Atlantic, Norwegian Sea, North Sea and Baltic Sea. Ambio, 23:(4-5) 288-292.
Falandysz J, Yamashita N, Tanabe S & Tatsukawa R. 1994c. Congener-specific data of polychlorinated biphenyl residues in human adipose tissue in Poland. Science of the total enviroment, 149: 113-119.
Falandysz J, Yamashita N, Tanabe S, Tatsukawa R, Rucinska L, Mizera T & Jakuczun B. 1994d. Congener-specific analysis of polychlorinated biphenyls in White-Tailed Sea Eagles *Haliaeetus albicilla* collected in Poland. Arch Environ Contam Toxicol, 26: 13-22.
Falandysz J, Strandberg L, Bergqvist PE, Kulp SE, Strandberg B & Rappe C. 1996. Polychlorinated naphthalenes in sediment and biota from the Gdansk Basin, Baltic Sea. Environmental Science and Technology, 30:(11) 3266-3274.
Falandysz J, Danisiewicz D, Strandberg L, Strandberg B, Bergqvist P-A & Rappe C. 1997a. DDTs, HCBz and PCBz in Stickleback from various sites in the Gulf of Gdánsk. Organohalogen Compounds, 32: 354-356.
Falandysz J, Danisiewicz D, Strandberg L, Strandberg B, Bergqvist P-A & Rappe C. 1997b. Pentachlorobenzene, hexachlorobenzene and DDTs in a pelagic food chain in the Baltic Sea. Organohalogen Compounds, 32: 370-373.
Falandysz J, Dembowska A, Strandberg L, Strandberg B, Bergqvist P-A & Rappe C. 1997c. Congener-specific data of PCBs in some species of fish from the Gulf of Gdánsk, Baltic Sea. Organohalogen Compounds, 32: 358-363.
Falandysz J, Florek A, Strandberg L, Strandberg B, Bergqvist P-A & Rappe C. 1997d. PCDDs and PCDFs in biota from the southern part of the Baltic Sea. Organohalogen Compounds, 32: 167-171.
Falandysz J, Strandberg B, Danisiewicz D, Strandberg L, Bergqvist P-A & Rappe C. 1997e. Spatial distribution of DDTs, HCBz and PCBz in plankton in the southern part of the Baltic proper. Organohalogen Compounds, 32: 249-353.
Falandysz J, Strandberg B, Strandberg L, Bergqvist P-A & Rappe C. 1997f. Concentrations and biomagnification of polychlorinated naphthalenes in black cormorants *Phalacrocorax carbo sinensis* from the Gulf of Gdansk, Baltic Sea. The Science of the Total Environment, 204: 97-106.
Falandysz J, Strandberg L, Bergqvist P-A, Strandberg B & Rappe C. 1997g. Spatial distribution and bioaccumulation of polychlorinated naphthalenes (PCNs) in mussel and fish from the Gulf of Gdansk, Baltic Sea. The Science of the Total Environment, 203: 93-104.
Falandysz J, Dembowska A, Strandberg L, Strandberg B & Rappe C. 1998a. Spatial distribution of PCBs in flounder, perch and lamprey from the Gulf of Gdansk, Baltic Sea. Organohalogen Compounds, 39: 223-228.
Falandysz J, Dembowska A, Strandberg L, Strandberg B & Rappe C. 1998b. Spatial distribution of PCBs in three-spined stickleback from the beach zone in the gulf of Gdansk, Baltic Sea. Organohalogen Compounds, 39: 229-234.
Falandysz J, Strandberg B, Strandberg L, Bergqvist PA & Rappe C. 1998c. Concentrations and spatial distribution of chlordanes and some cyclodiene pesticides in Baltic plankton. The Science of the Total Environment, 215: 253-258.

Falandysz J, Strandberg B, Strandberg L, Bergqvist P-A & Rappe C. 1998d. Spatial distribution of TCPM-H and TCPM-OH in blue mussel and fish from the Gulf of Gdansk, Baltic Sea. Bulletin of Environmental Contamination and Toxicology, 61: 411-418.

Falandysz J, Strandberg B, Strandberg L, Rappe C, T. M & Kalisinska E. 1998e. *Tris*(4-chlorophenyl)methane and -methanol in biota from the Polish coastal waters in the Baltic Sea. Organohalogen Compounds, 39: 439-444.

Falandysz J, Strandberg B, Strandberg L, Bergqvist P-A & Rappe C. 1999a. Cyclodiene pesticide residues in molluscs, crustaceans and fish in the Gulf of Gdansk, Baltic Sea. Organohalogen Compounds, 43: 147-150.

Falandysz J, Strandberg B, Strandberg L & Rappe C. 1999b. Tris(4-chlorophenyl)methane and tris(4-chlorophenyl)methanol in sediment and food webs from the Baltic south coast. Environmental Science & Technology, 33:(4) 517-521.

