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Dietary intake of hexachlorobenzene in Catalonia, Spain

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Abstract

To assess the dietary intake of hexachlorobenzene (HCB) by the population of Catalonia, Spain, a total-diet study was carried out. Concentrations of HCB were determined in food samples randomly acquired in seven cities of Catalonia between June and August 2000. A total of 11 food groups were included in the study. HCB levels were determined by HRGC/HRMS. Estimates of average daily food consumption were obtained from recent studies. HCB intake was estimated for five population groups: children (aged 4 to 9 years), adolescents (aged 10 to 19 years), male and female adults (aged 20 to 65 years), and seniors (aged >65 years). In general, HCB residues in foods were rather low excepting dairy products with a mean concentration of 0.869 ng/g wet weight. Total dietary intakes of HCB (microgram per kilogram body weight/day) were the following: children (0.0064), adolescents (0.0031), female adults (0.0025), male adults (0.0024) and seniors (0.0019). All these values are considerably lower than the WHO tolerable daily intake (TDI), which is 0.17 $\mu g\ kg^{-1}\ day^{-1}$ for non-cancer effects and 0.16 $\mu g\ kg^{-1}\ day^{-1}$ for neoplastic effects in humans.

Keywords: Hexachlorobenzene; Food; Total diet; Dietary exposure; Health risks

1. Introduction

In recent years, monitoring programs to determine the occurrence of chemical contaminants in foodstuffs and to assess health risks derived from human consumption have been developed in various countries. The concentrations of residues of a number of persistent organic pollutants (POPs) have been measured (Kiviranta et al., 2001; Tsutsumi et al., 2001; Bocio et al., 2003). Among these pollutants, organochlorines pesticides such as DDTs, hexachlorocyclohexanes, chlordane compounds, hexachlorobenzene (HCB) etc were also determined (Lázaro et al., 1996; Urieta et al., 1996; Nakata et al., 2002; Newsome et al., 2000).

In Spain, little information is available for HCB contamination of foods (Herrera et al., 1994; Lázaro et al., 1996; Urieta et al., 1996; Badia-Vila et al., 2000). HCB is formed as a by-product during the manufacture of chemicals used as solvents, other chlorine-containing compounds and pesticides. Moreover, until recent decades HCB was widely used as a pesticide. According its chemical characteristics, HCB tends to remain in the environment for a long time breaking down

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very slowly (Meharg et al., 2000). For non-occupationally exposed populations, contaminated food, specifically fatty food, is the main way of HCB exposure (Fitzgerald et al., 2001; ATSDR, 2002; Hanaoka et al., 2002). In addition to potential damage on various tissues following chronic exposure to this pollutant, the US EPA has determined that HCB is a possible human carcinogen, while the IARC has classified HCB as possibly carcinogenic to humans (Group 2B) (ATSDR 2002).

During the period 2000–2002, an extensive study has been launched in Catalonia (Spain) to determine the presence of toxic metals and POPs. The concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCNs), and HCB in foods have been measured. The dietary intake of these contaminants was also estimated. Samples taken of each food item were individually analyzed in order to obtain detailed information on the concentrations of the contaminants in foods frequently eaten by the population of Catalonia. To estimate the dietary intake of the pollutants, the results were combined with food consumption data from Catalonia. The dietary intake of metals (Llobet et al., 2003a), PCDD/Fs (Llobet et al., 2003b), PCBs (Llobet et al., 2003c), PBDEs (Bocio et al., 2003) and PCNs (Domingo et al., 2003) were recently reported. In this paper, we present the estimated daily intake of HCB by the population of Catalonia. Estimated intake was determined for individual from five groups: children (aged 4 to 9 years), adolescents (aged 10 to 19 years), male and female adults (aged 20 to 65 years), and seniors (aged >65 years). The current results were compared with international guidelines applicable to HCB, as well as with previously reported data from a number of countries.

