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Polybrominated Diphenyl Ethers (PBDEs) in Foodstuffs: Human Exposure through the Diet

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Polybrominated diphenyl ethers (PBDEs) are used as flame retardants in a variety of materials, including synthetic polymers and textiles. Although these chemicals have been detected in environmental samples and human tissues, there is little information about human exposure to PBDEs through the diet. In the present study, we determined the concentrations of PBDEs in a number of food samples acquired in Catalonia (Spain) during 2000. The dietary intake of PBDEs was estimated for the general population living in this Spanish region. The highest PBDE concentrations were found in oils and fats, fish and shellfish, meat and meat products, and eggs, while the lowest levels corresponded to fruits, vegetables, and tubers. The dietary intake of PBDEs for an adult male was 97.3 ng/day (assuming not detected (ND) = $^1/_2$ limit of detection (LOD)) or 81.9 ng/day (assuming ND = 0) The greatest contribution to these values corresponded to fish and shellfish, with approximately one-third of the total intake. TetraBDEs and pentaBDEs were the homologues showing the highest percentages of contribution to the sum of total PBDEs. The comparison of the current dietary intake with the suggested lowest observed adverse effect level value of 1 mg/kg/day for the most sensitive endpoints for toxic effects of PBDEs results in a safety factor over 5 orders of magnitude in relation to PBDE exposure from food.

KEYWORDS: Polybrominated diphenyl ethers (PBDEs); levels in food; dietary intake; general population; Catalonia, Spain

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are dicyclic aromatic ethers with highly lipophilic characteristics. They are widely used as flame retardants, being added to different materials, such as plastics for electronic appliances, paints, and textiles, to prevent them for catching fire. Technical PBDE products are manufactured as mixtures of penta-, octa- and decabromodiphenyl ethers, corresponding to the average bromine content (*1*, *2*). In the mid-1990s, the annual world production of flame retardants was 600,000 metric tons. From this total, about 50,000 tons were PBDEs (*3*).

PBDEs are structurally similar to well-known environmental pollutants such as dioxins and PCBs. Due to their relatively low reactivity and high hydrophobicity, they are persistent environmental substances, and for certain congeners, bioaccumulative. The tetra- to hexabrominated congeners exhibit the greatest bioaccumulation and toxicological potentials (*1*, *2*, *4*). Because of the use of these chemicals as flame retardants, they may migrate from the products during their entire life span.

In recent years, PBDE residues have been found in sediments, marine mammals, fish, and bird eggs (*1*, *2*, *5*−*7*). Moreover, PBDEs have also been identified in human blood, adipose tissue, and breast milk (*8*−*12*). There is evidence that, in recent years, PBDE concentrations are rising in human tissues and biota, with a rapid increase (*1*, *4*, *13*). For example, it has been reported that milk from Swedish mothers showed an exponential increase from 1972 to 1997, with a rate of increase that doubled with 5-year increments (*14*).

On the basis of available toxicity data and structural and mechanistic similarities with polychlorinated biphenyls (PCBs), the most sensitive toxic endpoints for PBDEs are probably thyroid hormone disruption and decreases of free thyroxin hormone (T4), neurobehavioral toxicity, and for some congeners, possibly cancer (*1*, *4*, *15*, *16*). However, unlike dioxins and PCBs, PBDEs are not strong inducers of the aryl hydrocarbon hydroxylase receptor (AhR) (*3*).

The environmental fate of PBDEs appears to be analogous to the fate of other structurally similar environmental pollutants such as PCBs, for which the main route of human exposure is via food (*5*). Although sources other than food may also

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Table 1. PBDE Concentrations (ng/kg wet weight) in Food Samples Collected in Catalonia, Spain[a]

