Residues of Hexachlorobenzene in Baltic Fish and Estimation of Daily Intake of this Compound and Pentachlorobenzene with Fish and Fishery Products in Poland*

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Abstract

An attempt has been made to review pollution of the Baltic fish and fishery products by hexachlorobenzene (HCBz) and to estimate daily intake of HCBz and pentachlorobenzene (PCBz) with marine and freshwater fishes and fishery products in Poland. The calculated arithmetic mean weighed concentrations of HCBz in muscle tissue of the Baltic fish are 22 ng/g wet weight in herring in 1973-1985 (n = 3115) and 4.3 ng/g in 1986-1992 (n = 371); 14 ng/g in sprat in 1971-1985 (n = 939); 6.4 ng/g in salmon in 1976-1982 (n = 151); 14 ng/g in eel in 1981-1982 (n = 480); 200 ng/g m cod-liver in 1973-1978 (n = 72) and 85 ng/g in 1979-1990 (n = 918); 0.65 ng/g in cod in 1981-1992 (n = 527); 2.2 ng/g in flatfishes in 1974-1992 (n = 396); 0.96 ng/g in perches in 1983-1992 (n = 70) and 2.8 ng/g in other fish species in 1981-1992 (n = 192). The intake of HCBz has been assessed for 60 to 92 ng/person/day in the years 1973-1985 and from 45 to 56 ng/person/day (mean 52 ng) in 1989-1996, while of PCBz for 7.8 to 11 ng/person/day (mean 9.4 ng) in 1989-1996.

Keywords: hexachlorobenzene, HCBz, pentachlorobenzene, PCBz, fish, Baltic Sea, dietary intake, food, pollution, organochlonnes.

Introduction

Hexachlorobenzene (HCBz) is a dioxin-like compound that binds to the Aryl hydrocarbon (Ah) receptor, resulting in dioxin-like effects and bioaccumulates. Exposure to HCBz results in induction of hepatic cytochrome P4501A1 and P4501A2 activities, hepatic porphyrin accumulation and excretion, alterations in thyroid hormone levels and metabolism, alterations in retinoid levels, liver damage, reduction in reproduction, splenomegaly, increase in mortality, neurological alterations, teratologic effects, and immunotoxic effects [1]. Hexachlorobenzene

is a persistent compound, which has become a wide-spread environmental pollutant. A main source of environmental pollution with HCBz can be municipal solid waste incineration and some industrial chemical processes [2]. Hexachlorobenzene posses fungicidal activity and has been used world-wide [3]. Pentachlorobenzene (PCBz) is an intermediate and a by-side product of chemical industrial processes such as synthesis of the fungicide pentachloronitrobenzene (PCNBz) and in technical formulations of HCBz [4-6]. It is also formed during combustion of municipal solid waste and can be found in flue gas as a main constituent (50%) of emitted con-

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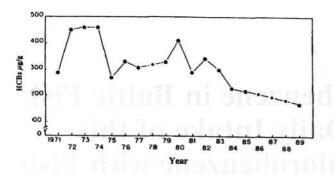
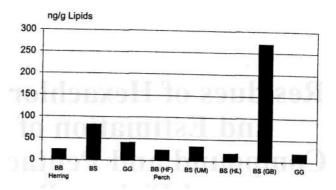


Fig. 1. Time-trends of hexachlorobenzene in cod-liver oil of the southern Baltic Sea origin (after 18)

geners of chlorobenzene [7-9]. Another indirect source of PCBz in the environment is metabolism of HCBz; PCBz is further degraded to tetra-, tri-, di- and monochlorobenzene [10, 11]. In aquatic environments PCBz is easily bound to settling particulate matter and sediment, and is bioaccumulated by fish and other marine biota [12]. Pentachlorobenzene is a relatively persistent compound - its half-life time in sediment is - 18 days [13], while in muscle tissue of fish it is > 7 days [14], and seems, like HCBz, to be a widespread environmental pollutant. Concentrations of PCBz determined in Baltic and the North Sea fish are - 5-10 fold lower than of HCBz [15, 16].

