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POLYCHLORINATED BIPHENYLS AND CHLORORGANIC PESTICIDES. ASSESSMENT OF HEALTH RISK ASSOCIATED WITH THE CONSUMPTION OF SEAFOOD

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Abstract. Polychlorinated biphenyls and chlororganic pesticides were used as tracers for nonpolar organic contaminants cycling in the Baltic Sea. A comparison of chlororganic contents of herring in different areas of the Baltic Sea suggests the northern part of the Gulf of Riga as the reference area for the Baltic Sea for the distribution of chlororganic compounds. Herring were collected in the Estonian coastal sea area. The studied organochlorine components were α-HCH; HCB; β-HCH; γ-HCH; δ-HCH; aldrin; p,p'DDE; p,p'DDD; p,p'DDT and polychlorinated biphenyl isomers IUPAC 49; 52; 101; 105; 118; 138; 153, and 180. The summary DDT (DDE+DDD+DDT) concentrations in the muscle tissue of 2-year-old herring varied from 102–585 ng g^{-1} (extractable fat) and for two most important polychlorinated biphenyl isomers, IUPAC 138 and 153, from 28 to 237 ng g^{-1} and from 47 to 422 ng g^{-1} (extractable fat), respectively.

The levels of organochlorines in the organism of the Baltic herring in the coastal areas of Estonia do not constitute any human health risk.

Key words: Baltic herring, PCB, pesticides, Baltic Sea, health risk.

INTRODUCTION

Environmentally hazardous substances, such as polychlorinated biphenyls (PCB) and chlororganic pesticides (COP), are acutely toxic, persistent, and bioaccumulative (i.e. they become concentrated in food chains to reach toxic levels).

Following the decision of the Baltic Marine Environment Protection Commission, a systematic monitoring of harmful substances in selected species of the Baltic Sea biota was carried out in Estonia (Роотс, 1992; Roots, 1994, 1995; Roots & Aps, 1993). In studying PCB compounds, one has to consider both the partition coefficient (between 1-octanol and water) and the stereochemistry of compounds. The determination of separate PCB congeners is necessary because congeners vary differ in their toxic properties and their half-life in organisms can considerably.

The species chosen as test organisms and the sampling procedures recommended for monitoring harmful substances in biota are intended to provide a picture of the levels of harmful substances in the studied organisms and to reveal trends in their levels over time. HELCOM has selected herring (*Clupea harengus membras* L.) and cod (*Gadus morhua* L.) as study organisms in the Baltic Monitoring Program (BMP), because:

– they can be caught in all parts of the Baltic (this is important for comparison);

– they are easy to collect;

– they are of suitable size for preanalytical sample treatment;

– their biology is fairly well known;

– they are important commercial species.

The monitoring program is based on measurements of obligatory (mandatory) and tentative determinants (Haarich, 1994). We selected Baltic herring and the toxicants presented by Haarich (1994) as a base for our studies. In 1994, Estonian fishermen caught 45 416 t of fish from the Baltic Sea. From that amount herring made up 34 494 t and sprat (*Sprattus sprattus* (L.)) 9079 t. In connection with the decrease of salinity in the northern part of the Baltic Sea, the catches of cod have noticeably decreased, amounting to 227 t, that is 0.7% from the overall amount of the fish caught (Estonian Environment 1994, 1995).

Trends of environmental contaminants have to be studied in defined areas (the same place every year) (Роотс & Пейкре, 1978; Haahti, 1994; Roots, 1995). An important role is played by the time and place of catching when different regions are compared (Роотс & Пейкре, 1978; Роотс, 1992). The prespawning age classes of herring seem to be local or regional species at young age (Parmanne, 1990; Bignert et al., 1993). In all probability, samples of younger fish originate from the same stock and are thus representatives of the local environment (Roots & Lukki, 1990, 1994; Bignert et al., 1993; Haarich, 1994).

There were two main reasons for selecting the Estonian coastal zone as a sampling area:

– the importance of fish in the food of the people living in the coastal areas is relatively big;

– air pollution from local sources inside Estonian boundaries is small.

Appropriate regions would be the northern part of the Gulf of Riga (Table 1) and the western part of the Gulf of Finland (Roots & Aps, 1993). The Gulf of Riga was chosen as a research area because the accumulation of chlorinated hydrocarbons in the Baltic herring in this area has been insufficiently studied so far (Roots, 1994, 1995, 1996).