food group	vegetables (n=8)[b]	tubers (n=2)	pulses (n=2)	cereals (n=4)	fruits (n=6)	fish and shellfish (n=8)	meat and meat products (n=15)	eggs (n=2)	milk (n=2)	dairy products (n=2)	fats and oils (n=3)
tetraBDE	4.0 (3.9)	0.5 (0)	2.3 (2.0)	2.2 (0)	0.4 (0)	158.3 (158.2)	23.5 (23.3)	17.3 (17.3)	8.0 (8.0)	10.7 (10.7)	169.7 (169.7)
pentaBDE	1.4 (1.3)	0.5 (0)	0.6 (0)	2.2 (0)	0.4 (0)	115.9 (115.8)	24.9 (24.7)	25.8 (25.8)	5.2 (5.2)	23.4 (23.4)	157.7 (157.7)
hexaBDE	0.4 (0)	0.9 (0)	1.1 (0)	4.5 (0)	0.7 (0)	47.4 (47.0)	13.5 (12.8)	11.9 (11.9)	0.5 (0)	2.0 (0)	139.7 (138.0)
heptaBDE	0.7 (0)	1.8 (0)	2.2 (0)	8.9 (0)	1.4 (0)	5.4 (3.0)	23.9 (22.5)	4.4 (3.3)	1.1 (0)	4.0 (0)	77.0 (73.7)
octaBDE	1.4 (0)	3.7 (0)	17.9 (0)	17.9 (0)	2.9 (0)	6.8 (1.4)	23.4 (19.1)	4.7 (0)	2.1 (0)	7.9 (0)	43.7 (30.3)
sum PBDE	**7.9 (5.2)**	**7.4 (0)**	**10.7 (2.0)**	**35.7 (0)**	**5.8 (0)**	**333.9 (325.3)**	**109.2 (102.4)**	**64.5 (58.3)**	**16.9 (13.2)**	**47.9 (34.1)**	**587.7 (569.3)**

[a] For each food group, two values are given. The upper and lower (in parentheses) values were calculated assuming that when a congener was below the detection limit the concentration was equal to one-half of the respective limit of detection (upper value) or zero (lower value). [b] n = number of composite samples analyzed.

influence total PBDE intake in humans, to date, there is an evident lack of data on these hypothetical sources of human exposure. In a recent extensive review of occurrence, dietary exposure, and toxicology of these chemicals, it was concluded that, although sufficient data were available on PBDE levels in fish, less was known about their concentrations in other major food groups or about possible differences in food levels between countries or regions (*5*).

As for most countries, the levels of PBDEs have not been previously investigated in Spain. During 2000−2002, a wide study has been launched in Catalonia (NE Spain) to determine human exposure from dietary sources to several environmental pollutants. Results about metals, dioxins and furans, and PCBs have been recently reported (*17*−*19*). The main objective of the present study was to measure the concentrations of PBDEs in a number of foods acquired in Catalonia and to calculate the dietary intake of these compounds by the population living in this Spanish region.

MATERIALS AND METHODS

Sampling. From June to August 2000, food samples were randomly acquired in local markets, big supermarkets, and grocery stores from seven cities (Barcelona, Tarragona, Lleida, Girona, L'Hospitalet de Llobregat, Badalona, and Terrassa) of Catalonia, 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 and tubers (lettuce, tomato, potato, green beans, cauliflower); fresh fruits (apple, orange, pear), and eggs. The second group included cow milk (whole, semiskimmed) 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, 2 composite samples were analyzed for each food item. Each composite was made up of 10 individual samples, which were collected in five different places. 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 one composite sample was analyzed for each food item. This composite was made up of eight individual samples of similar weights, which were collected in four different places of a same city. The sums of the tetra-to-octabrominated congeners were determined for each sample. A total of 54 samples were analyzed.

