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RESEARCH ARTICLE

Potential risk assessment of polybrominated diphenyl ethers (PBDEs) by consuming animal-derived foods collected from interior areas of China

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Abstract Polybrominated diphenyl ethers (PBDEs) are one class of brominated flame retardants (BFRs). Although studies have reported the occurrence of PBDEs in freshwater fish species from several locations, to our best knowledge, there was no comprehensive data on PBDEs in foods of animal origin, such as pork, egg, and milk samples from interior areas of China, where pork and eggs are the major constituents of diet. The levels of PBDEs (BDE-28, 47, 99, 100, 153, 154, and 183) were determined in samples of animal-derived foods widely consumed by the population of Hubei in east-central China and the middle reaches of the Yangtze River. Two hundred six samples of animal-derived foods were randomly acquired in 17 sites of Hubei in 2010. The highest medium concentration of $\sum_7$PBDEs was found in chicken eggs (0.191 ng/g wet weight (ww)), followed by duck eggs (0.176 ng/g ww), pork (0.050 ng/g ww), carps (0.047 ng/g ww), and cow milk (0.013 ng/g ww). The estimated dietary intake of Σ_7PBDEs for a standard adult of 60 kg body weight based on medium and 95th percentile concentrations with consumption of animal-derived foods in Hubei province were 157.5 and 1960.3 pg/kg body weight/day, respectively. Chicken eggs (65.9 %) and pork (23.4 %) were the largest contributors to dietary intake of $\sum_7$PBDEs through animal-derived foods. The same PBDE sources were exposed in Hubei province via principal component analysis (PCA), and the particular congener profile in samples of animal-derived foods revealed the possible exposure history of octa-BDEs and penta-BDEs in the local region. The large margins of exposure (MOE) calculated following the European Food Safety Authority (EFSA) approach for three important congeners, BDE-47, BDE-99, and BDE-153, indicated that the estimated dietary exposures were unlikely to be a significant health concern to in Hubei.

Keywords Polybrominated diphenyl ethers · Animal-derived foods · Source · Risk assessment · Interior areas of China

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Introduction

Polybrominated diphenyl ethers (PBDEs) are one class of brominated flame retardants (BFRs). They are structurally akin to the PCBs and other polyhalogenated compounds, consisting of two halogenated aromatic rings. PBDEs are a group of persistent and bioaccumulative chemicals widely used in electronic appliances, plastics, textiles, polyurethane foam, and various other consumer products. Commercial production of PBDEs began in the 1970s (Eckley and Selin 2004), and reports of their presence in the environment began surfacing in the early 1980s (Staskal et al. 2008). There are three commercial PBDE products. Each of these products consists of a rather narrow range of congeners and is named after the dominating congeners in the bromination pattern as penta-, octa-, and decabromodiphenyl-PBDEs (Darnerud

2003; Szlinder-Richert et al. 2010). Generally, the penta-BDEs seem to cause adverse effects at the comparably lowest dose, whereas much higher doses were needed for effects of the deca-BDEs (Darnerud 2003). As additives, PBDEs are physically mixed rather than chemically bound into product applications. As a result, they can be released into the environment when the consumer products are used (Yu et al. 2011). PBDEs have been detected in abiotic and biotic matrices that include air and sediments and animal and human tissues (de Wit 2002; Kierkegaard et al. 2009; Law et al. 2006; Li et al. 2008; Schecter et al. 2008). PBDEs can be bioaccumulated and biomagnified in the aquatic and terrestrial food webs (Hu et al. 2010; Voorspoels et al. 2007; Zhang et al. 2010). At present, PBDEs seem to have developmental neurotoxicity and endocrine disruption in animal experiments (Costa and Giordano 2007; Darnerud 2008), and they have shown to reduce fertility in humans at levels found in households (Harley et al. 2010). Moreover, it has been demonstrated that some PBDEs, similar to dioxins, might act via Ah receptor-mediated pathways as either agonists or antagonists (Alaee and Wenning 2002; Szlinder-Richert et al. 2010). These findings have led the Stockholm convention to introduce nine new organic chemicals to the persistent organic pollutants (POPs) list which included tetra-, penta-, hexa-, and hepta-BDEs in May 2009 (UNEP/POPS/COP.4/17 2009). These findings have also led to bans in commercial use and production of penta- and octa-BDE mixtures in the European Union, Japan, and USA (Alaee et al. 2003).

