DDT and PCB in Marine Animals from Swedish Waters

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Analyses of pesticide residues in a wide range of marine organisms from the coastal waters of Sweden show that there is a marked contamination in the Baltic. There are signs of an increase in polychlorinated biphenyls (PCB) from north to south in this area. Exceptionally large amounts of residues were found in white tailed eagles from the archipelago of Stockholm.

CHLORINATED pesticides can spread in living matter, and they are now present all over the world. In 1966 (ref. 1), it was discovered that some previously unknown substances isolated in analyses of pesticide residues were polychlorinated biphenyls (PCB). In Sweden, they occur in the natural environment in the same amounts as the chlorinated pesticides[1-3]. Recently, PCB has been found in organisms in the Netherlands, Great Britain[3-5] and the United States[6]. PCB is used almost exclusively in industry, and it seems to be more persistent than DDT. We have assessed PCB and DDT (including metabolites) contamination in Swedish marine ecosystems by analysing 176 samples. The hydrographical conditions along the Swedish coasts are not uniform so the residues are classified according to the four regional subdivisions shown in Fig. 1. Few samples were taken from the Sound and so this area has been included in the Baltic proper.

The species studied were: (1) mussel (*Mytilus edulis*); (2) herring (*Clupea harengus*); (3) plaice (*Pleuronectes platessa*); (4) picked dogfish (*Squalus acanthias*); (5) cod (*Gadus morhua*); (6) salmon (*Salmo salar*); (7) grey seal (*Halichoerus grypus*); common seal (*Phoca vitulina*) and ringed seal (*Pusa hispida*); (8) guillemot (*Uria aalge*); (9) white tailed eagle (*Haliaeetus albicilla*); (10) eggs of white tailed eagle; (11) heron (*Ardea cinerea*). Three samples of separated fish oil were also analysed. Two were taken from the Norway pout (*Boreogadus esmarki*) and one from herrings from the northern part of the west coast. One sample (largely herring oil) was also extracted from fish from the southern Baltic proper. This was unseparated.

Homogenates of one or more mussels were analysed. In the fish, axial muscle tissue extracted from the dorsal side above the lateral line, approximately a third of the length from the tail was analysed. In nine out of eleven salmon, the samples had to be taken immediately behind the head (only heads were available). Most of the samples from the seals were taken from the blubber in the tail. In two seal pups from the Gulf of Finland, the samples were from the fore leg, and in one grey seal from the Baltic proper, the sample was liver. It is reported[3], however, that in seals the chlorinated hydrocarbon content in the fat is approximately constant throughout the different parts of the body. As a rule, the specimens, or parts of the specimens, were sent to the Swedish Museum of Natural History immediately after capture and were stored at $-20°$ C. Three seal tails from the archipelago of Stockholm and one from the Gulf of Bothnia were, however, kept by the hunter for six months in a non-heated room. In the homogenized eggs of the guillemot the fat varied from 3·6 to 11 per cent probably because homogenization before aliquotation was not quite perfect. Samples were taken from both the pectoral muscle and brain of the white tailed eagle. All the eagles were found dead and in some cases in a state of decay, and all the eggs were either addled, rotten or dry. In the heron, pectoral muscle only was analysed.

DDE was the principal metabolite of DDT found.

