

Intake of Hexachlorobenzene with Food in Poland

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Abstract

Hexachlorobenzene exposure from food in Poland in 1970-1996 was calculated by multiplying its annualized mean consumption rates by residue concentrations quantified in food. Estimated daily dietary intakes of hexachlorobenzene were 1.3 to 1.5 $\mu\text{g per capita}$ in 1970-1996, on average. Grains, potatoes, vegetables and fruits, meat, fish and dairy products nearly equally contributed as a source of hexachlorobenzene exposure in total diet in Poland.

Keywords: hexachlorobenzene, HCBz, food, intake

Introduction

Hexachlorobenzene (HCBz) has been a contaminant in the global environment for more than 80 years. In practice the residues of HCBz are detectable almost in every examined sample of human adipose tissue or milk [1-3]. Specific physico-chemical properties like lipophilicity, moderate volatility, permeability through biological membranes and high resistance to degradation are main features determining behaviour of HCBz in the environment and its migration via food webs. Due to the widespread occurrence of HCBz in the environment as a contaminant, food contains this compound in small concentrations. Also, food is a main source of HCBz intake for human beings.

An occurrence of the residues of HCBz in food may be connected both with fungicidal properties of this substance and the fact that HCBz was used in the past as fungicide in dressing a seed and pollution with this compound of the environment. Total quantity of HCBz imported to Poland in 1962-1972 was about 194.6 tons, and range of the imported quotas was from 8.0 tons in 1972 (no import in 1963) to 45.1 tons in 1968 [4]. From 1945 the cumulative world production of HCBz is 1-2 million tons. Shortly after 1970 many countries abolished the use

of HCBz as a fungicide but sporadically this substance was used until the 1980s, *e.g.* in the former East Germany and the Soviet Union [5].

Various chemical synthesis and processes with HCBz present as a technical impurity, substrate, intermediate and/or final product seem to be the main source of environmental contamination by this compound [6-8]. Among agricultural, forest and city soils (0-10 cm layer) sampled in Poland in the 1990s definitely the highest degree of contamination by HCBz was observed at the industrialized and urbanized sites in the southern part of Poland - from the cities of Chorzow, Katowice, and Krakow [9].

A degree of contamination with HCBz found in a human's body is apparently greater for individuals inhabiting industrialized regions rather than agricultural [3]. The human body burden of HCBz can be relatively high for some people. For example, in Slovakia women's milk contained HCBz at a concentration of 5300 ng/g lipid in 1977, 3500 ng/g in 1978, 680 ng/g in 1980-84 and 420 ng/g in 1990 [3]. The data on HCBz concentrations in human adipose tissue of Poles living in the cities of Skierniewice, Poznan and Gdansk have indicated that Poland doesn't belong amongst countries especially polluted with this compound. The range of the mean concentrations of

HCBz was from 220 to 360 ng/g lipid weight in 1979-1990, and considerably greater concentrations were noted in the former Czechoslovakia (840 ng/g lipid), Italy (2300 ng/g lipid) and Navarra in Spain (3400 ng/g lipid) [2,10].

Butter in Germany in 1970-1983 contained residues of HCBz from 12 to 1800 ng/g, on average, but greater contamination took place at the beginning of the 1970s [11].

Apart from the butter a decreasing time-trend of contamination with HCBz was observed in adipose fat of hogs in Germany in 1977-1988, *i.e.* the concentration dropped from approximately 40 ng/g to approximately 1 ng/g [12].

In Poland contamination of food by HCBz is monitored from many years and residue concentrations of this compound in 1970-1996 were low, on average

Table 1. Residues of hexachlorobenzene in food µg/kg).