In general, ingestion of contaminated food, especially of animal origin with high fat content, was the main dietary source of human exposure to persistent organic pollutants (POPs) (Djien Liem et al. 2000; Liu et al. 2011; Roosens et al. 2010). In animal origin foods, fish is considered a major exposure source for dietary POP exposure (Akutsu et al. 2008; Frederiksen et al. 2009; Voorspoels et al. 2007), even if only a small amount (about 10 %) of fish consumption in diet represents the main contamination route for humans (Voorspoels et al. 2007). Although studies have reported the occurrence of PBDEs in freshwater fish species from several locations (Gao et al. 2009; Meng et al. 2007; Xian et al. 2008), to our best knowledge, there was no comprehensive data on PBDEs in foods of animal origin, such as pork, egg, and milk samples from interior areas of China. The objective of this work was to determine the levels and profiles of PBDEs in pork, eggs, fish, and milk from Hubei province, an interior area of China. In Hubei, pork and eggs are the major constituents of diet, while marine fish were little consumed. The fact that marine fish products were the main contamination route for human dietary exposed to PBDEs was more or less well known (Chen et al. 2010; Darnerud et al. 2006; Gomara et al. 2006; Törnkvist et al. 2011). Then, the dietary intake of PBDEs for general population was calculated and the sources of PBDEs were also discussed.

Materials and methods

Food sampling and preparation

As shown in Fig. 1, a total of 206 samples were collected from local markets in 17 sites of Hubei province in 2010. Hubei covers an area of 185 900 km^2 with a population of 51 million. It locates in the heartland of China and the middle-lower reaches of the Yangtze River (Xie et al. 2009). The samples bought in markets included carps, pork, chicken eggs, duck eggs, and cow milk produced and permitted to sale in local areas (Table 1). Muscle from four carps or pigs of the same sampling site was taken randomly and combined into one composite sample for laboratory analysis, and the weight of every sample was about 300 g. Ten eggs from the same site were composed of one sample. Every sample of cow milk was 0.5 L. Since some areas produce little cow milk, there were only 13 samples collected from 8 cities. The edible filet tissues of carps, pork, eggs, and milk were fully homogenized and freeze-dried prior to analysis.

Materials and reagents

The standard solution containing congeners of BDE-28, 47, 99, 100, 153, 154, and 183 was 2.5 mg/L and purchased from AccuStandard (New Haven, CT, USA). BDE-77 solution was 1.0 mg/L and also purchased from AccuStandard. Silica gel 60 was purchased from ICN Biomedical GmbH (Oslo, Germany). Florisil was purchased from Merck (Darmstadt, Germany).

Chemical analysis

Analysis of PBDEs in carps, pork, eggs, and milk was performed following the procedure described by Liu et al. (Liu et al. 2008) with slight modification. The freeze-dried sample of 3–5 g was spiked with known amount of internal standards of BDE-77 (Eljarrat et al. 2008; Thomsen et al. 2002) and was extracted with 120 mL hexane/methylene chloride (5:1) by Soxhlet extraction for 20 h. Gravimetric lipid determination was performed after solvent evaporation. Then, bulk lipids were dissolved in 100 mL hexane and removed by stirring for 10 min with acid-modified silica gel (44 % H_2SO_4, *w*/*w*) in water bath at 50 °C. A glass column filled with anhydrous sodium sulfate, florisil, acidified silica, and silica gel was used for further cleanup. PBDEs were eluted with 150 mL hexane/methylene chloride (1:1). The eluate was evaporated in a rotavapor and further reduced to 20 μL under gentle nitrogen gas before GC–MS. Sample analysis was performed on an Agilent GC–MS 7890-5975C, operating in negative chemical ionization (NCI) mode. A column DB-5MS (15 m × 0.25 mm × 0.1 μm) was used for the separation of seven PBDE congeners. The column temperature was held at