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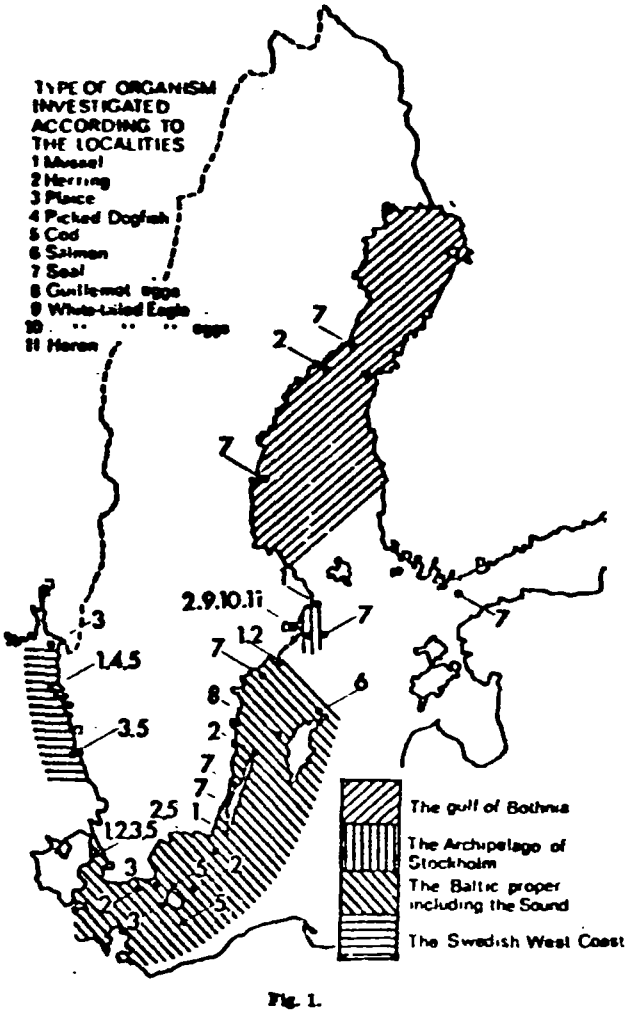


Fig. 1.

DDD was found in small concentrations, not usually more than 10 per cent of the ε DDT, in most samples. (ε DDT stands for the sum of DDT and DDE—DDE was multiplied by 1·11 to correspond to the original DDT.) For seals from the archipelago of Stockholm and for salmon from the Baltic proper which had relatively large amounts of DDD, ε DDT stands for the sum of DDT, DDE and DDD.

We analysed all the samples using gas chromatography and electron capture detection. Confirmations were carried out with double column systems combined with treatments with sulphuric acid and potassium hydroxide[7].

We found that the principal PCB components were PCB numbers 7–14 (low number is equal to a low chlorination degree), but the mussels also contained the lower numbers (the PCB composition being similar to clophen A 50). The PCB amounts given in Table 1 are, nevertheless, the sum of all PCB components found in the samples although the relative amounts of the different numbers vary. The results suggest that the low PCB numbers are metabolized or excreted faster than the higher numbers, so that there is an increase of the latter as it passes through a food chain.

An attempt to quantitate PCB has been made in Sweden[7], but because the method is still rather rough the values may be correct only within a factor of 2. The method is indirect because all the PCB components have not been isolated yet, and it is based on a combination of mass spectrometry, micro-colorimetric and electron capture detection. For each twenty samples a blank on the solvent and a residue-free plaice sample with added pesticides (lindane, aldrin, *pp'*DDE, dieldrin, *pp'*-DDD and *pp'*-DDT) were also analysed. Recovery was about 80 per cent, but as normal in residue analysis[8] we made no correction.

The mean figures in Table 1 are calculated from the average value of each locality within the four regions. The residue values are based both on fresh tissue and on hexane extractable fat. The figures for fresh tissue are comparable with reported values from other countries. We believe, however, that the fat values are of particular interest from an ecological point of view. Fat is important in the transport of energy and as a carrier of the residues between the species in the food chain. Furthermore, there is a great seasonal variation in fat content within the populations. The difference between the greatest and the smallest residue level in the extractable fat of individual, of one species at a given time, in a given place, is less than in fresh tissue. The fat content of herring muscle, for example, varied from about 1 per cent in the spring to about 10 per cent in the autumn. Our discussion will therefore be based largely on residue levels in extractable fat.

Mussels from the west coast of Sweden have lower average values of ε DDT and PCB than from the Baltic proper and the archipelago of Stockholm. The level of ε DDT in mussels from the Dutch, West German and British coasts[8,9] has an upper limit similar to that in mussels from the Baltic proper and the archipelago of Stockholm.