In a study by de Boer [16] spatial differences were found in distribution of HCBz concentrations in livers of cod from the North Sea. Cod caught in the southern part of the North Sea were more contaminated with HCBz than specimens from the central part, while those from the northern part showed lowest contamination. When examining cod-liver oil produced of codfish caught from various areas of the North-eastern Basin of the Atlantic Ocean in the 1980s it was found that those originating from the southern part of the Baltic Sea were most contaminated with HCBz [17]. In a time-trend study of organochlorines in cod-liver oil made of fish caught in the southern Baltic in the years 1971-1989 a slow rate of



'Fig. 3. Spatial variations of HCBz concentration in herring and perch in the Baltic Sea: Bothnian Bay (BB), Bothnian Sea (BS), Gulf of Gdansk (GG), Harufjarden (HF), Umea (UM), Hornslandet (HL) and Gavlebukten (GB) [20].

decrease of HCBz concentrations was observed after the year 1973 (Fig. 1) and indicated a continuous input of that chemical [18]. Concentrations of HCBz have further decreased in Baltic herring muscle tissue and in cod liver in 1987-1996; however, rates varied depending on the site, and for some sites a steady state condition was observed (Fig. 2) [19].

Based on recent studies of geographical distribution and degree of contamination with organochlorines of herring and perch in the Baltic Sea the elevated concentrations of HCBz (Fig. 3) and indicating continuous input from point emission source and higher pollution burden have been observed in the Gavlebukten in the Bothman Sea [20].

Culinary treatments such as frying, baking, grilling, smoke-curing or boiling of fish decrease (~ 20 %) HCBz content of fish [21]. Also, industrial processing of fish manufacturing of canned seafood results in a decrease of HCBz concentration in the final product [22, 23].

The aim of this study is to review available data on pollution of Baltic fish with HCBz and PCBz and to assess daily intake of those compounds with fish and fishery products in Poland.

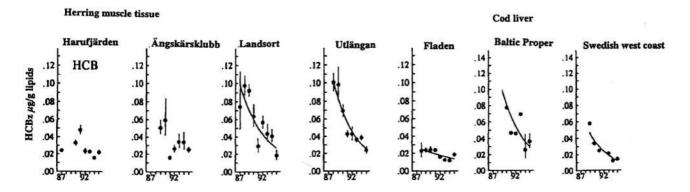


Fig. 2. Concentrations of HCBz in herring muscle and cod liver $\mu g/g$ lipid weight) at various locations in the Baltic Sea; adapted after [19].

Materials and Methods

The basis for calculation of dietary intake of organochlorine residues with fish and fishery products in Poland are explained in detail in another paper [24]. All the available data on HCBz content of Baltic fish and fishery products were used to calculate arithmetic mean weight concentration of this compound in muscle tissue of a particular fish species and families or groups of fish and in edible livers of cod in a particular year or period (Table 1). The data displayed in Table 1 are presented chronologically. The same procedure as mentioned above was used to calculate intake of PCBz but the number of data available for this compound is far less than for HCBz.