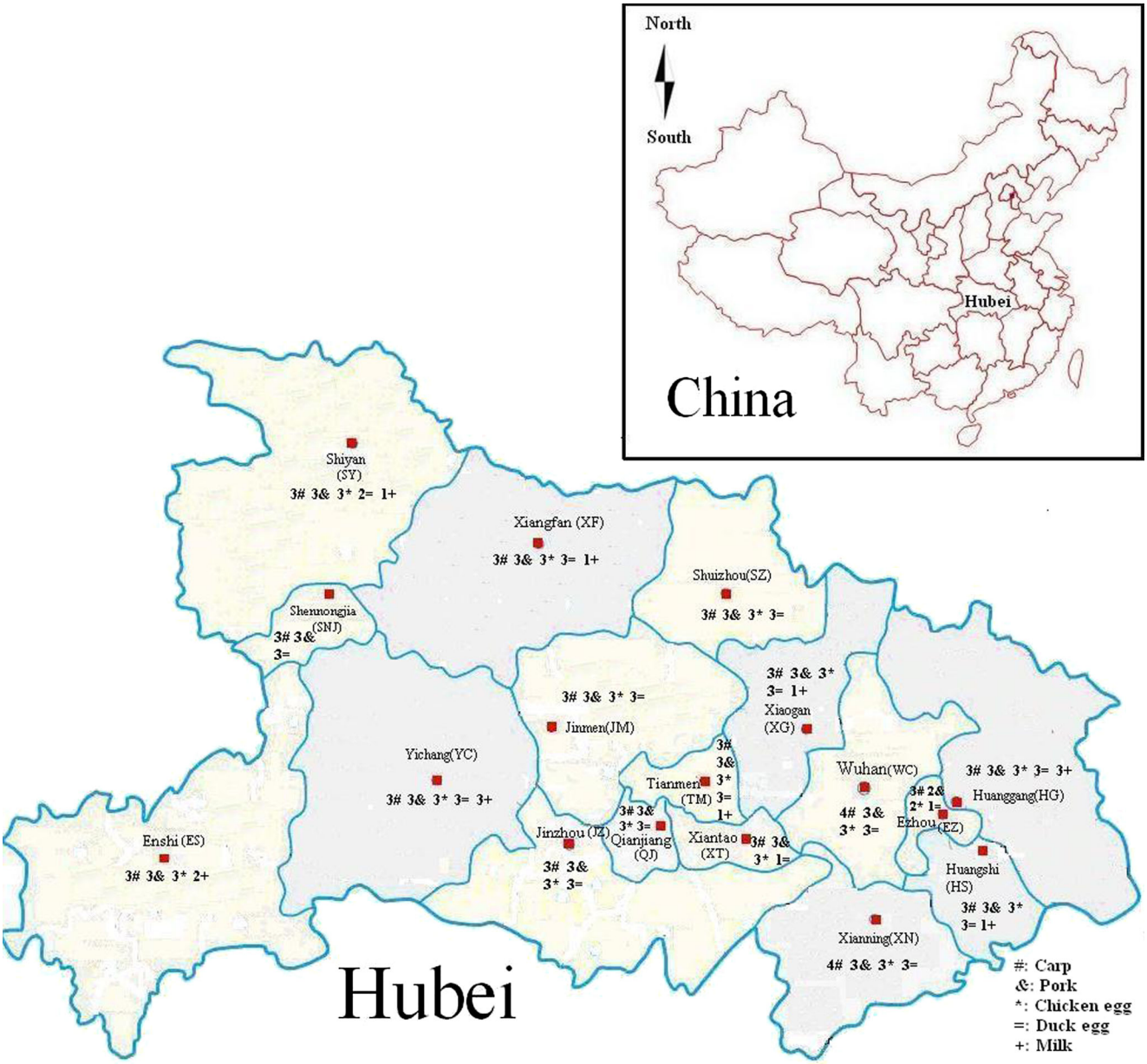

Fig. 1 Sample locations with number and symbols to indicate the amount and type of food samples in Hubei province

120 °C for 2 min then programmed at a rate of 26 °C min^{-1} to 310 °C, hold 3 min. Methane (CH_4) was used as the reagent gas. The ion source and transfer line temperature were 220 and 280 °C, respectively. Tri- to hepta-BDEs were detected using BDE-77 as internal standard, monitoring m/z 79 and 81.