Falandysz J, Trzosinska A, Szefer P, Warzocha J & Draganik B. 2000a. The Baltic Sea, especially southern and eastern regions. In: Seas at The Millennium: An Environmental Evaluation (Sheppard C, Ed.): Elsevier Science Ltd, pp. 99-120.

Falandysz J, Strandberg L, Strandberg B, Bergqvist P-A & Rappe C. 2000b. Pentachlorobenzene and hexachlorobenzene in fish in the Gulf of Gdansk. Polish Journal of Environmental Studies, 9:(2) 129-132.

Fein GG, Jacobson JL, Jacobson SW, Schwartz PM & Dowler JK. 1984. Prenatal exposure to polychlorinated biphenyls: Effects on birth size and gestational age. Journal of Pediatrics, 105: 315-328.

Fiedler H, Buckley-Golder D, Coleman P, King K & Petersen A. 1999. Compilation of EU dioxin exposure and health data: environmental levels. Organohalogen Compounds, 43: 151-154.

Foster WG. 1995. The reproductive toxicology of Great Lakes contaminants. Environ. Health Perspec., 103:(Suppl 9) 63-69.

Fromberg A, Cederberg T, Hilbert G & Büchert A. 2000. Levels of toxaphene congeners in fish from Danish waters. Chemosphere, 40: 1227-1232.

Fürst P. 2000. PCDDs/PCDFs in human milk - still a matter of concern? Organohalogen Compounds, 48: 111-114.

Fürst P, Fürst C & Wilmers K. 1994. Human milk as a bioindicator for body burden of PCDDs, PCDFs, organochlorine pesticides, and PCBs. Environmental Health Perspectives, 102:(1) 187-193.

Gilman AP, van Oostdam JC, Hansen JC, Odland JO, Tchachtchine V, Walker J, Lagerkvist BJ, Olafsdottir K, Weber JP, Bjerregaard P & Klopov V. 1997. The circumpolar AMAP maternal blood monitoring study: 1995-1996. ISBN 82-7655-060-6.

Granby K & Kinze CC. 1991. Organochlorines in Danish and west Greenland harbour porpoises. Marine Pollution Bulletin, 22:(9) 458-462.

Gustafsson K, Björk M, Burreau S & Gilek M. 1999. Bioaccumulation kinetics of brominated flame retardants (polybrominated diphenyl ethers) in blue mussels (*Mytilus edulis*). Environmental Toxicology and Chemistry, 18:(6) 1218-1224.

Haglund PS, Wiberg KI & Zook DR. 1995. Identification and Quantification of Monobromopolychlorobiphenyls in Baltic Ringed Seal and a Technical PCB (Clophen A50). Environ. Sci. Technol., 29:(11) 2801-2805.

Haglund PS, Zook DR, Buser HR & Hu J. 1997. Identification and quantification of polybrominated diphenyl ethers and methoxy-polybrominated diphenyl ethers in Baltic Biota. Environmental Science and Technology, 31:(11) 3281-3287.

Hagmar L, Linden K, Nilsson A, Norrving B, Akesson B, Schütz A & Möller T. 1992. Cancer incidence and mortality among Swedish Baltic Sea fishermen. Scand. J. Work Environ. health, 18: 217-224.

Hagmar L, Hallberg T, Leja M, Nilsson A & Schütz A. 1995. High consumption of fatty fish from the Baltic Sea is associated with changes in human lymphocyte subset levels. Toxicology Letters, 77: 335-342.

Hagmar L, Becher G, Heikkilä A, Frankman O, Dyremerk E, Schütz A, Ahlborg UG & Dybing E. 1998. Consumption of fatty fish from the Baltic Sea and PCB in whole venous blood, plasma and cord blood from delivering women in the Aland/Turku Archipelago. Journal of Toxicology and Environmental Health, Part A, 53: 581-591.

Hall AJ, Law DE, Harwood J, Ross HM, Kennedy S, Allchin CR, Campbell LA & Pomeroy PP. 1992. Organochlorine levels in common seals (*Phoca vitulina*) which were victims and survivors of the 1988 phocine distemper epizootic. The Science of the Total Environ., 115: 145-162.

Hansen E. 2000. Substance flow analysis for dioxins in Denmark. Environmental Project No 570 2000. Milijoprojeckt. Danish Environmental Protection Agency.

Haraguchi K, Athanasiadou M, Bergman A, Hovander L & Jensen S. 1992. PCB and PCB methyl sulfones in selected groups of seals from Swedish waters. Ambio, 21:(8) 546-549.

Harding KC & Harkonen TJ. 1999. Development in the Baltic grey seal (Hailochoerus grypus) and the ringed seal (Phoca hispida) populations during the 20th century. Ambio, 28:(7) 619-627.

Heide-Jorgensen MP, Härkönen T & Åberg P. 1992. Long-term effects of epizootic in Harbor seals in the Kattegat-Skagerrak and adjacent areas. Ambio, 21:(8) 511-516.

