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Environmental impact of PCBs in the marine environment

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Introduction

Since Jensen [1966] found PCBs in environmental samples, there has been much interest in the contamination of the environment by that group of chemicals. Due to the Yusho-illness (Japan 1968) research was focused on the effects of PCBs on human health. These results of occupational exposure had, in fact, been known since the 1930s. Detrimental effects of PCBs have been shown in numerous laboratory experiments and as a consequence, in case of deviating population parameters found in the field, elevated levels of PCBs were associated with, for example, an observed population decline. To establish a causal relationship between the presence of PCBs and biological disorders is often rather complicated, but this should neither lead to a diversion of the actual problem nor to ignorance of a potential problem.

In this paper three approaches will be used in order to assess the environmental impact of PCBs in the marine environment. First, literature on experimental and epidemiological effects will be reviewed, although this review will not be comprehensive. Only that information which can be used to assess the hazards for marine ecosystems is included. The second section will be devoted to implications arising by interpreting and comparing experimental findings to situations elsewhere, where similar symptoms were observed and elevated PCB-levels found. Finally two case studies, where effects are attributed to the impact of PCBs, will be discussed in detail to elucidate the complex nature and extent of the noxious actions of those pollutants.

Effects of PCBs

Experimental effects

The effects of PCBs have been amply described at all trophic levels in experiments. Acute mortality as a result of PCB poisoning was found in bacteria, algae, protozoa, shrimps, molluscs, fish [Stalling and Mayer, 1972], birds [Koeman *et al.*, 1973], and mammals [Aulerich *et al.*, 1973; den Boer, 1983]. Acute oral LD_{50} of PCBs in rats, rabbits, and mice ranges from 1–10 g/kg bodyweight. There is some indication that young animals may be more sensitive than adults, and females more sensitive than males. Symptoms associated with administration of a toxic dose of PCB (Aroclor 1242) in rats, consisted of diarrhoea, adipsia, oliguria, anorexia, erythema of limits followed by taxia, coma, and death. Death occurred up to 14 days following administration of the toxic dose. Twenty-four hours after administration of a single toxic dose of Aroclor 1242, gross pathology examination revealed that all organs appeared normal except for the liver and kidneys. Dermal toxicity of PCBs is also important, in rabbits LD_{50} by application of PCBs on the skin is about 2.0 g/kg bodyweight.

However, because the high concentrations of PCBs cited above are rarely found in the environment, aspects of subacute toxicity of PCBs appear to be of greater concern. One of the major biochemical effects of PCBs is the induction of microsomal enzyme activities in the liver (mixed-function oxydase (MFO)). Not only PCBs themselves are metabolised by MFO enzymes but the rates of metabolism of other compounds such as steroids, including hormones, are also affected. Values reported for threshold of enzyme induction vary between 0.5 and 2.5 mg/kg. The potency of individual PCB-congeners appears to depend largely on chlorine substitution patterns [Safe *et al.*, 1982].

Vos and Koeman [1970] found porphyria in PCB-treated chickens and suggested that this was due to the increase of mitochondrial ALA-synthetase. Debets [1981] found that apparently a PCB-congener has to be metabolised before it can show porphyrogenic activity. Except porphyria, PCBs administered at relatively high concentrations (50 mg/kg or higher in their diet) to laboratory animals have been shown to cause disfunction of the thyroid, changes in liver enzymes, changes in liver to bodyweight ratio, various disorders in the liver and alteration of the level of utilisation of corticosteroids A, D, and E.

Inhibited growth was observed in phytoplankton by Moore and Harris [1972]. Duke *et al.* [1970] noted inhibited shell growth in oysters, and Johansson *et al.* [1972] described various metabolic effects in brown trout.

Immunosuppression was found in mallard ducklings by Friend and Trainer [1970]. They proved to be less resistent to parasites. The same was observed in guinea pigs [Vos and de Roy, 1972] and coturnix quail [Dieter, 1974].

Impaired reproduction is the most obvious effect of PCBs in experimental studies. Again, in several trophic levels effects of PCBs have been demonstrated. Reduced reproduction was found in fish, crustaceans, birds and mammals such as mice, rats, and rhesus monkeys. Impaired reproduction of daphnias was found by Maki and Johnson [1975]. A reduced hatchability of eggs of brook trout was noted by Freeman and Idler [1973], whereas Kihlstroem *et al.* [1974] reported a decreased number of young in an experiment with zebra fish.

The effects on birds are generally shown by reduced egg production and hatchability [Dahlgren and Linder, 1971; Dahlgren *et al.*, 1972]. Examples of experimental PCB effects on reproduction in mammals are manifold. A prolonged oestrous cycle was found in mice by Orberg and Kihlstroem [1973] and Linder *et al.* [1974] observed fewer pups born in rats. A clear effect was demonstrated on a fish-eating mammal (mink) by Jensen *et al.* [1977] and den Boer [1983], who found a reduced reproductive rate, i.e. fewer pups born per female.