DDT contamination in individual plaice, cod and in fish oil is greater in the Baltic proper than off the west coast of Sweden, and there is little or no overlapping of the residue values. In the case of PCB, a plaice from the very polluted Idefjord in the west coast increases the average value for the west coast samples from 0·69 to 3 p.p.m. Other individual samples of fish from the west coast contain PCB residues of the same magnitude as residues in specimens from the Baltic. This may be the result of coastal contamination. The picked dogfish, which lives more in the open sea, seems to have the same amount of ε DDT, but a smaller amount of PCB than other fish from the west coast of Sweden.

The samples of herring from the Baltic proper, the archipelago of Stockholm and the Gulf of Bothnia had large residues of ε DDT. No herring from the west coast of Sweden has been studied. Recently we analysed eight herrings from the west coast. On fat basis the mean values were: ε DDT, 1·9; DDT, 0·86; and PCB, 0·75, in good agreement with the fish oil values (see Table 1). Juvenile herring from the Dutch Wadden Sea (coastal waters)[8,9] contained DDE residues in fresh tissue with an upper limit of the same magnitude as in herring from the Baltic area.

Residues of PCB and ε DDT in one sample of fish oil from the southern Baltic proper were about five to ten times greater than the three samples from the west coast of Sweden. The figures for the latter are similar. This is in accord with residues in fat in fish from the respective areas.

One herring from the Sound had a much larger content of PCB (23 p.p.m.) than the herring from the rest of the Baltic area. This increased the average value in this area from 4·6 to 6·8 p.p.m. Cod and plaice from the Sound also had a larger PCB content than specimens from the Baltic proper and off the west coast. This suggests that there is local contamination of PCB in the Sound. Furthermore, the mean levels of PCB noted in herring were smaller in the Gulf of Bothnia than in the rest of the Baltic.

The vagrant Baltic salmon had about ten times more chlorinated hydrocarbons residue than a sample reported from Great Britain[8].

The seals from the Baltic proper, the archipelago of Stockholm and the Gulf of Bothnia contained large amounts of residues of both ε DDT and PCB, about ten times greater than amounts found in Great Britain[8], Canada[8] and the Netherlands[10]. One specimen from

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the archipelago of Stockholm had 310 p.p.m. in fat, increasing the mean value for this part of the coast from 117 p.p.m. to 170 p.p.m. The levels of PCB decrease from south to north. This fact, together with lower average values of PCB in herring from the Gulf of Bothnia, indicates decreasing PCB contamination from south to north in the Baltic. Seal pups, one week old, also contained large amounts of ε DDT and PCB residues, and seal milk from the stomach of one of these pups had the same level.

Guillemot eggs from the Baltic proper provided additional information on the contamination of the Baltic. The means of ε DDT and PCB in eggs (wet weight) were 80 p.p.m. and 16 p.p.m. respectively, which for ε DDT is about ten times the levels reported from Great Britain[9,11-13]. The Baltic levels, both for ε DDT and PCB, are about four to five times greater than levels reported from the west coast of the United States[6], if the figures for residues in fat are compared. In guillemot eggs from the Baltic, as much as 87 per cent of ε DDT was DDE.

The population of white tailed eagles in Sweden seems to be on the decline. Only about two of ten pairs nesting in the archipelago succeeded in hatching during the past few years (E. Larsson, personal communication). All specimens of white tailed eagle studied up to the present contained at least three accumulating substances known to be toxic. These are DDT metabolites, PCB and mercury compounds[14]. Figures for four birds are reported in Table 1. A fifth specimen is not included because the fat content was not determined, but the pectoral muscle and brain tissue of this bird (wet weight) contained 190 and 39 p.p.m. respectively of DDT (as DDE), and 230 and 24 p.p.m. of PCB. Two specimens from northern Sweden, one of which died by accident, contained 2·6 p.p.m. (1·8–3·4) of DDT (as DDE) and 2·9 p.p.m. (1·8–3·9) of PCB in muscle tissue, the second having 1·8 p.p.m. DDT (as DDE) and 1·2 p.p.m. PCB in brain tissue. The figures from the archipelago of Stockholm are thus a hundred times larger, suggesting that there is a strong contamination in this area.