Results and Discussion

When analyzing residue concentrations of HCBz in Baltic herring presented in Table 1 it is visible that fish caught in the years 1973-1985 were more contaminated with this substance than the specimens netted in the 1990s. The arithmetic mean weighted concentration of HCBz for Baltic herring caught before the year 1985 is > 10 ng HCBz/g wet weight, while after the year 1985 it is < 10 ng/g wet wt. Similarly, the livers of the Baltic codfish (mainly cod) netted in the years 1973-1978 are characterized by greater residual concentrations (~ 200 ng HCBz/g wet wt) than of codfish caught after 1985 (< 100 ng/g wet wt). A time-trend data of HCBz for Baltic herring and the livers of Baltic codfish presented in Table 1 correspond well with presented earlier time-trend data of HCBz in cod-liver oil produced of the livers of cod caught in the southern part of the Baltic Sea as well as in the livers of cod caught in the North Sea. The arithmetic mean weighed concentration of HCBz in cod-liver oil produced in Poland of the livers of codfish caught in the southern Baltic Sea in the years 1971-1980 is 370 ng/g, and in cod-liver oil produced in the years 1981-1989 is 260 ng/g (Table 1). A decreasing time-trend of the residual concentrations of HCBz observed recently for the Baltic herring and cod can be explained due to subsequent withdraw of HCBz as a seed-dressing agent for agricultural purposes in the Baltic States in the 1970s, and decreased emission to the atmosphere from the upgraded or newly created the municipal solid waste and hazardous waste incinerators. Nevertheless, the integrated data on residual concentrations of HCBz in environmental matrices such as the Baltic herring, the livers of cod and cod-liver oil (Table 1) indicated that from the year 1980 there is only a slow decrease of HCBz concentrations for the entire Baltic and indicated a continuous input of that chemical. In the case of the muscle tissue of the Baltic cod and sprat as well as of the other species, families and groups of fish indicated in Table 1 the number of available data on residual concentrations of HCBz for the years 1971-1992 is too small to suggest any time-trends.

Fish due to a relatively low activity of the monooxygenase enzymes have a small capacity to metabolise persistent, lipophilic and bioaccumulative environmental contaminants such as organohalogenated compounds. A main rout of exposure to fish of organohalogenated environmental contaminants which are characterized by a relatively high value of the n-octanol/water partition coefficient (log $K_{\text{o/w}} > 5$) is the alimentary track (~ 90%). Accordingly, the residual concentration of HCBz found in the edible parts of fishes is dependent of the type of food available to fish, which is related territorially to a particular food webs, and on spatial differences of the pollution pattern, which determine daily intake of the compound by fish. Consequently, large and old specimens of the particular fish species, which are territorially connected with a particular fishing ground or water body, and a relatively higher contaminant intake rates should be characterized by greater residual concentrations of HCBz and similar compounds than specimens of smaller body size and young.

When examining the spatial differences in pollution load of organohalogenated chemicals in the Baltic Sea using herring and perch as bioindicators it was indicated that perch from a specific site in the Bothnian Sea, which neighbors a highly industrialized site in Sweden, are much more contaminated with HCBz (270 ng/g lipid weight) when compared to fish in the Gulf of Gdansk and also of some other sites in the northern part of the sea, which contained from 18 to 33 ngHCBz/g on a lipid weight basis (Fig. 2).

A tolerance of the residual concentration of HCBz in edible parts of fish and fishery products established in Sweden in the early 1980s is 200 ng/g wet weight [62]. The concentrations of HCBz above 200 ng/g wet weight were recorded in muscle tissue of a small percentage of eels and in larger extent in cod liver (Table 1).

The calculated in this study the arithmetic mean weighted concentration of HCBz in muscle tissue of the Baltic fishes are: 0.65 ng/g wet weight in cod (1981-1992; n = 527); 22 ng/g wet weight in herring in 1973-1985 (n = 3115) and 4.3 ng/g wet weight in 1986-1992 (n = 371); 14 ng/g wet weight in sprat (1971-1985; n = 939); 2.2 ng/g wet weight in flatfishes (1974-1992; n = 396); 6.4 ng/g in salmon-like fishes (1976-1982; n = 151); 14 ng/g wet weight in eels (1981-1983, n = 480); 0.96 ng/g wet weight in perch-like fishes (1983-1992; n = 70) and 2.8 ng/g wet weight in fish considered as "a group of unsorted fishes" (1981-1992; n = 192). In the case of cod liver the arithmetic mean weight concentration of HCBz was 200 ng/g wet weight in 1973-1978 (n = 72) and 85 ng/g wet weight in 1979-1990 (n = 918).

An assessed data on intake rates of HCBz contaminating fish and fishery products available in Poland in the years 1973 - 1996 are presented in Table 2. All available and published data on the amount and the structure of fish consumption in Poland [63] were taken into account when calculating the intake rates of HCBz in 1973-1985, while when assessing HCBz intake rates in 1986-1996 the calculations were based on a somewhat simplified consumption (figures presented elsewhere) [24].