Mink are extremely sensitive to the effects of PCBs. In mink, reproduction is severely affected at a dietary level of only 5 mg/kg Aroclor 1254, and a slight effect was already noted at a dietary level of 1 mg/kg [Ringer *et al.*, 1972]. The effects on reproduction are probably caused by an enhanced breakdown of some steroid hormones.

At present, there is no evidence to indicate what is the no-effect level of PCBs.

It should be emphasised that some effects attributed to PCBs can also be caused by impurities in commercial PCB mixtures like dibenzofurans.

Epidemiological effects

In the literature, some altered functioning of processes in organisms has been linked to PCB contamination. However, because of lack of knowledge about the impact of other environmental conditions which might have changed, clear indications for the effects of PCBs only exist for higher levels in the food chains, i.e. birds and mammals.

(1) Birds

High mortality among cormorants in the coastal region of the Netherlands very probably was caused by PCB poisoning [Koeman *et al.*, 1973]. The same was noted in guillemots, of which massive numbers were found dead in the Irish Sea [Parslow and Jefferies, 1973]. Abnormal chickens were observed in common and roseate tern on Great Island (New York) by Hays and Risebrough [1972].

(2) Mink

In the mid-1960s, reproduction decreased in fish-eating mammals which were held in mink farms in North America. These animals have traditionally been fed fish originating from the Great Lakes. Experimental studies showed that the observed decreased reproduction was caused by PCB contamination of the fish [Aulerich *et al.*, 1973].

(3) Otters
The continuing decline of the otter in Sweden, particularly in the coastal areas, is at least partly ascribed to PCB poisoning. This is based on the fact that analyses of PCB content of muscles taken from otters from the Baltic revealed values ranging from 140 to 300 mg/kg [Sandegren *et al.*, 1980], which exceeds the level of 50 mg/kg where the fecundity of the closely related mink was strongly reduced [Jensen *et al.*, 1977].

(4) Sea lions
Sea lions in Southern California exhibited a high rate of premature births. Comparison of PCB and DDE residue levels in blubber of females who produced full-term pups, appeared to be 6.6 and 8.0 times lower, respectively, than those found in females with premature births. However, the effects of PCBs could not be separated from those of DDE and some pathogens [DeLong *et al.*, 1973].

(5) Belugas
The St Lawrence beluga (*Delphinapterus leucas*) population has dramatically declined because of hunting. High levels of both PCBs and total DDT (up to 576 and 225 mg/kg on wet weight basis) have been found in blubber tissue. It is suggested that these organochlorines have led to the observed pathological lesions and the physiological and reproductive disorders, preventing the population from recovering [Martineau *et al.*, 1985].

(6, 7) Baltic seals and harbour seals in the Dutch Waddenzee: Populations of ringed, grey, and harbour seals in the Baltic, and harbour seals in the Dutch Waddenzee have declined sharply. Effects of pollutants (i.e. PCBs) on impaired reproduction will be discussed in detail later on.

Implications for intercomparison and transplantation/transposal

Specific bio-effects

To assess the effects of PCBs on the marine ecosystem requires knowledge of specific bio-effects caused by that group of chemicals. A bio-effect should give a marked reproducible change and be related to a physiological process in a given organism, hence indicating a specific response to a single xenobiotic. For a range of contaminants it has been proven possible to develop, under laboratory conditions, toxicity tests which often can be expressed in terms of acute mortality (described as LC_{50} values). Extrapolation of these tests to environmental conditions is extremely difficult. As for other chemicals, this procedure is complicated for PCBs by the fact that organisms are frequently exposed to several environmental contaminants simultaneously. These compounds may act in a synergistic or antagonistic manner, but quantitative data on potential interactions between different chemicals are scarce. Further, environmental conditions in estuaries especially fluctuate widely throughout the year as a result of changes in geochemical processes. Marine organisms can partly adapt to physical changes but the simultaneous occurrence of pollutants and environmental stress may prove to be more harmful [Nimmo and Bahner, 1974].

Analytical techniques and sampling

Assessment of the environmental effects of PCBs in the marine environment implies comparison of residue levels in water, sediment, and biota from different geographical regions. Reviews and quantitative comparisons of organochlorine residue levels have, for example, been made for various marine mammal species [Holden, 1978; Wagemann and Muir, 1981]. Sampling procedures and analytical techniques greatly influence the reliability of acquired data as has been discussed by Duinker *et al.* [1980], Duinker and Hillebrand [1983a] and Aguilar [1985]. In reviews this problem is often insufficiently referred to. The same holds for the representativeness of certain tissues due to incomplete knowledge of kinetics after exposure [Aguilar, 1985; Reijnders, 1986, 1987].