Analytical Methods and Instrumentation. Food samples were homogenized and blended using a domestic mixer. Composite samples were lyophilized previously to analyses of PBDEs, which were performed in accordance to the US EPA 1625 method (semivolatile organic compounds by isotope dilution GC/MS). Prior to extraction, dried samples were homogenized. Five to 10 g of the freeze-dried solid samples were mixed with a small amount of Na_2SO_4 and spiked with a mixture of $^{13}C_{12}$-marked standards. Samples were extracted for 24 h with the following organic solvents (Soxhlet-extraction): toluene for vegetables, fruits, cereals, eggs, milk, and dairy products; hexane/dichloromethane (1:1) for meat, fresh fish, and mussels, and petrolether for fish in oil. For oil and margarine, 2 g of the sample was dissolved in hexane and immediately used for the cleanup procedure. The cleanup procedure and fractionation of the sample aliquot was carried out as a multiple cleanup, using adsorption chromatography, 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 involved the reduction of the PBDE-containing fractions to the volume necessary for the analysis. Recovery rates were calculated against external reference standards. Standards were used to calculate the amounts of PBDE of their own congener group (e.g., tetra- for tetras, penta- for pentas, etc.), with the exception of OctaBDE that was calculated using $^{13}C_{12}$-HeptaBDE #183. The cleaned extract was analyzed by high-resolution gas chromatography/high-resolution mass spectrometry (Fisons CE 800 gas chromatograph coupled with a VG Autospec Ultima system with electronic impact and a multiple ion detection mode (with a resolution of >10 000)). A DB-XLB column (60 m, 0.32 mm i.d., 0.25μm dF) was used. The following internal standards were used to quantify PBDEs.: $^{13}C_{12}$-TetraBDPE #47, $^{13}C_{12}$-PentaBDPE #99, $^{13}C_{12}$-HexaBDPE #153, $^{13}C_{12}$-HexaBDPE #154, $^{13}C_{12}$-HeptaBDPE #183. One ng of each standard was used. Mean recovery rates ranged from 79%, for the sum of pentaBDEs (range 55−106%) and hexaBDEs (range 54−115%), to 85% (54−114%) for the sum of heptaBDEs. The detection limits varied from 5 to 40 ng/kg dry weight, depending on the specific food and the respective congeners.

Dietary Exposure Estimates. Average daily consumption data were obtained from recent studies carried out in Catalonia (*20*, *21*). PBDE intake was estimated for each food group assuming that when a congener was below the detection limit, the concentration was either equal to zero (not detected (ND) = 0) or one-half of the respective limit of detection (LOD) (ND= $^1/_2$ LOD). Results are presented for both assumptions.

RESULTS AND DISCUSSION

The concentrations of PBDEs in foodstuffs acquired in various locations of Catalonia in 2000 are given in **Table 1**. Results (wet weight) show the sum levels of tetra-, penta-, hexa-, hepta-, and octaBDEs, as well as the sum concentrations of total PBDEs (tetra to octa). Data are presented for 11 food groups. Calculations assuming that ND = $^1/_2$ LOD and ND = 0 are given for each food group. The highest concentration of total PBDEs was found in oils and fats (587.7−569.3 ng/kg), followed by fish and shellfish (333.9−325.3 ng/kg), meat and meat products (109.2−102.4 ng/kg), and eggs (64.5−58.3 ng/kg). In all these groups, a predominance of the homologues tetra- and pentaBDEs, followed by hexaBDEs, was observed in the sum of total PBDEs. By contrast, PBDEs were not detected in the groups of fruits, cereals, and tubers (**Table 1**). On the other

Table 2. PBDE Concentrations in a Number of Food Samples Collected in Catalonia, Spain[a]

food	ng/kg lipid weight	ng/kg wet weight
vegetables		8 (5)
tubers		7 (0)
pulses		11 (2)
cereals		36 (0)
fruits		6 (0)
white fish	2359 (2052)	88 (37)
shellfish	3140 (2961)	88 (83)
tinned fish	2117 (1997)	260 (246)
blue fish	10839 (10804)	1019 (1016)
pork and pork products	597 (565)	172 (166)
chicken	247 (0)	10 (0)
beef and beef products	290 (248)	42 (36)
lamb	261 (182)	31 (21)
eggs	530 (482)	64 (58)
dairy products	677 (557)	48 (34)
whole milk	630 (525)	24 (20)
semiskimmed milk	618 (402)	10 (6)
vegetable oils and fats	805 (795)	804 (794)
margarine	188 (145)	155 (120)

[a] Data were calculated assuming that when a congener was below the detection limit, the concentration was equal to one-half of the respective limit of detection. Values in parentheses were calculated assuming that ND = 0.

hand, the concentrations of PBDEs in a number of food groups are also presented (**Table 2**). Data are given in ng/kg lipid weight and ng/kg wet weight. In both cases, blue fish showed the highest levels, followed by the remaining fish and shellfish items (lipid weight basis) or by oils and fats (wet weight basis). In recent years, a number of investigators have determined PBDE levels in various marine and freshwater species. However, data on PBDE concentrations in other major food groups are very scarce. In a recent review, Darnerud et al. (*5*) reported PBDE levels between 26 and 36 900 μg/kg lipid in various fish species from Swedish lakes and rivers. In the same review, the mean PBDE level in mixed meat products and eggs from Sweden was reported to be between 360 and 420 ng/kg fat.