Helander B, Olsson A, Bignert A, Litzén K, Asplund L & Bergman Å. 1998. Abnormal dehydration in eggs of the white-tailed sea eagle (*Haliaeetus albicilla*) in Sweden - a persisting effect from high exposure to organochlorines? Organohalogen Compounds, 39: 423-426.

HELCOM. 2001a. Convention on the Protection of the Marine Environment of the Baltic Sea Area. Conclusions of the 4th Periodic Assessment (full assessment not published). http://www.helcom.fi/monas/FPA4_Conclusions.htm.

HELCOM (2001b). http://www.helcom.fi/

Helle E, Olsson M & Jensen S. 1976a. DDT and PCB levels and reproduction in ringed seal from the Bothnian Bay. Ambio, 5:(4) 188-189.

Helle E, Olsson M & Jensen S. 1976b. PCB levels correlated with pathological change in seal uteri. Ambio, 5:(5-6) 261-263.

Hilbert G, Cederberg T & Büchert A. 1996. Time trend studies of chlorinated pesticides, PCBs and dioxins in Danish human milk. Organohalogen Compounds, 30: 123-126.

Hsu C-C, Yu M-LM, Chen Y-CJ, Guo Y-LL & Rogan WJ. 1994. The Yu-cheng rice oil poisoning incident. In: Dioxins and Health (Schecter A, Ed.), New York: Plenum Press, pp. 661-684.

Hummert K, Vetter W & Luckas B. 1995. Levels of Alpha-HCH, Lindane, and enantiomeric ratios of Alpha-HCH in marine mammals from the northern hemisphere. Chemosphere, 31:(6) 3489-3500.

Hunter D, Hankinson S, Laden F, Colditz GA, Manson J, Willett WC, Speizer FE & Wolff MS. 1997. Plasma organochlorine levels and the risk of breast cancer. New Eng. J. Med., 337:(18) 1253-1258.

Ishaq R, Lundgren K, Strandberg B, Tysklind M & Broman D. 2000. Accumulation of polychlorinated napthalenes (PCNs) in Baltic Sea sediments. Organohalogen Compounds, 47:(21-23).

Iwata H, Tanabe S, Sakai N & Tatsukawa R. 1993. Distribution of Persistent Organochlorines in the Oceanic Air and Surface Seawater and the Role of Ocean on their Global Transport and Fate. Environmental Science and Technology, 27:(6) 1080-1098.

Jacobs MN, Johnston PA, Wyatt CL, Santillo D & French MC. 1997. Organochlorine pesticide and PCB residues in pharmaceutical, industrial and food grade fish oils. Int. J. Environment and Pollution, 8:(1/2) 74-93.

Jacobson JL & Jacobson SW. 1996. Intellectual impairment in children exposed to polychlorinated biphenyls in utero. New England Journal of Medicine, 335: 783-789.

Jansson B-O & Dahlberg K. 1999. The environmental status of the Baltic Sea in the 1940s, today, and in the future. Ambio, 28:(4) 312-319.

Jenssen BM. 1996. An overview of exposure to, and effects of, petroleum oil and organochlorine pollution in Grey seals (*Halichoerus grypus*). The Science of the Total Environment, 186: 109-118.

Johnston PA, Stringer RL, Santillo D, Stephenson AD, Labounskaia I & McCartney HMA. 1997. Towards zero-effluent pulp and paper production. Environmental Science and Pollution Research, 4:(3) 130.

Jonsson P. 2000. Sediment burial of PCBs in the offshore Baltic Sea. Ambio, 29:(4-5) 260-267.

Jonsson P, Grimvall A, Cederlöf M & Hildén M. 1996. Pollution threats to the Gulf of Bothnia. Royal Swedish Academy of Sciences, pp. 22-27.

Kannan K & Falandysz J. 1997. Butyltin residues in sediment, fish, fish-eating birds, Harbour Porpose and human tissues from the Polish coast of the Baltic Sea. Mar. Poll. Bull., 34:(3) 203-207.

Karlström Ö. 1999. Development of the M74 syndrome in wild populations of Baltic salmon (*Salmo salar*) in Swedish rivers. Ambio, 28:(1) 82-86.

Kautsky L & Kautsky N. 2000. The Baltic Sea, including Bothnian Sea and Bothnian Bay. In: Seas at the Millennium: An Environmental Evaluation (Sheppard C, Ed.): Elsevier Science Ltd, pp. 99-120.

Kennedy S. 1996. Infectious diseases of cetacean populations. In: The conservation of whales and dolphins (Simmonds M & Hutchinson J, Eds.): John Wiley & Sons. ISBN 0-471-96561-8.

Kiviranta H, Purkunen R & Vartiainen T. 1998. Levels of PCDD/Fs and PCBs in human milk in 1994 in Finland: Decrease in concentrations from 1987 to 1994. Organohalogen Compounds, 38: 121-124.

Kiviranta H, Vartiainen T, Verta M, Tuornisto JT & Tuornisto J. 2000. High fish-specific dioxin concentrations in Finland. The Lancet, 355: 1883-1885.

