

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^{2,3}. Recently, PCB has been found in organisms in the Netherlands, Great Britain⁴⁻⁶ and the United States⁷. 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) piked 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 aemarki*) 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 reported⁸, 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.

DSW 029722

STLCOPCB4013684

the archipelago of Stockholm had 310 p.p.m. in fat, exceeding the mean value for this part of the coast from 177 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 α -DDT and PCB residues, and seal milk from the stomach of one of these pups had the same level.

Gullmote eggs from the Baltic proper provided additional information on the contamination of the Baltic. The means of α -DDT and PCB in eggs (wet weights) were 40 p.p.m. and 18 p.p.m. respectively, which for α -DDT is about ten times the levels reported from Great Britain¹¹⁻¹². The Baltic levels, both for α -DDT and PCB, are about four to five times greater than levels reported from the west coast of the United States¹³, if the figures for residues in fat are compared. In gullmote eggs from the Baltic, as much as 87 per cent of α -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 of Stockholm 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 compounds¹⁴. 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 2.0 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 gullmote, fish to herring and fish to white tailed eagles. 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 herring, 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 α -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 metabolism¹⁴.

As previously mentioned, the seals from the archipelago

Table 1. CONCENTRATION OF ORGANOCHLORINE COMPOUNDS IN SWEDISH MARINE ORGANISMS 1956-58

	No. in sample	α -DDT	p.p.m. in fat DDT	PCB	α -DDT	p.p.m. in fresh tissue DDT	PCB	Per cent fat
Swedish West Coast								
Muskel	17	1 (0.4-5)	0.6 (0.2-1.8)	2 (0.5-7.0)	0.02 (0.004-0.04)	0.007 (0.002-0.08)	0.084 (0.011-0.33)	1.3 (0.66-2.6)
Oct. 1956								
Dec. 1957								
Plaice	3	1 (0.3-2)	n.d.	5 (0.4-14)	0.006 (0.003-0.009)	0.004 (trace-0.006)	0.021 (0.002-0.056)	0.8 (0.4-0.5)
Sept. 1956								
Cod	4	1 (0.4-2)	n.d.	7.3 (1.0-16)	0.005 (0.001-0.006)	0.003 (n.d.-0.006)	0.019 (0.006-0.030)	0.30 (0.19-0.34)
Sept. 1957								
Picked dogfish	7	1.8 (0.23-3.9)	0.91 (0.15-2.3)	1.8 (0.51-2.4)	0.15 (0.023-0.33)	0.081 (0.015-0.21)	0.15 (0.054-0.30)	9.0 (6.7-14)
Aug. 1956								
Fish oil	3	2.1 (1.5-2.6)	1.2 (0.8-1.4)	0.74 (0.44-1.0)				100
Oct. 1958								
Baltic Sea proper incl. the Sound								
Muskel	40	6 (0.9-10)	1.8 (0.5-2.9)	4.3 (1.9-8.5)	0.03 (0.009-0.07)	0.02 (0.003-0.023)	0.03 (0.008-0.067)	0.02 (0.48-1.6)
Oct. 1956, Dec. 1956								
Dec. 1957, Jan. 1958								
Herring	18	17 (4.1-37)	9.7 (1.4-21)	6.8 (0.5-23)	0.08 (0.003-2.3)	0.40 (0.012-1.3)	0.27 (0.009-1.0)	4.4 (0.7-12)
April, Sept. 1956-58								
Plaice	9	2.7 (1.4-7.2)	2.1 (0.4-7.2)	2.7 (1.7-4.8)	0.018 (0.006-0.034)	0.013 (0.003-0.029)	0.017 (0.010-0.033)	0.05 (0.58-0.71)
Sept. 1957								
Cod	5	19 (13-31)	9.8 (3.5-19)	11 (3.2-30)	0.063 (0.027-0.11)	0.032 (0.008-0.068)	0.033 (0.012-0.067)	0.32 (0.23-0.44)
Sept. 1957								
Salmon*	11	21 (80-63)	14 (7.7-30)	2.9 (1.1-8.2)	3.4 (0.28-7.1)	1.5 (0.06-3.1)	0.30 (0.014-0.54)	11.0 (1.2-20)
Autumn 1958								
Fish oil	1	16 (10-24)	7.3 (3.8-14)	3.6 (1.4-9.0)				100
Oct. 1958								
Seal (grey) liver	1	96	41	44	3.0	1.7	1.8	4.1
Seal (common and grey)	2	130 (110-160)	43 (27-68)	30 (16-43)	55 (58-74)	22 (21-32)	15 (8.5-21)	52 (48-55)
Sept., Nov. 1958								
Eggs from gullmote	9	870 (900-790)	250 (7.3-36)	250 (140-390)	46 (20-81)	1.2 (0.7-2.3)	16 (7.4-21)	7.0 (3.6-11)
May 1958								
The Archipelago of Stockholm								
Muskel	15	3 (1.4-7)	1 (1-1.9)	5.2 (3.4-7.0)	0.04 (0.01-0.061)	0.02 (0.01-0.024)	0.037 (0.032-0.044)	1.1 (0.94-1.3)
Oct. 1958, Dec. 1957								
Herring	4	7.7 (4.3-11)	3.0 (2.0-5.2)	6.1 (3.3-8.5)	0.23 (0.04-0.90)	0.11 (0.044-0.18)	0.17 (0.078-0.23)	2.6 (2.2-2.8)
May 1955								
Seal (grey)*	3	170 (87-310)	17 (11-21)	30 (16-56)	36 (25-54)	4.2 (3.4-6.8)	6.1 (5.7-8.4)	27.1 (11.5-37.3)
May 1958								
White tailed eagle	4	25,000 (18,000-36,000)	n.d.	14,000 (8,400-17,000)	230 (200-400)	n.d.	100 (150-240)	1.5 (0.9-2.0)
March-June 1955-56								
Pectoral muscle	3	1,000 (1,700-2,100)	n.d.	910 (480-1,500)	100 (90-110)	n.d.	47 (29-70)	6.1 (4.6-8.0)
Brain								
Eggs from white tailed eagle	5	1,000 (810-1,600)	n.d.	540 (250-800)	n.e.	n.e.	n.e.	5.0 (3.4-9.1)
May-June 1956								
Merlon	1	14,000	n.d.	2,400	71	n.d.	43	0.51
April 1957								
Gulf of Bothnia								
Herring	4	8.2 (5.2-8.1)	3.5 (2.0-4.8)	1.5 (0.92-2.0)	0.28 (0.15-0.42)	0.14 (0.06-0.31)	0.085 (0.026-0.091)	4.4 (2.1-6.8)
May-Oct. 1958								
Seal (ringed)	2	180 (110-180)	54 (34-87)	13 (9.7-16)	63 (48-82)	30 (28-31)	6.8 (5.0-8.5)	54 (52-55)
Gulf of Finland								
Seal pup (grey)	2	42 (41-43)	23 (22-23)	6.5 (6.0-7.0)	25 (24-26)	14 (13-14)	3.9 (3.4-4.4)	60 (56-63)
March 1958								
Seal milk	1	26	21	4.5	11	6.5	1.4	31
March 1958								