Table 1a

Concentration of chlororganic pesticides in the muscle tissue of the Baltic herring (Gulf of Riga)

No.	Length, cm		Weight, g	Age	Sex		Matu-rity	Fat %	Concentration, ng g^{-1} (fat)									
	L	L_s			♂	♀			α-HCH	HCB	β-HCH	γ-HCH	δ-HCH	Aldrin	p,p'DDE	p,p'DDD	p,p'DDT	sum. DDT
1	12.3	10.3	10.3	2	+		III	3.8	16.3	17.7	10.0	6.7			99.3	81.4	10.6	191.3
2	13.6	12.2	12.3	2		+	IV	2.3	2.0	42.0		0.8			155.2	189.8	46.5	391.5
3	12.4	11.3	10.0	2		+	IV	2.6							32.0	58.8	24.4	115.2
4	13.0	11.4	12.2	2		+	IV	3.9	4.0	15.5	29.9	8.1	2.6		39.8	216.6	73.3	329.7
5	12.2	10.8	9.1	2	+		III	3.2		15.4		4.3			195.0	126.1	64.6	385.7
6	13.6	12.3	12.8	2		+	III	0.8	1.8			9.1			170.0		48.7	218.7
7	11.7	10.4	9.0	2	+		III	0.9	2.9	8.9		19.9			363.0		141.2	504.2
8	13.3	11.0	12.5	2	+		IV	1.7	19.4				1.2		168.8	21.7		190.5
9	12.1	10.7	9.6	2		+	IV	1.6	6.3	6.2		11.2			77.9		24.4	102.3
10	12.1	10.7	9.4	2		+	IV	4.2	0.6	6.4		3.0			81.6		25.8	107.4
11	13.4	12.2	12.2	2	+		III	1.2	9.3	23.7	9.9	11.4		2.5	186.7		50.6	237.3
12	12.0	10.6	9.4	2	+		IV	1.3	4.0	9.9	10.6	6.4			177.3	7.8	80.1	265.2
13	11.6	10.3	8.1	2		+	III	3.5	4.9	12.1		6.5			166.4	21.5	61.8	249.7
14	12.6	11.3	11.1	2		+	IV	1.2	1.3	19.4		2.9		1.1	235.7		58.4	294.1
15	12.9	11.4	12.0	2		+	III	2.2							113.9		44.0	157.9
16	12.3	11.0	10.3	2	+		III	3.8	26.8	11.9	7.8	26.1		2.5	88.0	110.5	47.2	245.7
17	13.6	12.2	12.3	2		+	IV	2.3	8.2	34.7		10.8			302.5	169.9	112.4	584.8
18	12.4	11.0	10.0	2		+	IV	2.6	14.3	36.5		8.2			183.2	62.1	45.4	290.7
19	13.0	11.4	12.2	2		+	IV	3.9	21.5	13.6	13.8	25.9	8.4		168.3	200.3	48.8	417.4

Table 1b

Concentration of polychlorinated biphenyls in the muscle tissue of the Baltic herring (Gulf of Riga)

No.	Length, cm		Weight, g	Age	Sex		Maturity	Fat %	Concentration, ng g^{-1} (fat)							
	L	L_s			♂	♀			49	52	101	118	138	153	180	105
1	12.3	10.3	10.3	2	+		III	3.8			26.0	53.0	27.9	49.7	13.2	12.8
2	13.6	12.2	12.3	2		+	IV	2.3	31.7	67.6	162.5	390.6	119.7	226.6	101.9	64.2
3	12.4	11.3	10.0	2		+	IV	2.6	19.1	45.9	46.9		68.0	134.0	26.4	17.2
4	13.0	11.4	12.2	2		+	IV	3.9	7.5	18.9	35.0	83.2	61.5	102.7	12.1	18.1
5	12.2	10.8	9.1	2	+		III	3.2	10.2	34.6	43.5	96.3	63.0	108.6	18.1	20.3
6	13.6	12.3	12.8	2		+	III	0.8			31.8	295.8	236.9	421.7	135.2	73.3
7	11.7	10.4	9.0	2	+		III	0.9			145.5	170.4	183.1	363.7	99.9	23.1
8	13.3	11.0	12.5	2	+		IV	1.7	52.1		87.2	130.7	107.7	215.8	58.9	7.2
9	12.1	10.7	9.6	2		+	IV	1.6	62.0		70.7	100.4	66.1	118.3		7.3
10	12.1	10.7	9.4	2		+	IV	4.2	15.5		35.4	51.9	62.4	87.8	99.8	3.9
11	13.4	12.2	12.2	2	+		III	1.2	76.9		99.7	118.0	87.1	165.3	5.2	5.9
12	12.0	10.6	9.4	2	+		IV	1.3	54.9		74.2	103.3	77.0	149.4	37.6	
13	11.6	10.3	8.1	2		+	III	3.5	9.7		64.1	108.2	69.8	125.9	21.9	6.2
14	12.6	11.3	11.1	2		+	IV	1.2	28.9		139.0	153.4	159.1	256.7	240.4	14.5
15	12.9	11.4	12.0	2		+	III	2.2	15.3		34.4	73.0	79.6	122.0	101.1	6.1
16	12.3	11.0	10.3	2	+		III	3.8		9.4	30.5	57.7	40.4	46.6	7.0	
17	13.6	12.2	12.3	2		+	IV	2.3	27.6	55.4	159.9	204.5	227.0	269.7	70.6	
18	12.4	11.0	10.0	2		+	IV	2.6	34.4	19.1	59.4	97.1	83.9	112.3	19.4	
19	13.0	11.4	12.2	2		+	IV	3.9	17.9	19.0	47.3	92.6	75.7	83.3	10.0	