Product	Country and year	No.	HCBz	Reference
Grain	India, 1989	3	0.03 (0.01-0.04)	30
	Poland, 1981-85	–	0.6	16
Rice	Thailand, 1989-90	1	0.12	32
	Vietnam, 1991*	3	0.03 (<0.01-0.05)	33
Legumens	India, 1989	4	0.07(0.02-0.16)	30
	Thailand, 1989-90	2	0.02 (<0.01-0.03)	32
	Vietnam, 1991	6	0.04 (<0.01-0.18)	33
Vegetables	Poland, 1981-85	–	0.9 (0.3-1.4)	16
Potatoes	Poland, 1981-85	–	0.3	16
Vegetable oil	India, 1989	5	1.5 (0.09-2.8)	30
	Thailand, 1989-90	4	0.016 (0.08-0.35)	32
	Vietnam, 1991	1	1.2	33
Spices	India, 1989	5	0.22 (<0.01-0.54)	30
Butter	India, 1989	4	1.7 (0.86-2.4)	30
	Thailand, 1989-90	1	1.9	32
	Vietnam, 1990-91	2	5.0 (0.2-2.4)	30
	Germany, 1972	97	93	10
	Germany, 1980	267	27	10
	Germany, 1983	238	21	10
	Czechoslovakia, 1976-77	36	500	10
	Czechoslovakia, 1981-83	47	78	10
	Czechoslovakia, 1991	53	14	10
Lard	Poland, 1987-88	3(150)	3.7 (2.0-4.6)	31
	Thailand, 1989-90	1	0.4	32
Cattle fat	Poland, 1987	2(100)	5.9 (5.6-6.1)	31
Sheep fat	Poland, 1988	1(12)	14	31
Rabbit fat	Poland, 1988	1(10)	6.8	31
Duck fat	Poland, 1988	1(32)	15	31
Geese fat	Poland, 1988	1(18)	4.8	31
Poltry fat	Thailand, 1990	2	19(14-23)	32
Turkey fat	Poland, 1988	1(10)	18	31
Wild boar fat	Poland, 1988	1(50)	8.3	31
Roe-deer fat	Poland, 1988	1(50)	10	31
Stalk fat	Poland, 1988	1(50)	10	31
Animal fat	Vietnam, 1990-91	3	0.41 (0.29-0.65)	33
Meat	Vietnam, 1990-91	2	0.11 (0.03-0.18)	33
Pork	Spain, 1991	18	0.5	34
Caviar	Vietnam, 1990-91	3	3.8 (1.9-7.2)	33
Shrimps	Vietnam, 1990-91	1(4)	0.03	33
Fish and shrimps	India, 1989	42	0.07 (<0.01-0.55)	30
Crab	Vietnam, 1990-91	1(3)	0.17	33

* Data include rice imported to Poland in 1989

Table 2. Assessment of dietary intake of hexachlorobenzene in Poland.

Food item	Concentration ^a µg/kg	Intake (µg per capita/year)						
		1970	1975	1980	1985	1990	1995	1996
Cereals	0.6	70	72	76	71	69	72	72
Rice	0.05	0.11	0.10	0.16	0.10	0.13	0.13	0.13
Potatoes	0.3	57	52	47	43	43	41	41
Vegetables	0.9	100	98	91	95	107	108	104
Fruits ^b	0.9	30	31	34	26	26	37	45
Meat and meat products								
– cattle	1.2	19	19	22	19	21	20	20
– pork	0.74	22	30	26	22	27	25	26
– poultry	2.5	19	25	28	18	25	23	23
– other	9.8	20	25	22	25	25	24	43
– variety meats ^c	2.0	7.8	10	9.8	8.8	10	9.2	9.4
Animal fat	3.7	30	30	30	26	30	27	26
Vegetables fat	0.93	6.1	7.0	7.3	7.4	7.1	13	14
Fish*		34	31	30	22	16	19	20
Butter**	5.9	35	44	53	50	46	22	23
Milk***	0.1	26	26	26	27	24	20	20
Eggs	2.5	33	38	40	39	34	28	31
Total intake (µg/per capita/year)		509	538	542	499	510	488	518
Total intake (µg/per capita/day)		1.4	1.5	1.5	1.4	1.4	1.3	1.4

Note:^a - Based on a selected data from Table 1; ^b - received that pollution with HCBz is of the same magnitude as of the vegetables in Poland; ^c - an average value for a various kinds of meat of cattle, pork, poultry; * - dietary intake of HCBz taken from [33]; ** - assumed as fat (100%); *** - average fat content is 2%.