2. Materials and methods

2.1. Sampling

Between June and August 2000, food samples were randomly obtained in local markets, big supermarkets, and grocery stores from seven cities (Barcelona, Tarragona, Lleida, Girona, L'Hospitalet de Llobregat, Badalona and Terrassa) of Catalonia, Spain, which have populations between 150 000 and 1 800 000 inhabitants. For collection of samples two groups were made up. The first group included: meat of beef (steak, hamburger), pork (loin, sausage), chicken (breast) and lamb (steak); fish (hake, sardine) and shellfish (mussel); vegetables (lettuce, tomato, potato, green beans, cauliflower); fresh fruits (apple, orange, pear), and eggs. The second group included cow milk (whole, semi-skimmed) and dairy products (yogurt, cheese); cereals (bread, pasta, rice); pulses (lentils, beans); fats (margarine) and oils (olive, sunflower); tinned fish (tuna, sardine), and meat products (ham, hot dogs, salami). Because in the first group most products are usually retailed, their origins could be very diversified in the different cities. Therefore, in that group four composite samples were analyzed for each food item. Each composite was made up by 10 individual samples. In contrast, most food items included in the second group corresponded to brands/trademarks that could be obtained in many different places. Consequently, in this group only two composite samples were analyzed for each food item. Each composite was made up by 8 individual samples. A total of 108 samples were analyzed for HCB concentrations.

2.2. Analytical methods and instrumentation

Prior to extraction, samples were homogenized. Between 5 and 10 g of the freeze-dried solid sample were mixed with small amounts of sodium sulfate and spiked with $^{13}C_6$-HCB. Samples were extracted with the following organic solvents for 24 h (Soxhlet-extraction): toluene for vegetables, fruits, rice, pulses, pasta, eggs and milk and dairy products; hexane:dichloromethane (1:1) for meat and meat products and fresh fish and seafood, and petrolether for tinned fish. For oil and margarine samples, 2 g were dissolved in hexane, spiked with internal standards as described above and immediately used for the clean-up steps.

The clean-up procedure and fractionation of each sample aliquot was carried out using adsorption chromatography and gel permeation chromatography. A multi-step-clean-up was performed using a multilayer silica column (from top to bottom: sodium sulfate, silica, silica/sulfuric acid, silica, silica/potassium hydroxide, silica), alumina columns and gel permeation columns (BioBeads SX3). The final step was reduction of the HCB-containing fraction to the analytical needed volume. Prior to HRGC/HRMS analysis, a ^{13}C-labeled PCDD-standard was added for calculation of recovery ratios.

The cleaned extract was analyzed by HRGC/HRMS using a Fisons CE 8065 GC coupled with a VG AutoSpec Ultima system (EI and multiple ion detection (MID) mode, resolution $\geq$10.000). A 60-m DB-XLB-column (0.32 mm i.d., 0.25 μm Df) was used. HCB quantification was carried out using internal standards. Recovery ratios were in the range 50–126%, with a mean value of 92%. The detection limit was 5 ng/kg.

2.3. Dietary exposure estimates

The daily intake of HCB from each food item was calculated by multiplying the respective concentration in each food by the weight of that food group consumed by an average individual from Catalonia (Capdevila et al., 2000; Cucó et al., 2001). Finally, total dietary intake was obtained by summing these products for all food groups. For calculations, when HCB concentration was under the limit of detection (LOD), the value was assumed to be at one-half of the detection limits (ND = 1/2 LOD).

3. Results and discussion

The concentrations of HCB in a number of food samples (composites) collected in Catalonia are summarized in Table 1. Results are given in nanogram per gram wet weight. In addition, for those foodstuffs whose lipid content was higher than 1%, results are also expressed in nanogram per gram lipid. In general terms, HCB residues in foods were rather low excepting dairy products with a mean concentration of 0.869 ng/g wet weight. The most important contribution to this comparatively high value corresponded to cheese. Composites of cheese samples were based on 10 different commercial brands of semi-cured cheese with an average fat content of 30.4%. In contrast, the lowest HCB concentrations were detected in pulses (0.0006 ng/g wet weight) and fruits (0.0007 ng/g wet weight).

