Estimation of the dietary PBDE intake: A market-basket study from Belgium

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Introduction

Polybrominated diphenyl ethers (PBDEs) are used as flame retardants to improve fire safety in both commercial and domestic applications (WHO, 1994). Because these chemicals are persistent and lipophilic, their environmental levels have been continuously increasing since their introduction (de Wit, 2002). The increasing presence of PBDEs in human tissues is of particular concern because of their toxicological potential (Darnerud, 2003). A major uptake route of PBDEs is through the food we consume daily. Due to their lipophilic nature, these man-made chemicals are primarily found in lipid-rich food of animal origin, such as meat, fish and dairy products. The importance of the different routes of human exposure to PBDEs (dietary and atmospheric) is not completely understood at the moment and opinions about the contribution of the dietary uptake vary among publications. In general, the dietary uptake is estimated between 73 (Wijesekera et al., 2002) and 93 % (Harrad et al., 2004). The remaining proportion of the uptake can be attributed to inhalation and ingestion of airborne particles. However, the contribution of inhalation to the total PBDE exposure is susceptible to great variance depending on several factors, such as life style (indoor – outdoor), age (toddler – adult), home environment and work place exposure (Jones-Otazo et al., 2005).

In the present study, a market basket containing various meat, fish and dairy food products was analysed for their PBDE content. Additionally, selected fast food items were investigated. Based on the PBDE levels measured, an average daily dietary intake estimate of these pollutants was calculated.

Materials and Methods

Sample collection. Sample selection was both based on food items that are likely to contain relatively high levels of the above-mentioned pollutants and on food consumption estimates of the Belgian population (KB 03 March 1992). Selected products were purchased in April 2005 at 2 international European large-chain supermarkets and 1 regional "biological/organic" supermarket. Not all products were available in all stores. Fish and meat products were also purchased at a local fish-store and a local butcher's. Most popular fast-food items were sampled from 3 different restaurants. The samples were homogenized immediately after collection and stored at –20°C until further treatment.

Analysis. The following PBDE congeners (IUPAC numbering) were targeted for analysis: 28, 47, 99, 100, 153, 154, 183, and 209. In addition, brominated biphenyl (BB) 153 was also looked for, using BB 155 as internal standard (IS). BDE 77 was used as IS for BDE 28, 47, 99, 100, 153, and 154, and BDE 128 was used as IS for BDE 183. ^{13}C-labelled BDE 209 was used as IS for BDE 209.

The method used for the analyses has previously been described (Voorspoels et al., 2003) and is briefly summarized below. The homogenized samples were chemically dried using anhydrous Na_2SO_4, transferred into an extraction thimble, spiked with IS and Soxhlet extracted with hexane/acetone mixture (3:1, *v/v*). An aliquot of the extract was used for gravimetrical lipid determination. Clean-up was conducted by column chromatography on silica impregnated with concentrated sulphuric acid (48 %, *w/w*). PBDEs were analysed by gas chromatography-electron capture negative ionisation mass spectrometry (GC/ECNI-MS) operated in selected ion monitoring (SIM) mode (Voorspoels et al., 2003). For analysis of BDE 209, a short 12 m × 0.18 mm × 0.1 μm AT-4 (Alltech) column was used. The other PBDEs were resolved on a 25 m 0.22 × 0.25 mm × μm HT-8 (SGE) column.

Quality Assurance and Quality Control. The method quality control (QC) was done by regular analysis of procedural blanks and blind duplicate samples (RSD < 5 %). Instrumental QC was done by regular injection of solvent blanks and standard solutions. Procedural blanks of both PBDEs and PCBs were consistent (RSD < 12 %) and therefore the mean procedural blank value was used for subtraction. After blank subtraction, the limit of quantification (LOQ) was set at 3 times the standard deviation of the blank, which ensures > 99 % certainty that the reported value is originating from the sample.

Results and discussion

PBDE levels. This study is the first to report the levels of PBDEs in Belgian foods in a market basket survey. We could not observe any differences in PBDE levels in the foods between the different supermarkets and vendors, therefore average levels were calculated using all analyses regardless of the origin. This was not unexpected since Belgium is a small country and foods are often not transported over large distances. We found a wide variation in sum of PBDE congeners concentrations across the food groups sampled.