[5,14-29]. The aim of this work is to estimate a magnitude of dietary intake of hexachlorobenzene with total food by a general population in Poland.

Materials and Methods

Intake of HCBz with food in Poland was assessed, taking into account available data on degrees of food contamination by this compound and food consumption rates (Tables 1 and 2). This way of advance follows guidelines by the World Health Organisation [50] and was used earlier in some other studies [51-53].

An assessment of a degree of contamination with HCBz of cattle, pork, poultry and some other kinds of meat is based on the assumption of the existence of dynamic equilibrium of HCBz between different pools of body lipids and on absolute values of residue concentrations quantified in adipose fat of slaughtered and game animals in Poland [31]. It is assumed that the fat content of a definite kind of meat flesh is about 20%. In the case of butter (to simplify calculations it is assumed that lipid content of butter is 100%) and cow milk (lipid content 2%) a degree of contamination with HCBz was assessed taking into account data on average concentrations of HCBz quantified in the adipose fat of cattle. On the other hand, the case of hen eggs (assumed 20% lipid) was based on arithmetic mean weighed concentrations of HCBz quantified in adipose fat of poultry in Poland [31].

The data on the intake rate of HCBz through Baltic fish by Poles was taken from another paper [13].

In Poland in the past there were no extra episodes of HCBz spills or contamination of the environment or food resources with this chemical. A relatively higher degree of pollution with HCBz of soils in the area of Silesia and the city of Krakow can be explained by high industrialization - which is usually connected with higher emissions (incineration, combustion, metallurgy, metal reclamation, chemical industry) of this compound as well as higher urbanization.

In Poland there is a lack of large point sources of environmental pollution by HCBz. Also, the import quotas of this compound in the past were small. Additionally, HCBz poses favourable physico-chemical characteristics enabling its relatively easy volatilization from land surfaces and is prone for atmospheric transportation. So, in our assessment of daily dietary intake of HCBz in 1970-1996 we assumed that a degree of contamination in food of plant and animal origin by this compound (except Baltic fish) remained constant with no clear time-trend.

Results and Discussion

Table 1 presents available data on residue concentrations of HCBz in food in Poland and in some other countries available until 1996. When discussing residue con-

centrations of persistent contaminants in food, accuracy of the presented data is a key factor. It is known that due to a low recovery rate (< 50%) residue data on HCBz obtained after clean-up of the sample extract using Florisil gel and n-hexane or light petroleum as eluting agents are unreliable [21]. The recovery rate of HCBz is around 100% when using Florisil gel as adsorbent agent and after elution with a solvent or a mixture of solvents that are much more polar than n-hexane or light petroleum. Only data obtained using capillary gas chromatography for separation and characterized by adequate recovery rate of HCBz (> 90%) were considered and are included in Table 1. Because of the above reasons the number of data available on residue concentrations of HCBz in Polish food until 1996 is highly limited. Among others there is no validated data available on HCBz residues in milk and milk products as well as for others food items including some meat products.

Food in Poland usually contains residues of HCBz but the concentrations are very small. Edible animal fats, including butter usually contain HCBz at greater concentrations than other food products, and this kind of food home produced in Poland contains HCBz at concentrations below 20 µg/kg (Table 1). The data gathered in Table 1 indicate that food resources in the former East Germany (annualized mean up to 93 µg/kg) and Czechoslovakia (annualized mean up to 500 µg/kg) were periodically relatively highly contaminated with HCBz. The European food, when compared to some countries in Asia, seemed to be more contaminated with HCBz, on the average (Table 1).