Kjeller L-O & Rappe C. 1995. Time trends in levels, patterns and profiles for polychlorinated dibenzo-*p*-dioxins, dibenzofurans, and biphenyls in a sediment core from the Baltic Proper. Environmental Science & Technology, 29:(346-365).

Kocan A, Drobna B, Petrik J, Chovancova J, Patterson DG & Needham LL. 1995. Levels of PCBs and selected organochlorine pesticides in humans from selected areas of the Slovak Republic. Part III: Milk. Organohalogen Compounds, 26: 187-192.

Koistinen J, Vuorinen PJ & Paasivirta J. 1993. Contents and origin of polychlorinated diphenyl ethers (PCDE) in salmon from the Baltic Sea, Lake Saimaa and the Tenojoki River in Finland. Chemosphere, 27:(12) 2365-2380.

Koistinen J, Koivusaari J, Nuuja I & Paasivirta J. 1995a. PCDEs, PCBs, PCDDs and PCDFs in Black Guillemots and White-Tailed Sea Eagles from the Baltic Sea. Chemosphere, 30:(9) 1671-1684.

Koistinen J, Mussalo-Rauhamaa H & Paasivirta J. 1995b. Polychlorinated diphenyl ethers, dibenzo-*p*-dioxins and dibenzofurans in Finnish human tissues compared to environmental samples. Chemosphere, 31:(10) 4259-4271.

Koistinen J, Herve S, Paukku R, Lahtiperä M & Paasivirta J. 1997a. Chloroaromatic pollutants in mussels incubated in two Finnish watercourses polluted by industry. Chemosphere, 34:(12) 2553-2569.

Koistinen J, Koivusaari J, Nuuja I, Vuorinen PJ, Paasivirta J & Giesy JP. 1997b. 2,3,7,8-tetrachlorodibenzo-*p*-dioxin equivalents in extracts of Baltic white-tailed Sea Eagles. Environmental Toxicology and Chemistry, 16:(7) 1533-1544.

Koistinen J, Stenman O, Haahti H, Suonpera M & Paasivirta J. 1997c. Polychlorinated diphenyl ethers, dibenzo-*p*-dioxins, dibenzofurans and biphenyls in seals and sediment from the Gulf of Finland. Chemosphere, 35:(6) 1249-1269.

Koistinen J, Nyman M, Helle E & Vartiainen T. 1999. Dioxins in ringed seals (*Phoca hispida*) from the Baltic Sea and Spitsbergen. Organohalogen Compounds, 43: 365-368.

Korhonen M, Mannio J & Vartiainen T. 1995. The level of PCBs in aquatic biota in Finland. Organohalogen Compounds, 24: 453-456.

Korhonen M & Vartiainen T. 1997. Concentrations of PCDDs and PCDFs in Baltic herring (*Clupea harengus*) and Northern Pike (*Esox lucius*) in Finnish coastal area from 1989-1993. Organohalogen Compounds, 32: 299-304.

Kostamo A, Viljanen M, Pellinen J & Kukkonen J. 2000. EOX and organochlorine compounds in fish and ringed seal samples from Lake Ladoga, Russia. Chemosphere, 41: 1733-1740.

Larsson P, Backe C, Bremle G, Eklöv A & Lennart O. 1996. Persistent pollutants in a salmon population (*Salmo salar*) of the southern Baltic Sea. Can J Fish Aquat Sci, 53: 62-69.

Larsson P, Andersson A, Broman D, Nordbäck J & Lundberg E. 2000. Persistent organic pollutants (POPs) in pelagic systems. Ambio, 29:(4-5) 202-209.

Law RJ, Allchin CR & Harwood J. 1989. Concentrations of Organochlorine compounds in the blubber of seals from Eastern and North-eastern England, 1988. Mar. Poll. Bull., 20:(3) 110-115.

Lee KM, Kruse H & Wasserman O. 1996a. The pattern of organochlorines in mussels *Mytilus edulis* L. from the south west Baltic Sea. Arch Environ Contam Toxicol, 31: 68-76.

Lee KM, Kruse H & Wasserman O. 1996b. Seasonal fluctuation of organochlorines in *Mytilus edulis* L. from the south west Baltic Sea. Chemosphere, 32:(10) 1883-1895.

Lommel A, Kruse H, Müller E & Wasserman O. 1992. Organochlorine pesticides, octachlorostyrene, and mercury in blood of Elb River residents, Germany. Arch. Environ. Contam. Toxicol., 22: 14-20.

Looser R, Froescheis O, Cailliet GM, Jarman WM & Ballschmiter K. 2000. The deep-sea as a final global sink of semivolatile persistent organic pollutants? Part II: organochlorine pesticides in surface and deep-sea dwelling fish of the North and South Atlantic and the Monterey Bay Canyon (California). Chemosphere, 40: 661-670.