In a study carried out in freshwater fish from selected locations in Washington state, total PBDE concentrations ranged from 1.4 μg/kg (wet weight) in rainbow trout from a remote spring-fed stream to 1250 μg/kg (wet weight) in mountain whitefish from an urbanized river (*22*). On the other hand, in nine fish species collected from the Inland Sea of Seto (Japan), the concentrations of total PBDEs varied between 110 and 3 300 ng/kg (wet weight) (*23*), a range in which our current results in fish and shellfish are included. In marine fish and blue mussels from southern Greenland, Christensen et al. (*24*) found a total PBDE concentration in fish between 1800 and 12 000 ng/kg (wet weight), depending on location and species, while for mussels, the concentration was 110 ng/kg (wet weight).

Recently, Otha et al. (*25*) determined the concentrations of PBDEs in fish, meat, and vegetables and tubers sold in food markets from Hirakata (Japan). Total PBDE levels in fish tissues ranged from 17.7 to 1720 ng/kg (wet weight). Again, the results of the present study are within this range. The concentrations of PBDEs in vegetables and tubers detected by Otha et al. (*25*) were between 38.4 and 134.0 ng/kg (wet weight), values higher than the current results. Finally, PBDE concentrations in meat varied from 6.25 to 63.6 ng/kg (wet weight) for chicken and pork, respectively (*25*). In contrast to the levels found in vegetables and tubers, our current concentrations in meat are higher.

Table 3 shows data on food intake and dietary intake of PBDEs for a standard male adult of 70-kg body weight. Total

Table 3. Estimated Dietary Intake of PBDEs by the Adult Population of Catalonia, Spain[a]

food group	daily consumption[b] (g)	PBDE intake[c] (ng/day)
vegetables	226 (15.7)	1.8 (1.2)
pulses	24 (1.7)	0.3 (0.05)
cereals	206 (14.3)	7.4 (–)
tubers	74 (5.1)	0.6 (–)
fruits	239 (16.6)	1.4 (–)
fish and shellfish	92 (6.4)	30.7 (29.9)
meat and meat products	185 (12.8)	20.2 (18.9)
eggs	34 (2.4)	2.2 (2.0)
dairy products	106 (7.3)	5.1 (3.6)
milk	217 (15.0)	3.7 (2.9)
fats and oils	41 (2.8)	24.1 (23.3)
total intake	**1444 (100)**	**97.3 (81.9)**
		1.4 (1.2)[d]

[a] Results are given for a male adult of 70-kg body weight. [b] In parentheses, percentages of total consumption. [c] Data were calculated assuming that when a congener was below the detection limit, the concentration was equal to one-half of the respective limit of detection. Values in parentheses, were calculated assuming that ND = 0. [d] Total intake expressed in ng/kg body weight/day.

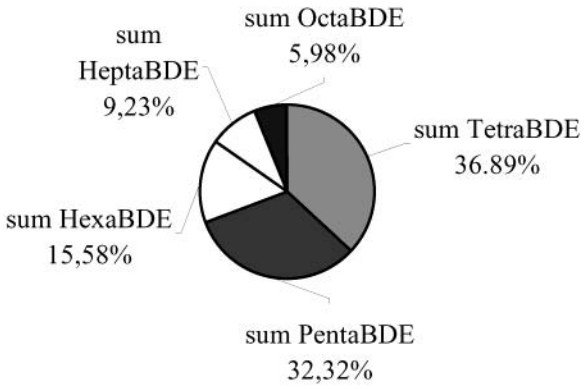

Figure 1. Percentages of contribution of the different congeners to the total dietary intake of PBDEs (ng/day). Calculations assuming ND = 0.

dietary intake was between 97.3 ng/day (ND = $^1/_2$ LOD) and 81.9 ng/day (ND = 0), or between 1.4 and 1.2, when the results are expressed in ng/kg body weight/day. The highest contribution to this intake corresponded to fish and shellfish, with approximately one-third of the total intake, followed by oils and fats and meat and meat products, with a contribution of about one-fourth each. In contrast, the lowest percentages corresponded to vegetables and tubers, fruits, and cereals. Although the dietary consumption of these foodstuffs in Catalonia is very notable, the low fat content of these products, and consequently their comparatively low PBDE content, explain the small contribution of these food groups to the total dietary intake of PBDEs.