An assessed intake of HCBz with fish and fishery products in Poland is from 22 to 34 μg per capita annually in 1973-1985 and from 16 to 23 μg in 1989-1996, i.e. from 60 to 92 ng and from 45 to 56 ng (mean 52 ng) per capita daily, respectively. For people eating 10 g (an average consumption rate per capita in Poland), 105 g or 95-160 g (net weight of cans' content) of the processed

Table 1. The concentrations of HCBz in Baltic fish and fishery products (ng/g wet weight).

Family, species and year	n	Mean	Range	Reference
1-1-	2	3	4	5
Gadidae			1.00	Ed Ne.
Cod Gadus morhua, whitin	o Merls	anoins n	nerlanous	
1981	98	0.77	0.2-6.1	25
1982-1983	47	<10*	<10-<10	26
1983	232	0.66	<0.1-2.7	27,28
1985	181	0.60	0.35-1.3	29
1986	13	0.45	0.43-0.47	29
1992 ·	3	0.46		15
Clupeidae Herring Clupea harengus				
1973	20	14	8-19	30
1974	63	11	8-18	31
1975	10	12	6-17	31
1976	15	40	9.7517/65	32
1977	1124	30	7-70	33
1979	205	28	10-48	33
1980-1981	1218	17	0.5-39	34-37
1982-1983	33	<10	<10-30	26
1983	299	12	2.1-150	28,32
1983	88	9	1-39	38
1985	40	15	14-16	32
1986	100	0.35	0.29-0.42	39
1986	47	5.1	4.4-5.5	29
1986	100	6.3		40
1987	100	6.2		40
1991	21	2.5	1.6-3.3	20
1992	3	3.7	1.0 0.0	15
Sprat Sprattus sprattus	, ,			1 25
1971-1974	42	<5		41
1975	30	18	17.00	42
1978	177	18	17-20	42
1981	102	6.4	0.8-62	43
1982-1983	47	<10	<10-10	26
1983	225	17	1.5-71	28,44
1983	31 285	18 14	8.1-35	45
1985		1 14	10-20	29
Pleuronectidae and Bothid		D/		
Flounder Platichthys flesus Psetta maxima	, plaice	Pleuroi	nectes plate	ssa, turbot
1974-1975	61	1.6	ND-20	30,31
1981	125	4.2	ND-27	35, 46, 47
1983	170	0.98	1	48
1985	15	3.0		29
1986	10	1.1	0.54-1.6	29
1992	15	0.90	0.60-1.5	15
Salmonidae	areas .		Orenande Statistica Statistica	
Salmon Salmo salar, brow gairdneri irideus	n trout	Salmo	<i>trutta</i> , trou	t Salmo
gairdneri irideus	22			
gairdneri irideus 1976	20	3.5	3-4	33
gairdneri irideus 1976 1978	20 8	3.5	3-4 32-110	33 33
gairdneri irideus 1976	20	3.5	3-4	33