Lopez-Carrillo L, Torres-Arreola L, Torres-Sanchez L, Espinosa-Torres F, Jimenez C, Cebrian M, Waliszewski S & Saldate O. 1996. Is DDT use a public healthe problem in Mexico? Environmental Health Perspectives, 104:(6) 584-588.

Luebke RW, Hodson PV, Faisal M, Ross PS, Grasman KA & Zelikoff J. 1997. Aquatic pollution-induced immunotoxicity in wildlife species. Fundamental and Applied Toxicology, 37: 1-15.

Masuda Y. 1994. The Yusho rice oil poisoning incident. In: Dioxins and health (Schecter A, Ed.), New York: Plenum Press, pp. 633-659.

McHugh B, Nixon E, Klungsoyr J, Besselink H, Brouwer A, Rimkus G, Leonards P & de Boer J. 2000. Survey of toxaphene concentrations in fish from European waters. Organohalogen Compounds, 47: 117-120.

Mitchell SH & Kennedy S. 1992. Tissue concentrations of organochlorine compounds in common seals from the coast of Northern Ireland. Sci. Tot. Environ., 115: 163-177.

Muir D, Kidd K, Koczanski K, Stern GA, Alaee M, Jantunen L & Bidleman T. 1997. Bioaccumulation of toxaphene congeners in freshwater and marine food webs. Organohalogen Compounds, 33: 34-38.

Müller G, Siebert U, Wünschmann A, Artelt A & Baumgärtner W. 2000. Immunohistological and serological investigation of morbillivirus infection in harbour porpoises (*Phocoena phocoena*) from the German Baltic and North Sea. Veterinary Microbiology, 75: 17-25.

Munro R, Ross H, Cornwell C & Gilmour J. 1992. Disease conditions affecting common seals (Phoca vitulina) around the Scottish mainland, September-November 1988. Sci. of Tot. Environ., 115: 67-82.

Mussalo-Rauhamaa H & Lindstrom G. 1995. PCDD and PCDF levels in human milk in Estonia and certain Nordic countries. Organohalogen Compounds, 26: 245-246.

Norén K. 1993. Contemporary and retrospective investigations of human milk in the trends of organochlorine contaminants in Sweden. Sci. Tot. Environ., 139/140: 347-355.

Norén K & Meironyté D. 2000. Certain organochlorine and organobromine contaminants in Swedish human milk in perspective of past 20-30 years. Chemosphere, 40: 1111-1123.

Norén K, Weistrand C & Karpe F. 1999. Distribution of PCB congeners, DDE, hexachlorobenzene, and methylsulfonyl metabolites of PCB and DDE among various fractions of human blood plasma. Arch. Environ. Contam. Toxicol., 37: 408-414.

Norstrom RJ & Muir DCG. 1994. Chlorinated hydrocarbon contaminants in arctic marine mammals. The Science of the Total Environment, 154: 107-128.

Öberg M, Stern N, Casabona H, Johansson N, Jensen S, Haglund P & Håkansson H. 1999. Levels of extractable organically bound chlorine and identified organohalogen pollutants in Baltic Sea herring oil, its fractions and rat liver. Organohalogen Compounds, 43: 401-404.

Olsson A & Bergman A. 1995. A new persistent contaminant detected in Baltic wildlife: Bis (4-chlorophenyl) Sulfone. Ambio, 24:(2) 119-123.

Olsson A, Vitinsh M, Plikshs M & Bergman Å. 1999. Halogenated environmental contaminants in perch (*Perca fluviatilis*) from Latvian coastal areas. The Science of the Total Environment, 239: 19-30.

Olsson M, Karlsson B & Ahnland E. 1994. Diseases and environmental contaminants in seals from the Baltic and Swedish west coast. Sci. Tot. Environ., 154: 217-227.

OSPAR. 1998. The Sintra Statement (Final Declaration of the Ministerial Meeting of the OSPAR Commission, Sintra 20-24th July 1998). OSPAR 98/14/1 Annex 45. OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic.

Pallemaerts M. 1998. Production, Toxics and Transnational law: An inquiry into the effectiveness of international and European Community Environmental Law. Part 1. Proefschrift voorgelegd tot het behalen van de graad van Doctor in de Rechten, Faculteit der Rechtsgeleerdheid Vrije, Universiteit Brussels, November 1998, pages 168-194

Päpke O. 1999. Background contamination of humans with dioxins and dioxin-like PCBs. Organohalogen Compounds, 44: 5-8.

Penninks AH. 1993. The evaluation of data-derived safety factors for bis(tri-*n*-butyltin)oxide. Food Add. Contam., 10: 351-361.

Polder A, Becher G, Savinova TN & Skaare JU. 1996. Dioxins, PCBs and some chlorinated pesticides in human milk from the Kola Peninsula, Russia. Organohalogen Compounds, 30: 158-161.

Polder A, Føreid S & Odland JO. 1998. PCBs, Toxaphenes and other chlorinated pesticides in human milk from the Archangels district, Russia. Organohalogen Compounds, 38: 155-158.

Ratcliffe DA. 1967. Decrease in eggshell weight in certain birds of prey. Nature, 215: 208-210.