The percentages of contribution of each PBDE homologue group to the total dietary intake of these pollutants are depicted in **Figure 1**. The highest contribution to total PBDE intake corresponded to tetraBDEs and pentaBDEs, with approximately one-third each. On the other hand, the estimated PBDE intake (ng/kg body weight/day) for the population of Catalonia, according to sex and age, is shown in **Figure 2**. Total PBDE intake through the diet ranged from 2.6 for boys under 9 years old to 0.9 for males in the group >65 years. For both sexes, the dietary intake of PBDEs was relatively constant from 10 to 65 years.

Information on PBDE dietary intake is very scarce. Searching in the scientific literature, we have only found four studies. A

Table 4. A Summary of Data of Human Exposure to PBDEs through the Diet

country	characteristics of the study	PBDE intake (ng/day)	remarks	reference
Sweden	market basket samples: fish, meat, dairy products, eggs, fats/oils,pastry	51 sum of congeners 47, 99, 100, 153, 154	calculations for intake were done assuming that ND = $^1/_2$ LOD	Darnerud et al. 2001 (5)
Sweden	foods of animal origin, Diet National Swedish inventory	females (18–74 years) mean: 40.8 sum of congeners 47, 99, 100, 153, 154	calculations for intake were done assuming that ND = 0	Lind et al. 2002 (26)
Canada	food basket study, most food samples of animal origin	44		Ryan and Patry 2001 (27)
UK	duplicate diet samples	median: 90.5 sum of congeners 47, 99, 100, 153, 154	calculations for intake were done assuming that ND = 0	Wijsekera et al. 2002 (28)
Catalonia (Spain)	total diet study, 54 samples belonging to 11 food groups	81.9 (lower) 97.3 (upper) sum of tetra- to octaBDEs	ND = 0 ND = LOD	this study

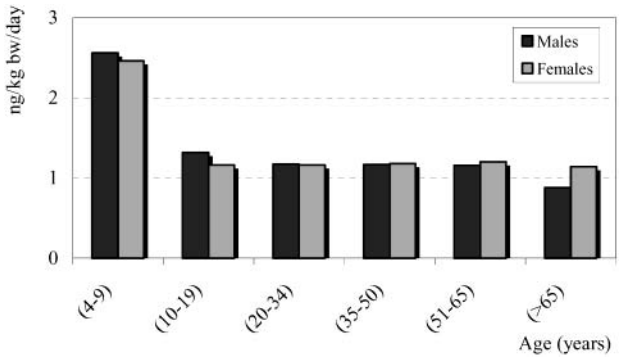

Figure 2. Estimated dietary intake of PBDEs by the general population of Catalonia, Spain, in relation to age and sex. Caculations assuming ND = 0.

summary of results of these studies, together with those of the current survey, are shown in **Table 4**. In an estimation of PBDE exposure from food in Sweden, the dietary intake was 51 ng/day (calculations were done assuming that ND = 1/2 LOD). This intake was higher than that found in a subsequent study, in which only food of animal origin was included (*26*). Total dietary intake for Swedish females (18−74 years old) was 40.8 ng/day. However, in that case, when the concentration of a PBDE congener was below the detection limit, that concentration was assumed to be zero (ND = 0) (*26*). It is important to remark that, in both Swedish studies, only PBDE congeners 47, 99, 100, 153, and 154 were determined. A similar dietary intake, 44 ng/day, was also found for Canadian adults in a food basket study (*27*). About 75% of the daily intake of PBDEs corresponded to meat, while dairy products and fish contributed with approximately 7 and 4%, respectively. Finally, in a recent survey carried out in the United Kingdom, the median dietary intake of PBDE congeners 47, 99, 100, 153 and 154 was 90.5 ng/day (calculations were done assuming that ND = 0) (*28*). This intake is very similar to our current estimated lower intake, 81.9 ng/day.