Therefore the procedures of comparison of data from diverse sources are considered to be inadequate for a comparative assessment of the degree of hazard PCBs pose to marine organisms.

Residue levels

The chemical complexity of this group of contaminants renders interpretation of residue levels difficult. Mere comparison of total PCB levels can lead to large errors because kinetics of PCBs—deposition and excretion—are markedly influenced by their physicochemical properties (e.g. Boon and Duinker, 1985). Unfavourable stereochemistry resulting from different patterns of chlorine substitution leads to decreased metabolisation [Tanabe *et al.*, 1981; Boon *et al.*, 1987]. Lipophilicity and the related log p values (p = partition coefficient between n-octanol and water) are also of importance for metabolism (e.g. Sigiura *et al.*, 1978; Hidaka *et al.*, 1984). Therefore PCB concentrations should be based on the determination of individual congeners.

Furthermore, most PCB residue levels are expressed on the basis of a unit weight of extractable fat. Lipid composition seems to be a key factor in the equilibrium partitioning during the deposition process [Schneider, 1982; Aguilar, 1985]. Aguilar suggests the use of triglyceride content as a basis of comparison, because lipophilic xenobiotics are essentially linked to triglycerides. However, in juvenile sole, the highest PCB concentrations were measured in winter when fish contained the lowest lipid levels and also the lowest fractions of triglycerides [Boon *et al.*, 1984]. A further complication is created by the existing stratification of lipids in blubber because, for example, in marine mammals, it will affect distribution of different PCB congeners in that tissue [Aguilar, 1985].

The physiological state of an organism can considerably affect its contaminant burden. Circumannual cycles in blubber deposition [Drescher *et al.*, 1977; Aguilar, 1985] and changes in diet [Tanabe *et al.*, 1984] will be reflected in contaminant burdens in marine mammals. The reproductive state of an animal will also condition tissue pollutant burden. In most marine mammals PCB levels are higher in mature males than in mature females. Females transfer contaminants to their fetus and via lactation to their offspring [Duinker and Hillebrand, 1979; Reijnders, 1980; Helle, 1981]. In areas with moderate or high levels of pollution, the difference between sexes becomes less pronounced [Martineau *et al.*, 1985] or is not observable any more [Olsson *et al.*, 1975; Helle, 1980a].

Higher or equal levels of PCBs in females than in males does not necessarily automatically implicate pollution-related reproductive failure [Aguilar, 1983].

Case studies

Baltic seals

Seal populations in the Baltic Sea have decreased rapidly during recent decades. Lowered reproduction is believed to be an important reason [Helle, 1980b]. Reproductive success has been severely affected by sterility as a consequence of pathological uterine occlusions [Helle, 1980a]. Between 50% and 60% of mature ringed and harbour seals and 40% of the grey seals exhibited these lesions [Helle *et al.*, 1976a; Olsson, 1977; Bergman *et al.*, 1981].

The Baltic is seriously polluted by several organochlorine compounds, including the DDT-family and PCBs. The respective levels are high in Baltic seals and are correlated with the observed reproductive disorders. It has been hypothesised that PCBs probably played a prominent role in the incidence of reproductive failures in the Baltic seals [Helle *et al.*, 1976a, 1976b; Bergman *et al.*, 1981]. This hypothesis has been strengthened by findings of Helle [1986], and Bergman and Olsson *et al.* [1975]. The outstanding pathological investigations of Bergman and Olsson give an excellent description of the disease complex of which PCBs and other organochlorines are a large part. These chemicals seem to interfere primarily with the endocrine system (e.g. immunosuppression and interrupted pregnancies).

What is the biological significance of these findings for the further existence of the Baltic seal populations? The highest average levels of PCBs in tissue of ringed seals from the Bothnian Bay were found in the second quarter of the 1970s and, at present, levels are roughly equal to those found ten years ago when levels were still rising [Helle, 1986]. The decreasing trend in PCB levels matches an identicial trend found in several fish populations along the Finnish coast [Paasivirta and Linko, 1980; Moilanen *et al.*, 1982]. The supposed decreasing levels of PCBs in general could lead to a possible recovery of the Baltic seal populations. However, if the pathological changes in female genital tracts are permanent, as seems to be the case, population simulations show that even in the best case, the population will nevertheless continue to decrease for a while before starting to recover very slowly [Helle, 1980b; Spence and Harwood, 1983].