* α -DDT stands for DDT + DDE + DDD. For salmon and seal, there was respectively 41 per cent DDD in α -DDT, the mean figure being 17 per cent.

n.e., Not estimated. n.d., Not detected.

DSW 029724

STLCPCB4013686

of Stockholm, stored in a non-heated room for six months, had a greater proportion of DDD (up to 40 per cent of Σ DDT), and simultaneously smaller levels of DDT itself, than all other seals. This could have been caused by post mortem metabolism by microorganisms¹². 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, transforming¹³ 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 animals¹⁴, and was found to be among the ten most potent chemicals among a hundred tested by injection in eggs¹⁵. Some teratogenic effects have been noted¹⁶. Effects of DDT, DDE and PCB on hepatic enzymes which increase the metabolism of progesterone, testosterone and oestradiol have also been published¹⁷. It is worth noting that bald eagles (*Haliaeetus leucocephalus*), fed a diet containing DDT, had between 58-80 p.p.m. DDT and DDD in their brains when they died¹⁸. Approximately the same values were found in robins and house-sparrows¹⁹. 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 1905 was 118 p.p.m. (mainly DDE) in muscle tissues²⁰. 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 added 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 Σ 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 bottom "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 winter, 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 5-8 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 halocline). 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.

We thank Mrs Kerstin Widmark, Mr Bruno Nucci, Mrs Elsa Nucci, Miss Gunnel Blomkvist and Miss Margareta Östberg at the Institute of Analytical Chemistry, the University of Stockholm. We also thank Mr Werner Berg and Mr Bengt Wallin at the Museum of Natural History, Stockholm, for their help in this work.

Received May 12, 1963.

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