Although in human tissues and fluids PBDE levels are still lower than those of other environmental contaminants such as PCBs, recent data suggest an increasing trend in human PBDE concentrations over time (*5*, *13*, *14*). Therefore, it is important to establish which are the human health risks derived from the intake of PBDEs through the diet. The following estimations were made for evaluation of PBDE risks in Catalonia. On the basis of the most sensitive endpoints for toxic effects of PBDEs, a lowest observed adverse effect level (LOAEL) value of 1 mg/kg/day was recently suggested as reasonable for compounds or mixtures belonging to the PBDE group (*5*). The comparison of the current dietary intake (1.4−1.2 ng/kg body weight/day) with the suggested LOAEL value of 1 mg/kg/day results in a safety factor over 5 orders of magnitude in relation to PBDE exposure from food. However, in a recent study, Wijesekera et al. (*28*) showed that, in an overall daily exposure to PBDEs, diet and inhalation contributed with 73 and 27%, respectively. This indicates that in addition to special dietary habits, the safety factor can be also notably influenced by other types of PBDE exposure. Taking into account the scarce information on the above, further investigations are clearly necessary.

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LITERATURE CITED

(1) McDonald, T. A. A perspective on the potential health risks of PBDEs. *Chemosphere* **2002**, *46*, 745−755.
(2) Rahman, F.; Langford, K. H.; Scrimshaw, M. D.; Lester, J. N. Polybrominated diphenyl ethers (PBDE) flame retardants. *Sci. Total Environ.* **2001**, *275*, 1−17.
(3) Manchester-Neesvig, J. B.; Valters, K.; Sonzgoni, W. C. Comparison of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in Lake Michigan salmonids. *Environ. Sci. Technol.* **2001**, *35*, 1072−1077.
(4) De Wit, C. A. An overview of brominated flame retardants in the environment. *Chemosphere* **2002**, *46*, 583−624.
(5) Darnerud, P. O.; Eriksen, G. S.; Jóhannesson, T.; Larsen, P. B.; Viluksela, M. Polybrominated diphenyl ethers: occurrence, dietary exposure, and toxicology. *Environ. Health Perspect.* **2001**, *109* (Suppl 1), 49−68.
(6) Dodder, N. G.; Strandberg, B.; Hites, R. A. Concentrations and spatial variations of polybrominated diphenyl ethers and several