Reijnders P. 1994. Toxicokinetics of chlorobiphenyls and associated physiological reponses in marine mammals, with particular reference to their potential fr ecotoxicological risk assessment. The Science of the Total Environment, 154: 229-236.

Reijnders P. 1996. Organohalogen and heavy metal contamination in cetaceans: observed effects, potential impact and future prospects. In: The conservation of whales and dolphins: Science and Practice (Simmonds M & Hutchinson J, Eds.), Chichester, UK: J Wiley & Sons, pp. 205-217.

Reijnders P & Brasseur S. 1992. Xenobiotic induced hormonal and associated development disorders in marine organisms and related effects in humans; an overview. In: Chemically-induced alterations in sexual and functional development: the wildlife/human connection (Colborn T & Clement C, Eds.), Princeton, New Jersey: Princeton Scientific Publishing Co. Inc., pp. 159-174.

Rimkus GG & Wolf M. 1995. Nitro musk fragrances in biota from freshwater and marine environment. Chemosphere, 30:(4) 641-651.

Roos A, Bergman A, Greyerz E & Olsson M. 1998. Time trend studies on ΣDDT and PCB in juvenile grey seals (*Halichoerus grypus*), fish and guillemot eggs from the Baltic Sea. Organohalogen Compounds, 39: 109-112.

Roots O. 1995. Organochlorine pesticides and polychlorinated biphenyls in the ecosystem of the Baltic Sea. Chemosphere, 31:(9) 4085-4097.

Roots O. 1998. Biogeomonitoring of toxic chloroorganic compounds in the ecosystem of the Baltic Sea. Ecological Chemistry, St Petersburg, 71:(1) 55-64.

Roots O & Talvari A. 1999. Bioaccumulation of toxic organic compounds and their isomers into the organisms of seals in west-Estonian archipelago biosphere reserve. Environmental Monitoring and Assessment, 54: 301-312.

Ross PS, de Swart RL, Reijnders PJH, van Loveren H, Vos JG & Osterhaus A. 1995. Contaminant-related suppression of delayed-type hypersensitivity and antibody responses in Harbour Seals fed herring from the Baltic Sea. Environ. Health Perspec., 103:(2) 162-167.

Rylander L & Hagmar L. 1995. Mortality and cancer incidence among women with a high consumption of fatty fish contaminated with persistent organochlorine compounds. Scand. J. Work Environ. Heath, 21: 419-426.

Rylander L & Hagmar L. 1999. No evidence for congenital malformations or prenatal death in infants born to women with a high dietary intake of fish contaminated with persistent organochlorines. Int Arch Occup Environ Health, 72: 121-124.

Rylander L & Hagmar L. 2000. Medical and psychometric examinations of conscripts born to mothers with a high intake of fish contaminated with persistent organochlorines. Scand J Work Environ Health, 26:(3) 207-212.

Rylander L, Strömberg U & Hagmar L. 1995. Decreased birthweight among infants born to women with a high dietary intake of fish contaminated with persistent organochlorine compounds. Scand J Work Environ Health, 21: 368-375.

Rylander L, Strömberg U & Hagmar L. 2000. Lowered birth weight among infants born to women with a high intake of fish contaminated with persistent organochlorine compounds. Chemosphere, 40: 1255-1262.

Sandau CD, Meerts IATM, Letcher RJ, McAlees AJ, Chittim B & Norstrom RJ. 2000. Identification of 4-hydroxyheptachlorostyrene in polar bear plasma and its binding affinity to transthyretin: A metabolite of octachlorostyrene. Environmental Science and Technology, 34:(18) 3871-3877.