MATERIAL AND METHODS

The herring samples were taken from trawl catches in the northern part of the Gulf of Riga in April 1992. Age of fish was determined using otoliths (Table 1).

The average body weight of herring, which had been relatively stable during 1940–60, started to increase in most regions of the Baltic Sea in the late 1970s (Fig. 1). High average values of body weight and length were

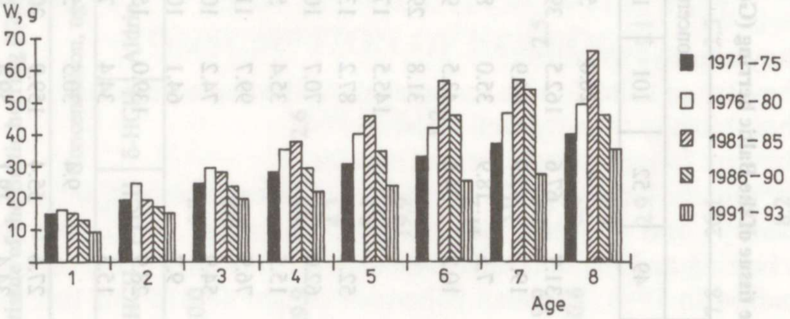

Fig. 1. Mean weight at age of herring in the Gulf of Finland in 1971–93 (from Raid & Lankov, 1995).

observed in all age groups of herring until the second half of the 1980s, when they started to fall unexpectedly. In the 1990s, also a drop in the average body weight of sprat occurred. The studies of the feeding of herring and sprat carried out during 1982–92 in the northeastern part of the Baltic Sea showed changes in the diet of the fish and also a rising number of fish with an empty stomach (Fig. 2). These changes are probably connected with the hydrological conditions prevailing in the Baltic Sea since the mid-1980s (Lumberg & Ojaveer, 1991). Hydrological conditions are, at least partially, responsible for the rapid decelaration in the growth of sprat and (especially) herring (Lankov, 1992; Raid & Lankov, 1989, 1995).

The fish samples (10 g) were frozen before further treatment. The samples were extracted with a mixture of acetone and 10% methyl-butyl ether in hexane (3.5:1) and *n*-hexane/methyl-butyl ether (9:1) (Jensen et al., 1983).

Dissolved lipid (0.1–0.2 g, fat) is then cleaned applying two methods: method I, a silica gel column treated with concentrated sulphuric acid; the