The intake of HCBz in Poland in 1970-1996 was estimated at about 1.3 to 1.5 µg *daily per capita* (Table 2). Meat and processed meat products, animal fats and butter altogether are the main source of HCBz in total diet of Poles. There are observed some fluctuations of the intake rate of HCBz with butter (Table 2), that are a result of variable and periodically decreased (1995-1996) consumption figures for this dairy product [52]. The dairy products (butter, milk and eggs) altogether are an important source of HCBz in the total diet of Poles as are meat and processed meat products or vegetables. Fish and processed fish products, however, contain greater concentrations of HCBz [13], when compared to other food items (Table 1), because of small consumption rates in Poland (6.1-8.1 kg *per capita* annually in 1970-1997) are only a minor source of this compound in the total diet of Poles (Table 2). In Poland in 1970-1996 cereals, potatoes and vegetables were as important a source of HCBz intake as meat and processed meat products. A relatively greater intake rate of HCBz with cereals, potatoes, vegetables and butter when compared to other food items (Table 2) is due to a relatively high consumption figure of such food in Poland. Additionally, HCBz nowadays is a compound contaminating the environment on a global scale, and its presence in a gas phase in the atmosphere enables absorption by the wax layer (cuticle) of the plant biomass, and subsequent bioaccumulation and transport through the food webs of herbivorous, omnivorous and carnivorous animals is a common feature.

Assessed intake rates of HCBz with total diet in Poland seem to correlate well with the available data on HCBz body burden of Poles, which is very low [1, 2].

Grains, potatoes, vegetables, fruits, meat and processed meat, fish and dairy-products are almost equivalently important as a source of HCBz in daily total diet of Poles. Table 3 reviews data on intake rates of HCBz with food obtained in various countries and when applying different intake rate calculation strategies (duplicate portion studies, total diet "market basket" studies, selective studies of individual foods). On the average, the daily dose of HCBz taken with food in Poland is, with some small exceptions, similar as reported in several other European countries, USA or Canada (Table 3). The data obtained in this study have indicated that assessed daily dietary intake rates of HCBz in Poland is four-fold lower than a tolerable dose in force in the United States of America.

Table 3. Average daily intake of HCBz in various countries

Country	HCBz (µg <i>per capita</i>)	Reference
India, 1989	0.13	30
Thailand, 1989	0.08	35
Vietnam, 1990	0.10	33
Japan, 1977	0.20	36
Japan, 1985	0.17	36
Japan, 1992-1993	0.15	37
Australia, 1970	47-100	38
Australia, 1990-1992	0.27	39
USA, 1982	1.1-2.0	40
USA, 1987	0.07	48
Canada, 1976-78	<1	38
Canada, 1985	0.04	41
Sweden, 1975	0.72	42
Sweden, 1990	0.30	42
Finland, 1983	1.7	43
Italy, 1971-1972	4.2	44
Italy, 1978-1984	1.2	44
Spain, 1979-1980	1.0	45
Spain, 1980-1981	16	45
Switzerland, 1971-1972	4.5 (2.2-7.4)	46
Switzerland, 1981-1983	1.1	47
The Netherlands, 1976-1978	1	38
Poland, 1970-1996	1.3-1.5	This study
FAO/WHO ADI*	36**	39
EPA RfD*	5	49

Explanation: * - Allowable daily intake (ADI) and RfD - originally expressed in mg/kg body weight daily, in this work, for the purpose of comparison, shown in µg *per capita* daily (the body weight 60 kg); (RfD: reference dose; dose which is not a cause a visible symptoms of poisoning when regularly used, divided by safety coefficient - usually by 100; a term analogous to ADI, and introduced by U.S. EPA, quote after [35]); ** - in 1990 ADI for HCBz was abolished.

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