Table 1 General information for samples

Sample	Sample size	Medium of water content (%)	Medium of lipid content (%)	Daily consumption (g)
Carp	53	81.1	2.1	17.7
Pork	50	43.3	41.3	44.2
Chicken egg	47	74.8	9.1	32.6
Duck egg	43	75.6	9.8	0.8
Milk	13	88.1	3.8	3.9

Quality assurance and quality control

In this study, method blank samples were performed every 11 samples. Spiking experiments of all PBDE standards (n=3) were done in solvent and five species of animal-derived foods, both at 1 ng/g ww (wet weight). The results showed that the recoveries were 74.4~105.2 %, and the relative standard deviation (RSD) was 5.4~13.0 % for PBDE standards in solvent. The recoveries were 71.0~93.3 %, and the RSD was 5.3~14.0 % in five species of animal-derived foods. The limit of

detection (LOD) was calculated as three times the procedural blank. LODs ranged 0.003 to 0.1 pg/g ww for seven PBDE congeners and internal standards of BDE-77. The laboratory was validated by successfully participating in the inter-laboratory comparison studies of PBDEs in pork organized by the Norwegian Institute of Public Health in 2010. The Z score value of sum PBDEs without BDE 209 in pork was 0.28.

Data analysis

The Statistical Package for Social Sciences (IBM SPSS Statistics for Windows, Version 19.0; SPSS Inc., Chicago, IL) was used for the quantitative data analysis. The differences among groups were assessed by the *t* test. The significant level was $P<0.05$ and two-tailed. Data were presented as medium and/or the range. Principal component analysis (PCA) was performed to find out the differences and similarities in PBDE profiles among samples.

Dietary survey

The average consumption of carps, pork, chicken eggs, duck eggs, and cow milk derived from the total diet study (TDS) of Hubei in 2010 was applied to reflect general dietary habits of local residents. Detail TDS of Hubei was performed as Chinese TDS (Li et al. 2007). TDS of Hubei in 2010 was reviewed and approved by Hubei Provincial Centre for Disease Control and Prevention (HB CDC) before the study began. The relative proportions of food items in the diets of adults from the general population of Hubei were determined by use of a questionnaire given to 1090 randomly selected individuals from all the 17 sampling sites. The participants provided their written informed consent to participate in this study. Dietary data in the questionnaire were collected through detailed in-person interviews and it was in written form. All participants were local residents who were interviewed to determine the amounts of 13 food species including animal-derived foods, vegetables, fruits, cereal products, and so on. Data collected for each food item included frequency of consumption and the quantity consumed on each occasion. Dietary intake was expressed as grams per day (g/d) for each person. Dietary exposure, expressed as pictograms per kilogram (basis weight) per day (pg/kg bw/day), was determined for each person based on a hypothesis that the weight of adult is 60 kg.

Results and discussion

Residue levels of PBDEs

A large variability in $\sum_7$PBDE concentrations was found between individual animal-derived foods of Hubei. The greatest concentration of $\sum_7$PBDEs (4.830 ng/g ww) was observed in one sample of pork, while PBDEs was not detected in a sample of milk. Medium concentrations of $\sum_7$PBD ranged from 0.013 ng/g ww in milk to 0.191 ng/g ww in chicken eggs (Table 2). Figure 2 provided an overview of the levels $\sum_7$PBDEs in the five species of animal-derived foods. And the ranking of medium concentrations of $\sum_7$PBDEs was following the descending order of chicken egg > duck egg > pork > carp > milk.