organochlorine compounds in fishes from the northeastern United States. *Environ. Sci. Technol.* **2002**, *36*, 146−151.
(7) Lindström, G.; Wingfors, H.; Dam, M.; van Bavel, B. Identification of 19 polybrominated diphenyl ethers (PBDEs) in long-finned pilot whale (Globicephala melas) from the Atlantic. *Arch. Environ. Contam. Toxicol.* **1999**, *36*, 355−363.
(8) Covaci, A.; de Boer, J.; Ryan, J. J.; Voorspoels, P.; Schepens, P. Distribution of organobrominated contaminants in Belgian human adipose tissue. *Environ. Res.* **2002**, *88*, 210−218.
(9) Meneses, M.; Wingfors, H.; Schuhmacher, M.; Domingo, J. L.; Lindström, G.; van Bavel, B. Polybrominated diphenyl ethers detected in human adipose tissue from Spain. *Chemosphere* **1999**, *39*, 2271−2278.
(10) Meironyté-Guvenius, D.; Bergman, A.; Koren, N. Polybrominated diphenyl ethers in Swedish human liver and adipose tissue. *Arch. Environ. Contam. Toxicol.* **2001**, *40*, 564−570.
(11) Sjödin, A.; Patterson, D. G., Jr.; Bergman, A. Brominated flame retardants from U.S. blood donors. *Environ. Sci. Technol.* **2001**, *35*, 3830−3833.
(12) van Bavel, B.; Hardell, L.; Kitti, A.; Liljedahl, M.; Karlsson, M.; Petterson, A.; Tysklind, M.; Lindström, G. High levels of PBDEs in 5% of 220 blood samples from the Swedish population. *Organohalogen Compd.* **2002**, *58*, 161−164.
(13) Ikonomou, M. G.; Rayne, S.; Addison, R. F. Exponential increases of the brominated flame retardants, polybrominated diphenyl ethers, in the Canadian Arctic from 1981 to 2000. *Environ. Sci. Technol.* **2002**, *36*, 1886−1892.
(14) Meironyté, D.; Koren, N.; Bergman, A. Analysis of polybrominated diphenyl ethers in Swedish human milk. A time-related trend study, 1972−1997. *J. Toxicol. Environ. Health* **1999**, *58*, 329−341.
(15) Darnerud, P. O.; Thuvander, A. Effects of polybrominated diphenyl ether (PBDE) and polychlorinated biphenyl (PCB) on some immunological parameters after oral exposure in rats and mice. *Toxicol. Environ. Chem.* **1999**, *70*, 229−242.
(16) Pijnenburg, A. M. C. M.; Everts, J. W.; de Boer, J.; Boon, J. P. Polybrominated biphenyl and diphenyl ether flame retardants: analysis, toxicity, and environmental occurrence. *Rev. Environ. Contam. Toxicol.* **1995**, *141*, 1−26.
(17) Llobet, J. M.; Falcó, G.; Casas, C.; Teixidó, A.; Domingo, J. L. Concentrations of arsenic, cadmium, mercury, and lead in common foods and estimated daily intake by children, adolescents, adults, and seniors of Catalonia, Spain. *J. Agr. Food Chem.* **2003**, *51*, 838−842.
(18) Llobet, J. M.; Domingo, J. L.; Bocio, A.; Casas, C.; Teixidó, A.; Müller, L. Human exposure to dioxins through the diet in Catalonia, Spain: Carcinogenic and non- carcinogenic risks. *Chemosphere* **2003**, *50*, 1193−1200.
(19) Llobet. J. M.; Bocio, A.; Domingo, J. L.; Teixidó, A.; Casas, C.; Müller, L. Levels of polychlorinated biphenyls in foods from Catalonia, Spain: Estimated dietary intake. *J. Food Protect.* **2003**, *66*, 479−484.
(20) Capdevila, F.; Llop, D.; Guillén, N.; Luque, V.; Pérez, S.; Sellés, V.; Fernández-Ballart, J.; Martí-Henneberg, C. Food intake, dietary habits, and nutritional status of the population of Reus-(X): Evolution of the diet and the contribution of macronutrients to energy intake. *Med. Clin. (Barc).* **2000**, *115*, 7−14.
(21) Cucó, G.; Arija, V.; Martí-Henneberg, C.; Fernández-Ballart, J. Food and nutritional profile of high energy density consumers in an adult Mediterranean population. *Eur. J. Clin. Nutr.* **2001**, *55*, 192−199.
(22) Johnson, A.; Olson, N. Analysis and occurrence of polybrominated diphenyl ethers in Washington State freshwater fish. *Arch. Environ. Contam. Toxicol.* **2001**, *41*, 339−344.
(23) Akutsu, K.; Obana, H.; Okihashi, M.; Kitagawa, M.; Nakazawa, H.; Matsuki, Y.; Makino, T.; Oda, H.; Hori, S. GC/MS analysis of polybrominated diphenyl ethers in fish collected from the Inland Sea of Seto, Japan. *Chemosphere* **2001**, *44*, 1325−1333.
(24) Christensen, J. H.; Glasius, M.; Pécseli, M.; Platz, J.; Pritzl, G. Polybrominated diphenyl ethers (PBDEs) in marine fish and blue mussels from southern Greenland. *Chemosphere* **2002**, *47*, 631−638.
(25) Ohta, S.; Ishikuza, D.; Nishimura, H.; Nakao, T.; Aozasa, A.; Shimidzu, Y.; Ochiai, F.; Kida, T.; Nishi, M.; Miyata, H. Comparison of polybrominated diphenyl ethers in fish, vegetables, and meats and levels in human milk of nursing women in Japan. *Chemosphere* **2002**, *46*, 689−696.
(26) Lind, Y.; Aune, M.; Atuma, S.; Becker, W.; Bjerselius, R.; Glynn, A.; Darnerud, P. O. Food intake of the brominated flame retardants PBDEs and HCBD in Sweden. *Organohalogen Compd.* **2002**, *58*, 181−184.
(27) Ryan, J. K.; Patry, B. Body burdens and food exposure in Canada for polybrominated diphenyl ethers (BDEs). *Organohalogen Compd.* **2001**, *51*, 226−229.
(28) Wijesekera, R.; Halliwell, C.; Hunter, S.; Harrad, S. A preliminary assessment of UK human exposure to polybrominated diphenyl ethers (PBDEs). *Organohalogen Compd.* **2002**, *55*, 239−242.

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