It is well known that residues of chlorinated hydrocarbons tend to be greater in the higher trophic levels than in the lower. Some simplified food chain relations can be deduced from the figures in Table 1. These are fish to seal, fish to guillemot, fish to heron and fish to white tailed eagle. In all cases, the increase in residues from prey to predator is at least ten-fold in both whole tissue and fat. In the eagle and heron, the increase is up to 100 times; but for both of these species some important food organisms are missing in this investigation. There are indications (Table 1) that the percentage of DDE and of ε DDT increases as it progresses from lower to higher trophic levels. This may be the result of decomposition of DDT within the organisms. In all the samples from birds, the residues are almost exclusively DDE, while in seals and fishes there is up to 50 per cent of DDT. This difference may be the result of differences in metabolism[15].

As previously mentioned, the seals from the archipelago

Table 1. CONCENTRATION OF ORGANOCHLORINE COMPOUNDS IN SWEDISH MARINE ORGANISMS 1966–68

	No. in sample	p.p.m. in fat			p.p.m. in fresh tissue			Per cent fat
		εDDT	DDT	PCB	εDDT	DDT	PCB	
Swedish West Coast								
Mussel	17	1	0·6	2	0·02	0·007	0·064	1·3
Oct. 1966		(0·4–5)	(0·3–1·3)	(0·5–7·0)	(0·005–0·04)	(0·002–0·03)	(0·011–0·23)	(0·66–2·6)
Dec. 1967								
Plaice	3	1		5	0·006	0·004	0·021	0·5
Sept. 1966		(0·6–2)	n.a.	(0·4–14)	(0·003–0·009)	(trace–0·006)	(0·002–0·056)	(0·4–0·5)
Cod	4	1		7·3	0·005	0·003	0·019	0·20
Sept. 1967		(0·6–2)	n.a.	(1·8–16)	(0·001–0·006)	(n.d.–0·006)	(0·006–0·030)	(0·19–0·34)
Pickled dogfish	7	1·5	0·01	1·5	0·15	0·001	0·15	9·6
Aug. 1968		(0·28–3·9)	(0·15–2·3)	(0·81–2·4)	(0·028–0·33)	(0·015–0·21)	(0·084–0·20)	(6·7–14)
Fish oil	3	2·1	1·2	0·74				100
Oct. 1968		(1·5–2·6)	(0·83–1·4)	(0·54–1·0)				
Baltic Sea proper incl. the Sound								
Mussel	40	6	1·8	4·3	0·02	0·02	0·03	0·92
Oct. 1966, Dec. 1966		(0·9–10)	(0·5–2·9)	(1·9–8·6)	(0·009–0·07)	(0·003–0·023)	(0·006–0·057)	(0·46–1·6)
Dec. 1967, Jan. 1968								
Herring	18	17	9·7	6·8	0·68	0·40	0·27	4·4