1 1000 1000	2	3	4	5
Anguillidae				
European eel Anguilla angui	illa			
1981	125	1 18	3-66	36,46,47
1982	128	13	1.5-210	50,51
1983	227	12	<0.5-330	The state of the s
Percidae	221	12	(0.5-550	33,31-33
Perchae Perch <i>Perca fluviatilis</i> , pikep	orch S	tizostas	lian luciana	rea
reich reica javianus, pikep	eren 3	uzostea	иоп шсюре	rcu
1983	6	0.9	0.5-1.9	35
1991	35	0.27	0.23-0.36	20
1991	10	3.2		20
1992 .	16	1.1	0.79-1.3	15
1992	3	0.85		15
Unsorted fish				
Garfish Belone belone, Four	bearde	d rock	ling Enchel	vopus
cimbrius, Lamprey Lamper				
rous blenny Zoarces viviparu				
tomus, Greater sand eel Hyp				
eel Ameodytes tobianus	J. JP.		, 200	
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1981	15	0.72	0.1-2.8	46
1981	7	1.1	0.42-2.8	35,46
1983	1	0.4		35
1983	2	31	27-35	35
1983	24	2.2	0.89-6.7	35
1986	20	3.1		29
1986	68	4.5	1.6-18	54
1992	55	0.62	0.36-0.93	15
Cod liver				
1973	42	1 220	10-1600	1 30
1974	- 5	190	50-310	30
1975	4	190	110-240	55
1978	10	280	110-240	23
1978	11	78	<1-230	56
1979	25	45	22-96	56
1980	9	44	30-110	56
1981	585	89	<10-530	1
				47,57-59
1982-1983	43	70	<10-170 <10-330	60
1983	210	96	<10-330	61
1985	40	34		39
1986	3	36	50.76	29
1990 (canned)	3	60	50-76	22
Cod-liver oil				
1971-1980	9	370	290-460	18
1981-1989	6	260	200-310	18
1980s	11	280	170-370	17
1980s (western Baltic Sea)	9	160	110-260	17
1980s (North Sea and	ì			
Norwegian Sea)	2	120	110-130	17
1980s (Iceland)	8	87	73-100	17
Cod hard roe		"	100	"
1975	1	8		55
	1	°		33
Herring hard roe			10	55
1975	2	6	4-8	55
Herring soft roe (milt)				
1975	2	4	3-4	55

Notes: * value omitted when calculating arithmetic mean weighted; ND, not detected.

Year	Daily intake (ng per capita)							
	Total	Cod	Herring	Sprat	Flatfishes	Other fishes*		
1973-1975	72-92	1.3-1.7	43-56	27-33	0.4-0.4	<0.4-1		
1976-1980	66-84	1.1-3.0	48-63	7-29	0.3-0.4	<0.7-1		
1981-1985	60-81	1.4-2.9	50-70	4.1-11	0.3-0.3	<0.3-<0.8		
1989	50	14**	36***					
1990	45	13**	32***					
1991	51	14**	37***					
1992	53	15**	38***	1				
1993	56	16**	40***					
1994	56	16**	40***					
1995	. 53	15**	38***					
1996	54	15**	39***					

Table 2. Estimation of daily intake of HCBz in fish and fishery products in Poland

Notes: *Salmon, Brown trout, Trout, European eel, Pike, Perch family and *Cyprinides* (for Pike and *Cyprinides* was assumed that HCBz content of muscle tissue is 0.9 ng/g wet weight *i.e.* as is for perch family) and in other marine fish - considered in fishery statistic in Poland as unsorted fish and other freshwater fish - also considered in fishery statistic as unsorted fish; **as lean fish of < 5 % lipids in muscle tissue; ***as fatty fish of > 5 % lipids in muscle tissue.

(canned) cod livers annually an assessed intake of HCBz with a such product is 0.6. 6.3 and 5.7-9.6 µg per capita, respectively [22]. On the other hand, for peoples eating cod-liver oil of Baltic origin (200 g of oil per annum - 1 bottle) an estimated intake of HCBz was 74 µg (1971-1980) and 52 µg per capita annually (1981-1996), while it was 19 µg for those eating the cod-liver oil produced in the United Kingdom, Norway or Island, i.e. 200, 140 and 52 ng per person daily, respectively. The lipid rich muscle tissue of fish such as herring and sprat, like it is in the case of the polychlorinated biphenyls (PCBs) and chlordane (CHLs) [24, 63], is a main source of intake of HCBz with a fish diet for a general population in Poland (Table 2). Obviously for a specific group of people frequently eating canned cod liver manufactured of Baltic cod those foodstuffs are a main source of daily intake of HCBz.

If one assumes that the concentration of pentachlorobenzene in muscle tissue of fatty fish (> 5% lipid, herring and sprat) was 0.75 ng/g wet weight and of lean fish (< 5%; other species) [15] was 0.29 ng/g wet weight an assessed daily intake of PCBz with fish and fishery products in Poland in 1989-1996 was from 7.8 do 11 ng (mean 9.4 ng) per capita.

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