Savitz DA. 1994. Re: Breast cancer and serum organochlorines: a prospective study among white, black, and asian women. Journal of the National Cancer Institute, 86:(16) 1255.
Schade G & Heinzow B. 1998. Organochlorine pesticides and polychlorinated biphenyls in human milk of mothers living in northern Germany: current extent of contamination, time trend from 1986 to 1997 and factors that influence the levels of contamination. The Science of the Total Environment, 215: 31-39.
Schecter A, Fürst P, Fürst C, Groebel W, Kolesnikov S, Savchenkov M, Beim A, Boldonov A, Trubitsun E & Vlasov B. 1990. Levels of dioxins, dibenzofurans and other chlorinated xenobiotics in human milk from the Soviet Union. Chemoshere, 20:(7-9) 927-934.
Schoula R, Hajslova J, Bencko V, Pustka J, Holadova K & Vizek V. 1996. Occurrence of persistent organochlorine contaminants in human milk collected in several regions of Czech Republic. Chemosphere, 33:(8) 1485-1494.
Schröter-Kermani C, Helm D, Herrman T & Päpke O. 2000. The German environmental specimen bank - application in trend monitoring of polybrominated diphenyl ethers in human blood. Organohalogen Compounds, 47: 49-52.
Senthilkumar J, Duda CA, Villeneuve DL, Kannan K, Falandysz J & Giesy JP. 1999. Butyltin compounds in sediment and fish from the Polish coast of the Baltic Sea. Environ Sci & Pollut Res, 6:(4) 200-206.
Siebert U, Wünschmann A, Weiss R, Frank H, Benke H & Frese K. 2001. Post-mortem findings in harbour porpoises (*Phocoena phocoena*) from the German North and Baltic Seas. J Comp Path, 124: 102-114.
Simmonds MP, Johnston PA & French MC. 1993. Organochlorine and mercury contamination in United Kingdom seals. The Veterinary Record, 20 March 1993 291-295.
Simmonds MP & Mayer SJ. 1997. An evaluation of environmental and other factors in some recent marine mammal moralities in Europe: implications for conservation and management. Environ. Rev., 5: 89-98.
Simonich SL & Hites RA. 1995. Global Distribution of Persistent Organochlorine Compounds. Sci., 269:(1995).
Sinkkonen S & Paasivirta J. 2000. Polychlorinated organic compounds in the Arctic cod lliver: trends and profiles. Chemosphere, 40: 619-626.
Sjödin A, Hagmar L, Klasson-Wehler E, Björk J & Bergman A. 2000. Influence of the consumption of fatty Baltic Sea fish on plasma levels of halogenated environmental contaminants in Latvian and Swedish men. Environmental Health Perspectives, 108:(11) 1035-1041.
Skei J, Larsson P, Rosenberg R, Jonsson P, Olsson M & Broman D. 2000. Eutrophication and contaminants in aquatic ecosystems. Ambio, 29:(4-5) 184-193.
Söderström M, Wachtmeister CA & Förlin L. 1994. Analysis of chlorophenolics from bleach kraft mill effluents (BKME) in bile of perch (*Perca fluviatilis*) from the Baltic Sea and development of an analytical procedure also measuring chlorocatechols. Chemosphere, 28:(9) 1701-1719.
Strandberg B, Strandberg L, Bergqvist P-A, Falandysz J & Rappe C. 1998a. Concentrations and biomagnification of 17 chlordane compounds and other organochlorines in Harbour Porpoise (*Phocoena phocoena*) and herring from the southern Baltic Sea. Chemosphere, 37:(9-12) 2513-2523.
Strandberg B, van Bavel B, Bergqvist P-A, Broman D, Ishaq R, Naf C, Pettersen H & Rappe C. 1998b. Occurrence, sedimentation and spatial variations of organochlorine contaminants in settling particulate matter and sediments in the northern part of the Baltic Sea. Environmental Science & Technology, 32: 1754-1759.
Strandberg B, Bandh C, van Bavel B, Bergqvist P-A, Broman D, Ishaq R, Näf C & Rappe C. 2000. Organochlorine compounds in the Gulf of Bothnia: sediment and benthic species. Chemosphere, 40: 1205-1211.
Strandberg L, Strandberg B, Bergqvist P-A, Rappe C & Falandysz J. 1998c. PCBs and organochlorine pesticides in breast muscles of White-Tailed Sea Eagles *Haliaeetus albicilla* breeding in Poland. Organohalogen Compounds, 39: 453-456.
Svensson BG, Hallberg T, Nilsson A, Schütz A & Hagmar L. 1994. Parameters of immunological competence in subjects with high consumption of fish contaminated with persistent organochlorine compounds. Int Arch Occup Environ, 65: 351-358.
Svensson BG, Mikoczy Z, Strömberg U & Hagmar L. 1995a. Mortality and cancer incidence among Swedish fishermen with a high dietary intake of persistent organchlorine compounds. Scand J Work Environ Health, 21: 106-115.
Svensson BG, Nilsson A, Jonsson E, Schutz A, Akesson B & Hagmar L. 1995b. Fish consumption and exposure to persistent organochlorine compounds, mercury, selenium and methylamines among Swedish fishermen. Scand J Work Environ Health, 21: 96-105.
Swedish National Food Administration. 1996. Livsmedelsverkets råd omfiskkonsumtion. Livsmedelsverket 9/96.
Tanabe S, Watanabe S, Kan H & Tatsukawa R. 1988. Capacity and mode of PCB metabolism in small cetaceans. Marine Mammal Science, 4:(2) 103-124.
Tanabe S, Iwate H & Tatsukawa R. 1994. Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals. The Science of the Total Environment, 154: 163-177.
Tanabe ST, Kannan K, Tabucanon MS, Siriwong C, Ambe Y & Tatsukawa R. 1991. Organochlorine pesticide and polychlorinated biphenyl residues in foodstuffs from Bangkok, Thailand. Environ. Poll., 72: 191-203.
UNEP. 1995. Decision 18/32 of the UNEP Governing Council: Persistent Organic Pollutants. United Nations Development Programme, May 1995.
Vallin L, Nissling A & Westin L. 1999. Potential factors influencing reproductive success in Baltic cod, *Gadus morhua*: A review. Ambio, 28:(1) 92-98.
Valters K, Olsson A, Vitinsh M & Bergman Å. 1999. Contamination sources in Latvia: levels of organochlorines in Perch (*Perca fluviatilis*) from Rivers Daugava and Lielupe. Ambio, 28:(4) 335-340.
van't Veer P, Lobbezoo IE, Martin-Moreno JM, Guallar E, Gomez-Aracena J, Kardinaal AFM, Kohlmeier L, Martin BC, Strain JJ, Thamm M, van Zoonen P, Baumann BA, Huttunen JK & Kok FJ. 1997. DDT (dicophane) and postmenopausal breast cancer in Europe: case-control study. British Medical Journal, 315: 81-85.
Vaz R, Slorach SA & Hofander Y. 1993. Organochlorine contaminants in Swedish human milk: studies conducted at the National Food Administration 1981-1990. Food Additives Contam., 10:(4) 407-418.
Vetter W. 1999. Presence of Q1, a new $C_9H_3Cl_7N_2$ organochlorine contaminant in evironmental samples. Organohalogen Compounds, 43: 433-436.
Vieth B, Heinrich-Hirsch B & Mathar W. 2000. Trends in dioxin intake and in human milk levels in Germany. Organohalogen Compounds, 47: 300-303.
vom Saal FS, Montano M & Wang M. 1992. Sexual differentiation in mammals. In: Chemically-induced alterations in sexual and functional development: The wildlife/human connection (Colborn T & Clement C, Eds.), Princeton, New Jersey: Princeton Scientific Publishing Co. Inc., pp. 17-85.