In a previous study, Herrera et al. (1994) determined the concentrations of HCB in Spanish meat products and meat of different species. On a fat basis, HCB levels averaged 49 μg/kg in lamb, varied between 8 and 18 μg/kg in pork and beef products, and amounted to 26 μg/kg in fresh poultry sausages. In a subsequent study of the same group, in which organochlorine pesticide residues were determined in total diet samples from Aragón (Spain), HCB contamination was found to be very low (mean concentration: 1.1 ng/g wet weight; maximum: $<$10 ng/g wet weight) (Lázaro et al., 1996). The highest mean HCB level was found in eggs (1.6 ng/g) followed by legumes (0.2 ng/g) and cereal-based products (0.2 ng/g). However, samples of dairy products, and oils and fats were not analyzed. In the current study, these samples concurrently with those of fish and shellfish and eggs were the food groups showing the highest HCB concentrations.

In other surveys carried out during the 1980s, Gartrell et al. (1985, 1986a,b) detected HCB in a variety of foods including dairy products, meats, fish, and oils and fats, with the highest mean levels corresponding to oils and fats (0.9 ng/g) and meat and fish, both with 0.2 ng/g. In a recent survey, HCB was detected only in 4.8% food composites from six Canadian cities (Newsome et al., 2000), while more recently HCB was detected in 59% of a wide variety of foodstuffs collected in Shanghai and its vicinity (China) (Nakata et al., 2002). In the present study, HCB was detected in 86% of the samples. Probably, it is due to a better sensitivity of the present analytical method rather than a widespread HCB presence in the Spanish environment.

Table 2 shows data on daily consumption of the analyzed foodstuffs classified into 11 groups: meat and meat products, fish and seafood, vegetables, tubers, fruits, eggs, milk, dairy products, cereals, pulses and oils and fats. Data are given for children, adolescents, male adults, female adults and seniors living in Catalonia. The dietary intake of

Table 1
Concentrations of hexachlorobenzene (nanogram per gram wet and lipid weight) in foodstuffs collected in Catalonia, Spain

	Wet wt.	Lipid wt.		Wet wt.	Lipid wt.
Meat and meat products			*Fish and seafood*		
Beef steak (4)	0.038	0.282	Hake (4)	0.038	2.103
Beef burger (4)	0.266	1.809	Sardine (4)	0.781	8.305
Pork loin (4)	0.075	0.283	Mussel (4)	0.025	0.887
Pork sausage (4)	0.353	1.114			
Breast chicken (4)	0.007	0.165	*Tinned fish*		
Lamb steak (4)	0.236	2.005	Tuna (2)	0.053	0.439
Ham (2)	0.023	0.217	Sardine (2)	0.385	3.134
Hot dogs (2)	0.319	1.139			
Salami (2)	0.241	0.821	*Fruits*		
			Apple (4)	0.0007	–
Vegetables and tubers			Orange (4)	0.0016	–
			Pear (4)	0.0005	–
Lettuce (4)	0.005	–			
Tomato (4)	0.007	–	Eggs	0.182	1.508
Cauliflower (4)	0.005	–			
Green beans (4)	0.010	–	*Pulses*		
Potato (4)	0.001	–	Lentils (2)	0.0005	0.031
			Beans (2)	0.0006	0.040
Cereals					
French bread (2)	0.004	0.231	*Oils and fats*		
Soft bread (2)	0.026	2.024	Olive oil (2)	0.085	0.086
Rice (2)	0.001	–	Sunflower oil (2)	0.154	0.154
Pasta (2)	0.002	0.179	Margarine (2)	0.019	0.023
Milk			*Dairy products*		
Whole (2)	0.017	0.453	Yogurt (2)	0.070	2.705
Semi-skimmed (2)	0.009	0.538	Cheese (2)	1.668	5.488

In parentheses, number of analyzed samples.

HCB for each of these five age and/or sex groups are also shown. The total daily intakes of HCB for the same five groups are also depicted in Fig. 1.

When the dietary HCB intake was expressed in microgram per day, the highest value corresponded to the groups of adolescents (0.168) and male adults (0.166), while the lowest intake of this contaminant corresponded to seniors (0.117). For all five groups, the consumption of dairy products was the main responsible of these intakes, which contributed with the minimum percentage (53.4%) for seniors and the maximum (64.8%) for children. Other food groups with a remarkable contribution to HCB intake were meat and meat products, and fish and seafood. These groups were followed at a great distance by eggs, and oils and fats. By contrast, the lowest contributions to total HCB dietary intake corresponded to pulses, vegetables and tubers, and fruits. This is an important result, as vegetables and fruits are items of a great consumption in Catalonia, a region in which predominates the Mediterranean diet.