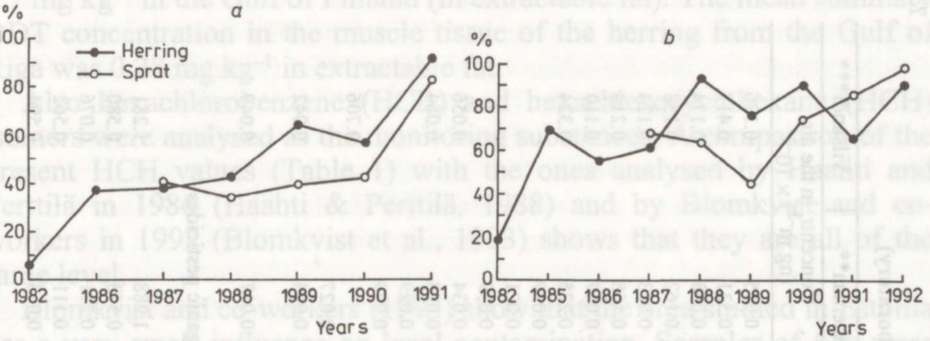

Fig. 2. Proportion of herring and sprat with empty stomachs in the northern Baltic in April 1982–91 (*a*) and in November–December 1982–92 (*b*) (from Raid & Lankov, 1995).

organochlorines were eluted with *n*-hexane; and method II, a silica gel column treated with concentrated ortophosphoric acid; the organochlorines were eluted with 20% CH_2Cl_2 in *n*-hexane (Jensen et al., 1983; Bergman et al., 1992).

The recovery of organochlorines from the extraction and clean-up procedures was measured. PCB and COP were analysed by capillary gas-chromatography (Varian 3400/3300) fitted with a 63 Ni electron capture detector. The column used was a 30 m DB-5 (Roots & Aps, 1993; Roots, 1995).

The determinations of the content of PCB and organochlorine pesticides in the organisms of herring in the Gulf of Riga indicated the need for a thorough study of the emission paths of these pollutants into the bay, particularly by atmospheric transport (Roots, 1990, 1995; Роотс, 1992).

The air samples (sampling absorbent polyuretane foam) of *c*. 1000 m^3 were collected by Estonian scientists from Estonian EMEP Stations at Lahemaa and Vilsandi during two weeks. Professor P. Larsson and co-workers from the Department of Ecotoxicology, Lund University, Sweden, analysed the samples (Larsson & Okla, 1991; Larsson et al., 1993). The results are presented in Table 2.

RESULTS AND DISCUSSION

Our data on herring from the Gulf of Finland (Roots & Aps, 1993) and the Gulf of Riga (Table 1) in 1991–92 showed lower DDT levels than the samples collected by Haahti and Perttilä in 1986 (Haahti & Perttilä, 1988). According to Haahti and Perttilä, the summary DDT concentrations in the

Table 2

Levels of chlororganic compounds in the air (analysed by Lund University Ecotoxicological Laboratory)

Domain No.*	Chlorobiphenyls No. (IUPAC)	Lahemaa** Concentr. in the air, (ng m^{-3}) × 10^{-2}	Vilsandi*** Concentr. in the air, (ng m^{-3}) × 10^{-2}
2	7, 9	0.063	
3	6	0.082	
6	15, 17, 18	0.465	0.257
7	24, 27	0.105	0.066
8	16, 32	0.212	0.023
11	26	0.035	0.034
12	25	0.178	0.102
13	28, 31	0.382	0.303
14	20, 33, 53	0.285	0.197
15	22, 51	0.080	0.039
16	45	0.150	0.148
19	52	0.531	0.334
20	49	0.276	0.172
21	47, 48, 75	0.275	1.655
22	35		
23	44	0.358	0.258
24	37, 42, 59	0.171	0.094
25	41, 64	0.346	0.195
27	40	0.213	
29	63	0.154	0.333
30	74	0.137	0.112
31	70	0.231	0.189
32	66, 95	0.472	0.500
36	92	0.233	0.256
37	84	0.220	0.345
38	90, 101	0.325	0.267
39	99	0.226	0.041
42	97	0.123	0.099

Domain No.*	Chlorobiphenyls No. (IUPAC)	Lahemaa** Concentr. in the air, (ng m^{-3}) × 10^{-2}	Vilsandi*** Concentr. in the air, (ng m^{-3}) × 10^{-2}
43	87, 115	0.095	0.126
46	77, 110	0.331	0.417
47	82, 151	0.098	0.174
50	118, 149, 123	0.162	0.158
53	146	0.023	0.124
54	105, 132, 153	0.168	0.214
55	141, 179	0.037	0.110
58	138, 158, 160	0.225	0.324
61	187	0.039	
62	183	0.021	
63	128	0.039	
66	174	0.034	0.056
67	177	0.028	0.084
68	156, 171, 202	0.029	
72	180	0.029	
75	200		0.706
77	170, 190	0.027	
79	199	0.064	0.042
80	196, 203		
84	194	0.017	0.049
	Chlororganic pesticides		
	α-HCH	1.288	1.281
	HCB	0.553	0.580
	γ-HCH	0.211	0.076
	p,p'DDE	0.111	0.562
	p,p'DDT	0.063	0.594

* After Schultz et al., 1989. **The sampling period 25 February to 9 March 1992; *** 28 March to 11 April 1992.

herring muscle ranged from 0.3 mg kg^{-1} in the Gulf of Bothnia to 0.7 mg kg^{-1} in the Gulf of Finland (in extractable fat). The mean summary DDT concentration in the muscle tissue of the herring from the Gulf of Riga was 0.28 mg kg^{-1} in extractable fat.