April, Sept. 1966–68		(4·1–37)	(1·5–21)	(0·5–23)	(0·003–2·3)	(0·012–1·3)	(0·009–1·0)	(0·7–12)
Plaice	6	2·7	2·1	2·7	0·018	0·013	0·017	0·65
Sept. 1967		(1·4–7·8)	(0·6–7·2)	(1·7–4·8)	(0·008–0·036)	(0·003–0·028)	(0·010–0·032)	(0·58–0·71)
Cod	5	19	9·8	11	0·063	0·032	0·033	0·32
Sept. 1967		(12–31)	(3·5–19)	(3·2–26)	(0·027–0·11)	(0·004–0·066)	(0·012–0·067)	(0·21–0·44)
Salmon*	11	31	14	2·9	3·4	1·5	0·30	11·0
Autumn 1966		(20–53)	(7·7–26)	(1·1–8·2)	(0·26–7·1)	(0·095–3·1)	(0·014–0·54)	(1·2–20)
Fish oil	1	16	7·3	3·5				100
Oct. 1966								
Seal (grey) liver	1	96	41	44	3·9	1·7	1·8	4·1
Seal (common and grey)	2	130	62	30	96	38	18	82
Sept., Nov. 1966		(110–150)	(57–66)	(18–43)	(58–74)	(31–22)	(8·5–21)	(46–55)
Eggs from guillemot	9	570	20	250	46	1·2	18	7·8
May 1965		(300–790)	(7·5–36)	(140–360)	(20–61)	(0·7–2·3)	(7·9–21)	(3·6–11)
The Archipelago of Stockholm								
Mussel	15	3	1	6·2	0·04	0·02	0·037	1·1
Oct. 1966, Dec. 1967		(1–4·7)	(1–1·8)	(3·4–7·0)	(0·01–0·061)	(0·01–0·024)	(0·022–0·044)	(0·94–1·3)
Herring	4	7·7	3·9	5·1	0·23	0·11	0·17	2·8
May 1965		(4·3–11)	(2·0–5·3)	(3·3–6·5)	(0·094–0·20)	(0·044–0·15)	(0·073–0·23)	(2·2–2·8)
Seal (grey)*	3	170	17	30	26	4·2	6·1	23·1
May 1966		(97–310)	(11–21)	(16–56)	(25–36)	(2·4–6·0)	(5·7–6·4)	(11·5–37·5)
White tailed eagle								
March–June 1965–66	4	25,000		14,000	230		190	1·5
Pectoral muscle		(18,000–36,000)	n.d.	(8,400–17,000)	(200–400)	n.d.	(150–240)	(0·9–2·0)
Brain	3	1,900		910	160		47	5·1
		(1,700–2,100)	n.d.	(480–1,500)	(90–110)	n.d.	(29–70)	(4·6–6·0)
Eggs from white tailed eagle	5	1,800		540				5·6
May–June 1966		(610–1,900)	n.d.	(250–880)	n.e.	n.e.	n.e.	(3·4–9·1)
Heron								
April 1967	1	14,000	n.d.	9,400	71	n.d.	48	0·51
Gulf of Bothnia								
Herring	4	6·2	3·5	1·5	0·26	0·14	0·065	4·4
		(5·2–8·1)	(2·9–4·5)	(0·93–2·9)	(0·15–0·42)	(0·091–0·21)	(0·028–0·091)	(2·1–6·8)
Seal (ringed)	2	120	56	13	63	30	6·5	54
May–Oct. 1966		(110–130)	(54–57)	(9·7–16)	(56–68)	(28–31)	(5·0–8·5)	(52–55)
Gulf of Finland								
Seal pup (grey)	2	42	23	8·5	25	14	3·0	60
March 1968		(41–43)	(22–23)	(8·0–7·0)	(24–26)	(13–14)	(3·4–4·4)	(56–63)
Seal milk								
March 1966	1	36	21	4·5	11	6·5	1·4	31