When the total dietary intake of HCB was expressed in microgram per kilogram body weight/day, the decreasing ranking by age/sex groups was found to be the following: children (0.0064), adolescents (0.0031), female adults (0.0025), male adults (0.0024) and seniors (0.0019). All these values are considerably lower than the WHO tolerable daily intake (TDI), which is 0.17 $\mu g\ kg^{-1}\ day^{-1}$ for non-cancer effects and 0.16 $\mu g\ kg^{-1}\ day^{-1}$ for neoplastic effects in humans (ATSDR, 2002). According to this, the

Table 2
Food intake (gram per day) and hexachlorobenzene (HCB) intake (nanogram per day) through the diet of children, adolescents, male adults, female adults and seniors of Catalonia, Spain

Food group	Children		Adolescents		Male adults		Female adults		Seniors	
	Food intake	HCB	Food intake	HCB	Food intake	HCB	Food intake	HCB	Food intake	HCB
Meat and meat products	140	24.25	167	28.92	185	31.98	125	21.65	114	19.74
Fish and seafood	51.5	13.20	62	15.90	92	23.59	79.3	20.34	80	20.51
Vegetables	125	0.73	162.5	0.94	226	1.56	202.3	1.17	189.5	1.10
Tubers	63.5	0.08	76.5	0.10	74	0.10	57	0.07	69.5	0.09
Fruits	196	0.14	202	0.14	239	0.17	226.6	0.16	268	0.19
Eggs	26.5	4.83	25.5	4.65	34	6.26	23.3	4.25	22.5	4.10
Milk	309	3.99	266.5	3.44	217	2.80	253.3	3.27	253.5	3.27
Dairy products	114	99.10	122.5	106.49	106	92.15	91.3	79.40	72	62.59
Cereals	200.5	2.13	221	2.34	206	2.19	138.3	1.47	156.5	1.66
Pulses	25.5	0.02	24	0.01	24	0.01	22.6	0.01	22	0.01
Oils and fats	33.5	2.88	36	3.1	41	3.53	31	2.67	29	2.49
Total HCB intake										
(μg/day)		0.153		0.167		0.166		0.136		0.117
(μg kg^{-1} day^{-1})		0.0064		0.0031		0.0024		0.0025		0.0019

intakes of HCB estimated in the present survey are equivalent to 3.76, 1.82, 1.47, 1.41 and 1.12% of the TDI (non-cancer effects) for children, adolescents, female adults, male adults and seniors, respectively.

A summary of HCB concentrations in foodstuffs of various countries is presented in Table 3. The current intakes are remarkably lower than those reported in the 1980s for some industrialized countries: Finland, 0.0242 μg kg^{-1} day^{-1} (Moilanen et al., 1986), the Netherlands, 0.0143 μg kg^{-1} day^{-1} (Greve, 1986), or the UK, 0.0029 μg kg^{-1} day^{-1} (Burton and Bennett, 1987). In turn, HCB intakes in the USA estimated for the Total