Results are presented as the average sum of all measured PBDEs for each food item (wet weight normalised) in Table 1. Also the fat content of each food item is provided. Because the number of congeners with levels below the LOQ in certain foods was sometimes substantial, such as for meat products, estimations were necessary. Sum of PBDE levels are therefore classified in low, medium and upper bound, for which levels below LOQ for individual congeners were replaced by 0, ½ LOQ or the LOQ, respectively. All further discussion on levels is based upon the averaged medium bound sum of PBDE levels.

Table 1. Average sum of PBDE-levels (pg/g ww) and lipid percentage in the different foods.

	Food	Fat %	Lower bound	Medium bound	Upper bound
Fish and seafood	Cod	0.3	40	48	56
	Sardines	7.2	1	52	105
	Shrimp	1.3	51	61	70
	Mackerel	16	168	196	224
	Trout	3.1	269	273	278
	Smoked salmon	13	1017	1022	1027
	Fresh salmon	13	1570	1575	1580
Meat products	Beef steak	0.5	0	17	31
	Chicken breast	1.3	1	18	31
	Minced meat	18	3	54	106
	Pork sausage	24	32	75	119
	Hamburger	9.6	39	80	121
	Pork chop	2.8	62	91	120
	Salami	34	63	92	122
	Meatloaf	27	112	138	164
Dairy products	Cheese	34	18	118	217
	Eggs	10	35	102	170
	Butter	83	279	545	813
Fast food	Quick King Fish	18	0	52	105
	Mc Donalds Filet-O-Fish	11	5	55	107
	Quick Double Chicken	18	48	78	108
	Mc Donalds Mc Chicken	12	0	52	105
	Quick Gaint	18	76	104	133
	Mc Donalds Big Mac	12	160	160	160
	Pizza Hut Super Supreme	13	73	102	131

Lower Bound = N.D. (not detected) was replaced by zero; Medium bound = N.D. was replaced by 1/2 LOQ; Upper bound = N.D. was replaced by LOQ

The results of the present study indicate that fish contain the highest PBDE levels (average of 460 pg/g ww, 10 – 90 percentiles 50 – 1240 pg/g ww), followed by dairy products (average of 220 pg/g ww, 10 – 90 percentiles 100 – 380 pg/g ww), fast-food (average of 86 pg/g ww, 10 – 90 percentiles 55 – 130 pg/g ww), and meat products (average of 55 pg/g ww, 10 – 90 percentiles 15 – 82 pg/g ww). One fresh salmon filet had the highest total concentration of PBDEs (2364 pg/g ww) of all foods, whereas cod had the lowest levels in fish (29 pg/g ww) and levels in beef steak and chicken breast were the lowest of all foods analysed (< 20 pg/g ww). BB 153 could not be determined above the LOQ in any sample. This was expected since levels of this compound are known to be rather low in Belgian environmental samples in general (Voorspoels et al., 2003; Voorspoels et al., 2006).

*Dietary intake.*No average food intake studies have been completed to date in Belgium. The constitution of the market basket in the present study was therefore partly determined by inquiries in supermarkets and local stores, and partly by a publication providing a theoretical estimate of the average of daily food consumption in Belgium (KB 03 March

1992). The fast food intake estimate was based upon one restaurant visit each two weeks. Intake calculations did not include any fruit or vegetables, which (should) contribute to a great extent to the total daily diet. Levels in these food items however, are expected to be low, seeing the low fat content of these products and the lipophilicity of the pollutants under investigation. However, some studies reported the presence of PBDEs in fruits and vegetables, cereals and even spices and sweets (Ohta et al., 2002; Kiviranta et al., 2004; D'Sylva, 2006). Those studies indeed revealed a low PBDE content in those foods and thus a low contribution to the total dietary PBDE intake of around 20 % of the total.

For the intake calculations of the present study, the average theoretical daily consumption of each food category or individual food (g) was multiplied with the corresponding concentrations (pg/g ww), which resulted in a total daily intake (pg/day) (Table 2).