Also hexachlorobenzene (HCB) and hexachlorocyclohexane (HCH) isomers were analysed as the monitoring substances. A comparison of the present HCH values (Table 1) with the ones analysed by Haahti and Perttilä in 1986 (Haahti & Perttilä, 1988) and by Blomkvist and co-workers in 1992 (Blomkvist et al., 1993) shows that they are all of the same level.

Blomkvist and co-workers (1993) show that the area studied in Estonia has a very small influence on local contamination. Samples of fish were collected during August and September 1992 at three different localities along the coast of the Baltic Republics and from a Swedish reference area (Kvädöfjärd). The localities were south of Hiiumaa Island in Estonia, Daugavgriva (the delta of the Daugava River) in Latvia, and Courland Lagoon in Lithuania. It is obvious that the Courland Lagoon results indicate a strong eutrofication. There is a similar tendency, although not as strong, in the Daugavgriva material (Blomkvist et al., 1993).

We recommend that besides herring's age, length, weight, sex, fat percentage, and degree of maturity, in the future the percentage of empty stomachs (and the content of different food in stomachs) should be considered as an additional parameter. One can assume that the rising number of fish with an empty stomach has contributed to the stability of chlororganic compound contents in herring organisms in the northern part of the Baltic Sea (Fig. 2) (Roots, 1996).

The input of PCB components to the Baltic Sea from the atmosphere (both wet and dry deposition) deserves special attention (Roots, 1990). Combined use of data on the PCB concentration and meteorological parameters will contribute to a better understanding of the direction and origin of the air masses above the Baltic Sea. The atmosphere is an important contributor of anthropogenic matter to the marine ecosystem (air–sea surface–plankton–herring). Above the Baltic Sea the concentration of PCB and their components proved especially high with southwesterly winds. This endangers the islands of Saaremaa and Hiiumaa, since these winds are prevailing on the Baltic Sea. Herring starts its active search for food in March–April. It is the time when the highest levels for some COP have been found in the air in Central Europe (Roots, 1995).

The analyses of PCB and pesticides made by the Lund University on the samples taken in three Baltic air research stations – at Vilsandi and Lahemaa in Estonia and at Salaspils near Riga in Latvia – showed that the air and rain water samples taken in Estonian stations were cleaner than the samples taken in Latvia (Larsson & Okla, 1991). This refers either to the long-range transportation or to a local emission centre situated near the Gulf of Riga.

Air and precipitation concentrations at the Latvian Salaspils station were respectively 7.6 and 8.7 times and those at the Lithuanian (Courland Lagoon) station 1.3 and 2.6 times higher in 1991–92 than the same concentrations at the Estonian Lahemaa station (Larsson et al., 1993; Roots, 1993). The same situation was observed by Blomkvist et al. (1993). Although the contamination analyses were made only on samples from one year and should be verified by additional studies, the results suggest that the most important risk substances, PCBs and DDTs, occur in rather low concentrations also in areas receiving wastes from such a big city as Riga (Blomkvist et al., 1993). The results of Hiiumaa Island are very promising and strongly support the opinion that the Gulf of Riga is a high class reference area.

The frequency of cancer in Estonia is below the European average. Estonia ranked 47th for male and 51st for female cancer (Thomson, 1994). However, the incidence rates of stomach cancer and leukemia in both sexes and cancer of the pancreas in males were higher in Estonia than those in other European countries on average. According to the pattern of these cancer sites, Estonia ranked 4–6th in Europe and was among the top 20 when 166 world populations were compared. The incidence rates of cancer of the thyroid and testes in males, non-Hodgkin's lymphoma in females, and breast and bladder in both sexes in Estonia were low compared with other European countries. Estonian site distribution of cancer is similar to that of Latvia and several other Eastern European countries (Thomson, 1994).