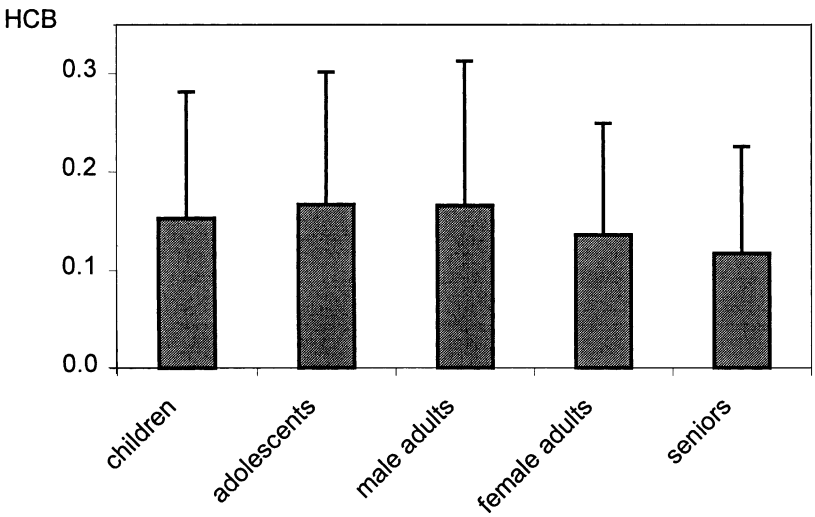

Fig. 1. Total dietary intake (mean ± S.D.) of hexachlorobenzene (microgram per day) by the population of Catalonia, Spain, according to age/sex groups.

Table 3
A summary of dietary intake of HCB in a number of regions and countries

Country	HCB intake* $\mu g\ kg^{-1}\ day^{-1}$	Reference
The Netherlands	0.0143	Greve, 1986
Finland	0.0242	Moilanen et al., 1986
United Kingdom	0.003	Burton and Bennett, 1987
Sweden	0.005	Vaz, 1995
The Netherlands	0.0014–0.0031	Brussaard et al., 1996
Basque Country, Spain	0.0029	Urieta et al., 1996
Shanghai region, China	0.008	Nakata et al., 2002
Catalonia, Spain	0.0024	This study

* Body weight was estimated to be 70 kg excepting in the Chinese study in which it was 60 kg.

Diet Analyses declined from 0.0011 and 0.0006 $\mu g\ kg^{-1}\ day^{-1}$ in 1988 to 0.0004 and 0.0002 $\mu g\ kg^{-1}\ day^{-1}$ in 1991, for 14–16-year-old males and 60–65-year-old women, respectively, (ATSDR, 2002). These values are of the same order of magnitude than the present results.

In the 1990s, Vaz (1995) reported a dietary HCB intake in Sweden of 0.005 $\mu g\ kg^{-1}\ day^{-1}$ via foods of animal origin, whereas in the Netherlands the results of the Dutch National Surveillance System showed a dietary HCB intake between 0.0014 and 0.0031, which is similar to that found in our survey. Very similar to the current dietary HCB intake was also that reported after a wide food surveillance carried out in another Spanish region, the Basque Country, between 1988 and 1990. A HCB intake of 0.0029 $\mu g\ kg^{-1}\ day^{-1}$ was found (Urieta et al., 1996). In contrast to these results, a lower HCB intake was recently reported by Nakata et al. (2002) for the region of Shanghai, China. An average daily intake of 0.046 μg HCB/person/day (0.0007 $\mu g\ kg^{-1}\ day^{-1}$ for a 70-kg person) was found. The authors remarked the very low levels of vegetables and cereals (most under detection limit) and the high consumption of these foods in China, which would explain the low HCB intake through the diet. However, in a recent Japanese study in which the specific dietary intake of HCB was not reported, a borderline significant correlation between HCB levels in serum and fish intake was observed (Hanaoka et al., 2002). In the present survey, fish and seafood had a notable contribution to the total dietary intake of HCB.

Taking into account international guidelines applicable to HCB, the current dietary intake of this contaminant by the population of Catalonia seem to pose very small health risks. Individuals consuming great amounts of certain foodstuffs: meat, fish and especially cheese, could result in an increased HCB exposure through the diet. However, the safety margin to prevent health risks would be still high.

With respect to other potential sources of HCB exposure for humans, it can be interesting to remark that the highest plasma levels of HCB even described were recently found in the inhabitants of a Catalonian village. However, these concentrations were attributed to the high air HCB levels in the area due to the activity of a chemical plant that manufactures organochlorine compounds (Herrero et al., 1999; Sala et al., 1999; Ballester et al., 2000).

In summary, although the present HCB intakes were similar to those reported for a number of countries, HCB is carcinogenic in humans and as recently noted by Hanaoka et al. (2002), this environmental contaminant has been pointed out to act as a dioxin-like compound, being its potential health effects at low-dose exposure still unclear.

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