Table 2. Estimated dietary PBDE intake (ng) in Belgium.

		EDC (g)	Lower bound intake	Medium bound intake	Upper bound intake
Daily	Fish and seafood	30	13	14	14
	Meat products	150	5.8	11	15
	Cheese	30	0.6	3.6	6.5
	Eggs	30	1.1	3.1	5.1
	Butter	5	1.4	2.7	4.1
	Fast food	20	1.0	1.6	2.3
	Daily total		23	35	48
	Uptake 50%		12	18	24
	Uptake 90%		21	32	43
Yearly	**Yearly dietary uptake estimates**				
	Uptake 50%		4200	6500	8700
	Uptake 90%		7700	11600	15600

EDC = Estimated daily consumption (g) (KB 03.03.1992)

It is noteworthy that the data used in the estimation the dietary exposure to PBDEs will possibly overestimate the actual dietary exposure because most analyses were done on fresh unprocessed foods that were not boiled, cooked, baked or fried prior to analysis (except the smoked salmon and fast food samples). Cooking processes have been shown to lead to losses of PCBs and other organochlorines in trout, via the loss of fat (Zabik et al., 1996). Other processes that occur during food preparation can also reduce the pollutant load of foods, such as volatilisation and extraction into the cooking oil. Several studies reported reductions of PCBs and depending on the applied cooking process, these reductions can vary between 15 and 65 % (Zabik et al., 1992; Salama et al., 1998; Schecter et al., 1998; Wilson et al., 1998). No data is currently available on the loss of PBDEs following food preparation processes, but it can be assumed that, based on the similar physical properties as PCBs, similar losses will also occur following cooking. Additionally, the uptake of PBDEs in the GI tract is not well studied and possibly varies with the degree of bromination. Therefore, PBDE uptake estimates of 50% and 90% were assumed to calculate the effective biological available (absorbed) amount from the GI tract. A value of 90 % is consistent with both the assumption of Stapleton et al. (2005) in their calculation of potential doses to children as a result of ingesting PBDE-contaminated house dust in homes in Washington, D.C., and with the systemic absorption of BDE 47 in mice, which exceeded 80 % (Staskal et al., 2005). The relevance of these data to humans or other mammals is unknown at the present time. As a result, also a 50 % absorption scenario was selected.

Total average dietary PBDE intake in the present study was estimated between 23 and 48 ng/day. This value is in accordance with what was previously reported by others in other countries. Some publications are currently available on the issue of dietary PBDE intake and levels range from 24 to 91 ng/day (Darnerud et al., 2001; Ryan and Patry, 2001; Lind et al., 2002 Bocio et al., 2003; Harrad et al., 2004; Kiviranta et al., 2004). Although these intake estimations were based on diets from geographical distinct areas, such as Canada, Finland, Spain, Sweden and the UK, it is fair to say that the daily intake estimations of the present study are similar. Furthermore, food items that were included in the various studies were not always comparable, as some authors also included levels in cereals, fruits, vegetables, beverages and even spices and sweets. However, contributions of these food items were rather low, not affecting the total estimated intake to a too great extent.

Although fish is only a minor constituent of the Belgian diet, it can be considered a major contributor to the total daily PBDE-intake (around 40 %) due to the high PBDE levels in fish (Figure 1), while meat products account for around 30 % of the total dietary intake of PBDEs (Figure 1). Dairy products only contribute to a lesser extent. Also other studies have reported a similar uptake distribution. Sjödin et al. (2003) have found that around 50% of the total dietary PBDE uptake in Sweden originated from fish, while the rest was from meat and dairy products. The fish contribution is slightly lower in the present study, but this can be related to the dietary habit of Belgians, which tend to be more meat than fish consumers.

Based on the present results, it can be concluded that PBDEs are present in the Belgian diet and that their daily intake is situated in the same range as in other countries. However, to have a more complete picture of the human dietary PBDE exposure, more food items, such as vegetables, fruits, and cereals should be included. Furthermore, food should be treated in the same manner as it would be when consumed.

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