The following questions need a proper answer:

– What is the amount of fish consumed per day that could be taken as basis for our calculations?

– From which standards shall we proceed in evaluating the risk poised by toxicants?

When considering the possible effects of consuming seafood, it is necessary to characterize communities and individuals according to the amounts they consume, since communities in different parts of the world are likely to show big differences in their consumption of seafood. For example, based on FAO food balance data, the European regional average consumption of seafood (fish and shellfish) is 60 g per person per day and in East Asia 79 g per person per day. The figure used to represent high consumption is 150 g per person per day (Reports and Studies, 1991).

The joint FAO/WHO Expert Committee on Food Additives (JECFA) generally sets the ADI (the acceptable daily intake) of a food additive on the basis of the highest "no-observed-effect" (NOEL) level in animal studies.

ADI is the daily dosage of a chemical which, during an entire lifetime, appears to be without appreciable risk on the basis of all the facts known at the time. Where an ADI has been established for a particular pollutant it will be noted whether the intake from seafood is likely to be a substantial part of the ADI (Reports and Studies, 1991). NOEL is the risk that may be incurred by especially susceptible groups, such as children and old people.

ADI and NOEL values used in our work are based upon the literature data (FAO/WHO, 1971, 1978, 1985; Reports and Studies, 1991).

For each level of seafood consumption, the health (carcinogenic) risk from marine pollutants varies according to their concentration. This is turn varies from place to place and from time to time. We took as base for assessing the health risk organochlorine concentrations in the muscle tissue of herring caught in Estonian coastal areas (gulfs of Riga and Finland) (Roots & Aps, 1993; Roots, 1995).

DDT and its metabolites are some of the most frequently reported organochlorine contaminants in the environment. An overall NOEL for the toxicity of DDT of 0.25 mg kg^{-1} body weight per day in humans and ADI of 0.02 mg kg^{-1} body weight was set by the Joint Meeting on Pesticide Residues (JMPR) in 1984 (FAO/WHO, 1985; Reports and Studies, 1991).

If a person weighing 60 kg consumes 150 g d^{-1} of fish containing 20 µg kg^{-1} of DDT and its metabolites, the daily intake will be 0.05 µg kg^{-1} body weight. This would constitute 1/400 of the ADI.

Hexachlorocyclohexane. Technical HCH contains a number of isomers of which gamma-HCH (lindane) is commonly used as an insecticide. In the Baltic Sea the concentrations of all three isomers (alpha, beta, and gamma) are similar (Roots & Aps, 1993).

If a 60 kg person eats 150 g d^{-1} fish containing 60 µg kg^{-1} (alpha-, beta-, and gamma-HCH), this will constitute 1/53 of the ADI (for gamma-HCH 0.008 mg kg^{-1} body weight was set by the JMPR). No ADI was established for the other isomers or for technical HCH.

Aldrin+dieldrin. These insecticides are not a problem for the Baltic Sea. If a 60 kg person consumes 150 g d^{-1} fish containing 0.5 µg kg^{-1} (max) of aldrin plus dieldrin, the daily intake will constitute 1/80 of the ADI (0.0001 mg kg^{-1} body weight).

Chlordane. The main components are alpha- and gamma-chlordane and the most toxicologically significant metabolite is oxy-chlordane.

If 150 g of fish containing 4 µg kg^{-1} gamma-chlordane plus oxy-chlordane (Roots & Aps, 1993) is consumed daily by a 60 kg person, this will constitute 1/50 of the ADI (0.005 mg kg^{-1} body weight).

Polychlorinated biphenyls. Human health studies of "Yusho" disease in Japan (1968) and in Taivan (1979) showed that contamination of food with PCB from an industrial source can be a cause of severe human diseases.

If a 60 kg person eats 150 g d^{-1} of fish containing 20 µg kg^{-1} PCB, this will amount to 0.05 µg kg^{-1} body weight per day. This is about 1/3000 of the estimated average daily intake in the accident in Japan in 1968 (Reports and Studies, 1991), which involved about 1000 persons, and 1/800 of the NOEL of 0.04 mg kg^{-1} body weight per day for monkeys.

To sum up, we can say that the levels of organochlorines found in the organism of the Baltic herring in the coastal areas of Estonia (Table 1) do not pose any human health risk. However, it is known that marine food is not the only route of human exposure and any overall risk assessment must take this into account (Roots, 1986).

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