* εDDT stands for DDT + DDE + DDD. For salmon and seal, there was respectively 41 per cent DDD in εDDT, the mean figure being 17 per cent.
n.e., Not estimated. n.d., Not detected.

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of Stockholm, stored in a non-heated room for six months, had a greater proportion of DDD (up to 40 per cent of ε DDT), and simultaneously smaller levels of DDT itself, than all other seals. This could have been caused by post mortem metabolism by microorganisms[20]. On the other hand, the ringed seal from the Gulf of Bothnia, stored in the same way as far as we know, had only about 5 per cent of DDD, but this low figure may be corrected with the uncertainty of DDD estimation at the time when the other seals were analysed. This difference between the archipelago seals and the other seals, however, could be related to the fact that the large amount of pollution and the land water relation in the archipelago of Stockholm produce a higher microbiological activity in the water, transforming[17] DDT to DDD and possibly DDE.

Little is known about the toxicity of PCB in the levels found by us. PCB has, however, been proved to cause pathological changes in laboratory animals[19], and was found to be among the ten most potent chemicals among a hundred tested by injection in eggs[19]. Some teratogenic effects have been noted[16]. Effects of DDT, DDE and PCB on hepatic enzymes which increase the metabolism of progesterone, testosterone and oestradiol have also been published[6]. It is worth noting that bald eagles (*Haliocetus leucocephalus*), fed a diet containing DDT, had between 58–86 p.p.m. DDT and DDD in their brains when they died[20]. Approximately the same values were found in robins and house-sparrows[21]. Brain tissue in white tailed eagles from the Stockholm area contained on average 100 p.p.m. (fresh tissue) of DDE. DDE, on the other hand, is less acutely toxic than DDT itself. The greatest value in North American bald eagles collected in the field in 1965 was 118 p.p.m. (mainly DDE) in muscle tissues[21]. The greatest level for sampled wild white tailed eagles from Sweden is 400 p.p.m. (mainly DDE) and the mean was 330 p.p.m. in muscle tissue (wet weight). Nothing is yet known about the PCB content in bald eagles from North America, but in eagles from our archipelago the mean value in muscle was 190 p.p.m. (brain 47 p.p.m.) and the greatest figure was 240 p.p.m. (brain 70 p.p.m.).

Neither the fresh tissue nor the original fat values were estimated in the addled eggs from white tailed eagles. Sometimes the water content was as low as 8 per cent (normally 70–75 per cent in fresh hen eggs). The values based on the fat content of these eggs are probably more reliable and we only report such figures. The levels, although high, were only 1/25 of those of the eagle fat. The residues of ε DDT and PCB in the guillemot eggs were about 50 per cent lower than those in the white tailed eagle eggs. The guillemots are vagrant in winter but most of the central Baltic population is believed to remain in the Baltic area the year round. It is not known whether the population is decreasing, but no drastic decline has been reported.

All species mentioned here (except mussel) are more or less vagrant, thereby giving a picture of the situation in a large water area. A relatively few analyses on a vagrant animal may not give a very accurate picture, but when different species are shown to contain more residues in the Baltic than in the North Sea and the Atlantic, the situation is more convincing.

In the Baltic proper, the Gulf of Bothnia and in the archipelago of Stockholm, the levels of chlorinated hydrocarbons are approximately ten times greater than the reported sparse figures for comparable species in the North Sea area and the Atlantic. This may be caused by several factors. The Baltic is surrounded by large areas of land and the water volume is comparatively small. Furthermore, the water exchange with the North Sea is very limited as a consequence of the thresholds (18 and 8 m deep) and the narrowness of the Danish sounds. Residues brought into the Baltic from the coasts and from air pollution will accumulate here to a great extent. This development is no doubt favoured by a generally low microbiological activity because of very low temperatures of the intermediate Baltic "winter water" during most of the year, and also of the deep water layers in the northern areas. As a consequence of the rather cold water, this is also largely true for the upper surface layer.

The brackish water character of the Baltic also favours a concentration of residues in living organisms. The substances in question are probably more readily transported in a horizontal direction by living matter from fresh water to the low salinity surface water (frequently 5–6 per mille or less) of the archipelagos and along the Baltic coasts. In the North Sea, the discrepancy between the salt seawater and fresh water especially when combined with the effect of the tidal movements can result in a faster decomposition of freshwater organisms and of organic matter, including the chlorinated hydrocarbons. The increased and more immediate contact here with the microbiological activity located in the bottom material may be important. Furthermore, there is in the Baltic proper a pronounced stratification of the water. A distinct halocline delimits the deep water of higher salinity upwards (usually in 50–70 m depth). In the warm season, the uppermost surface layer is separated from the "winter water" by a very marked thermocline (usually in 15–20 m depth, sometimes supported by a secondary haloline). As a result of these two discontinuity layers and, especially in coastal areas, of the absence of marked tidal currents, the vertical water exchange and movements are more or less blocked. The contact of sinking organic matter with the bottom material is thus to a great extent delayed. In the discontinuity layers of the Baltic Sea, living and dead plankton and other organic matter are concentrated, as a consequence of differences in the specific gravity of the water. The layers mentioned are important feeding areas for plankton and pelagic fish such as herring and sprat. In the Sound, in the archipelago of Stockholm and in the Idefjord, local sources of pollution can be important as the residue levels indicate.

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