Harbour seals in the Dutch Waddenzee

The population of harbour seal (*Phoca vitulina*) in the westernmost part of the Waddenzee has collapsed during the past few decades [Reijnders, 1976 and 1978]. Between 1950 and 1970 the population dropped from more than 3000 to less than 500 animals. Studies of population dynamics in different parts of the Waddenzee revealed that pup production was very low in the Dutch harbour seal population compared with the stable population in Schleswig Holstein, Germany [Reijnders *et al.*, 1981]. A comparative toxicological study [Reijnders, 1980] on the levels of heavy metals and chlorinated hydrocarbons in tissues of seals from the western and northern parts of the Waddenzee showed that only PCB-levels differed significantly. These differences were a reflection of the levels found in their prey species. A west-east trend in PCB levels in fish was found [den Boer, 1983]. The comprehensive studies by Duinker and Hillebrand [1979] and Duinker *et al.* [1982a, 1982b, 1984] provide a good insight to the route of transport of PCBs into the Waddenzee. They conclude that PCB pollution of the Waddenzee is predominantly caused by the River Rhine, which mainly affects the western (Dutch) part. Available experimental and epidemiological data on the ability of PCBs to interfere with mammalian reproduction led to the hypothesis that PCBs might be responsible for the low rate of reproduction in the Dutch seal population.

To test the detrimental effects of PCBs on seal production an experiment with captive seals was started at the Netherlands' Research Institute for Nature Management. Two groups of 12 harbour seals each were fed a diet containing different levels of pollutants. The average daily intake (over approximately two years) was 1.5 mg PCBs for group 1, receiving fish from the western part of the Waddenzee, and 0.22 mg for group 2, receiving fish from the Atlantic. Three males receiving Atlantic fish were alternated between both groups during the mating period. The reproductive success, shown in Table 1, was significantly lower in group 1 than in group 2 ($P<0.02$, Fisher's exact probability test). Blood samples were taken regularly for hormone analyses in order to detect whether hormonal regulation was affected and if so, in which phase of the reproductive cycle effects were most dramatic. The hormone profiles showed that the reproductive process was disrupted in the post ovulation phase, around the time of implantation. These findings corroborate with an experiment carried out simultaneously with mink [den Boer, 1983]. This was designed to test whether pure PCBs (Clophen A 60 and A 30 respectively) had the same effect on reproductive performance of American mink *Mustela vison* as PCB-polluted fish. The results showed that reproduction was inhibited at very low levels of PCB intake (25 μg/day). The effects of pure PCB were identical to those of the contaminated fish diet (from the western part of the Waddenzee). The precise mechanism, which might be the failing of the priming effect on the endometrium to appear, or a maternal rejection response, or impaired steroid binding, could however not be tested with the information available [Reijnders, 1986].

Table 1 Number of participating, ovulating, and pregnant seals in both experimental groups, during the season 1983–1984.

Group	1	2
No. of females	12	12
No. ovulating	12	12
No. pregnant	4	10

After the completion of the first experiment, group 2 was released into the North Sea and group 1 was fed 'clean' Atlantic fish for approximately 1.5 years. The experiment has just been completed and blood samples have yet to be analysed. Preliminary results indicate that the adverse effect of the polluted fish is reversible (Table 2).

These results show that the reproductive failure in harbour seals from the Dutch Waddenzee is related to feeding on fish from this polluted area. The available epidemiological and experimental data on effects and levels of PCBs in seals and mink fed on fish from this area suggest that these pollutants are the main cause of this failure.

Pollution by PCBs has large consequences for the further existence of a harbour seal population in the Dutch Waddenzee. In 1984, only 44% of the population consisted of animals born in the area itself [Reijnders, 1986]. Without influx from the German part of the Waddenzee and repatriation of orphaned seals by nursery stations the population would have become virtually extinct around 1984. Although the adverse effects of PCBs are probably reversible, this does not imply a quick recovery of the harbour seal population. Even if the discharge of PCBs into the Waddenzee would stop, PCBs will still reach the water column, fishes, and ultimately seals, by release from the sediment. Partitioning between the ambient water and the sediment will partly determine concentrations [Duinker *et al.*, 1983b]. At present it is not possible to predict how long it will take for PCB levels in seals to drop below the threshold at which reproduction is affected.

Summary

Abundant conclusive experimental evidence exists to demonstrate the noxious effects of PCBs on marine organisms. High residue levels of PCBs have been found especially in fish-eating animals in the higher trophic levels of marine communities.

Interpretation of the effects of PCBs in the marine environment is complicated because extensive knowledge upon metabolic and environmental synergisms is required and not yet available.

Only in a very few cases, where profound long-term population studies have been carried out, can environmental effects be attributed to PCBs.

Nevertheless, it can be concluded that, although more experimental work has to be carried out, PCBs show serious effects justifying great concern about the environmental impact of these pollutants.

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Table 2 Number of participating, ovulating, and pregnant seals receiving Waddenzee fish during the seasons 1982–1984 and Atlantic fish during 1984–1986.

	PCB-polluted diet	'Clean' diet	
Season	1982–84	1984–85	1985–86
No. of females	12	12	12
No. ovulating	12	12	12
No. pregnant	4	6	8

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