Vos JG, Dybing E, Greim HA, Ladefoged O, Lambré C, Tarazona JV & Brandt I. 2000. Health effects of endocrine-disrupting chemicals on wildlife, with special reference to the European situation. Critical Reviews in Toxicology, 30:(1) 71-133.

Wania F & Mackay D. 1993. Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions. Ambio, 22:(1) 10-18.

Wania F & Mackay D. 1996. Tracking the distribution of persistent organic pollutants. Environmental Science and Technology, 30:(9) A390-A396.

Wehler EK, Hovander L & Bergman A. 1997. New organohalogens in human plasma - Identification and qualification. Organohalogen Compounds, 33: 420-425.

Weisglas-Kuperus N. 1998. Neurodevelopmental, immunological and endocrinological indices of perinatal human exposure to PCBs and dioxins. Chemosphere, 37:(9-12) 1845-1853.

Weisglas-Kuperus N, Sas TCJ, Koopman-Esseboom C, Van der Zwan CW, De Ridder MAJ, Beishuizen A, Hooijkaas H & Sauer PJJ. 1995. Immunologic Effects of Background Prenatal and Postnatal Exposure to Dioxins and Polychlorinated Biphenyls in Dutch Infants. Pediatric Res. (1995), 38:(3) 404-408.

WHO. 1996. Levels of PCBs, PCDDs and PCDFs in human milk. Second round of WHO-coordinated study. Environmental Health in Europe Series No 3. EUR/ICP EHPM02 03 05. World Health Organisation.

Wiberg K, Rappe C & Haglund P. 1992. Analysis of bromo-, chloro- and mixed bromo/chloro-dibenzo-p-dioxins and dibenzofurans in salmon, osprey and human milk. Chemosphere, 24:(10) 1431-1439.

Wideqvist U, Jansson B, Reutergårdh L, Olsson M, Odsjö T & Uvemo U-B. 1993. Temporal trends of PCC in Guillemot eggs from the Baltic. Chemosphere, 27:(10) 1987-2001.

Witte J, Büthe A & Ternes W. 2000. Congener-specific analysis of toxaphene in eggs of seabirds from Germany by HRGC-NCI-MS using a carborane-siloxane copolymer phase (HT-8). Chemosphere, 41: 529-539.

Wolff MS, Toniolo PG, Lee EW, Rivera M & Dubin N. 1993. Blood levels of organochlorine residues and risk of breast cancer. J. of Nat. Canc. Institut., 85:(8) 648-652.

Wulff F & Rahm L. 1993. Accumulation of chlorinated organic matter in the Baltic Sea from 50 years of use - A threat to the environment. Marine Pollution Bulletin, 26:(5) 272-275.

Zakharov VM & Yablokov AV. 1990. Skull asymmetry in the Baltic grey seal: effects of environmental pollution. Ambio, 19:(5) 266-269.

Zook DR, Buser H-R, Bergqvist P-A, Rappe C & Olsson M. 1992. Detection of *tris*(chlorophenyl) methane and *tris*(chlorophenyl) methanol in ringed seal (*Phoca hispidus*) from the Baltic Sea. Ambio, 21:(8) 557-560.

GREENPEACE

April 2001

Greenpeace International
Keizersgracht 176
1016 DW Amsterdam
The Netherlands

Tel: (31) 20 523 6222
Fax: (31) 20 523 6200
www.greenpeace.org

ISBN: 90-73361-71-0