Similar to the study in Nanjing, a city in the Yangtze River Delta, China (Su et al. 2012), levels of ∑PBDEs in foods of Hubei were less in fish, but higher in meat than that in the USA (Schecter et al. 2004; Schecter et al. 2006b), Spain (Bocio et al. 2003), Sweden (Darnerud et al. 2006), Finland (Kiviranta et al. 2004), and Japan (Ohta et al. 2002) when compared to other market basket surveys. Compared with similar studies conducted in China, the levels of ∑PBDEs in fish from Hubei (medium of 0.047 ng ΣPBDE/g ww, 0.002–0.753 ng ΣPBDE/g ww) was lower than that of Nanjing (0.180 ng ΣBPDE/g ww, $P=0.002$) and Guangdong (0.231 ng ΣPBDE/g ww, $P=0.001$) (Meng et al. 2007) and notably lower than that in freshwater fishes from the lower reach of the Yangtze River (18 to 1100 ng/g lipid weight) (Xian et al. 2008), Hong Kong (13.2 ng/g ww) (Cheung et al. 2008), and the marine fish (91 ng/g ww) reported from four sea areas of China (Liu et al. 2011). The concentrations of ∑PBDEs in pork of Hubei (0.003–4.830 ng ΣPBDE/g ww) was comparable to meat of Nanjing (0.015–0.950 ng/g ww, $P>0.05$) (Su et al. 2012), Hong Kong (0.023–3.500 ng/g ww, $P>0.05$) (Chen et al. 2010), and Taiwan (mean±standard deviation 0.545±0.181 ng/g ww, $P>0.05$) (Chen et al. 2012). The main reason for this observation could be that carps had relatively low lipid content (2.1 %), and pork had great lipid content (41.3 %). High PBDE concentrations in diet have been found in food items with high lipid content like meat fat (Frederiksen et al. 2009). However, other factors like accumulation with time and seasonal variations were likely to contribute to the PBDE level in meat and fish (Darnerud et al. 2001).

The mean and range of ∑PBDE levels in chicken eggs across all 17 sites were not different significantly as those in duck eggs ($P>0.05$). The ∑PBDE levels in eggs in this study (mean of 0.417 ng/g ww, median of 0.189 ng/g ww, 0.002–4.674 ng/g ww) were closed to chicken eggs in Hong Kong (mean of 0.49 ng/g ww, 0.28–0.8 ng/g ww, $P>0.05$) (Chen et al. 2010), but higher than those reported median concentrations of ∑PBDE in Europe (0.03–0.162 ng/g ww) (Bocio et al. 2003; Covaci et al. 2009; Darnerud et al. 2006; Dirtu and Covaci 2010; Gomara et al. 2006; Törnkvist et al. 2011; Voorspoels et al. 2007) and North America (0.085 ng/g ww) (Schecter et al. 2006a).

∑PBDE levels in milk (medium of 0.013 ng/g ww, ND-0.590 ng/g ww) were higher than milk in Finland, Spain, and

Table 2 Summary of results on the PBDE levels in samples from Hubei province

	Carp			Pork			Chicken egg			Duck egg			Milk		
	Range	Medium	Mean±SD	Range	Medium	Mean±SD	Range	Medium	Mean±SD	Range	Medium	Mean±SD	Range	Medium	Mean±SD
BDE-28[a]	ND-0.665	0.00036	0.02109±0.09462	ND-0.121	0.00005	0.01241±0.02991	ND-0.390	0.00011	0.03447±0.07654	ND-0.395	0.00004	0.02223±0.07209	ND-0.227	0.00029	0.01912±0.06256
BDE-47[a]	ND-0.214	0.00109	0.01158±0.03068	ND-0.405	0.00135	0.02144±0.06277	ND-0.471	0.00314	0.04400±0.10015	ND-0.357	0.01140	0.04716±0.07476	ND-0.062	0.00099	0.01043±0.01888
BDE-99[a]	ND-0.124	0.00020	0.00684±0.01953	ND-0.035	0.00014	0.00346±0.00745	ND-0.156	0.00012	0.00839±0.02621	ND-0.148	0.00012	0.00855±0.02509	ND-0.253	0.00282	0.03149±0.06991
BDE-100[a]	ND-0.030	0.00005	0.00334±0.00669	ND-0.231	0.00009	0.00647±0.03381	ND-0.671	0.00009	0.01556±0.09778	ND-0.097	0.00009	0.00331±0.01495	ND	0.00003	0.00004±0.00002
BDE-153[a]	ND-0.291	0.00136	0.01783±0.04711	ND-2.189	0.00800	0.11054±0.33365	ND-2.339	0.00034	0.09647±0.35520	ND-2.044	0.00637	0.09977±0.31897	ND-0.035	0.00021	0.00612±0.01270
BDE-154[a]	ND-0.205	0.00693	0.01857±0.03249	ND-0.203	0.00280	0.01973±0.04194	ND-0.161	0.00019	0.01496±0.03234	ND-0.362	0.00020	0.02558±0.06435	ND-0.031	0.00016	0.00490±0.00908
BDE-183[a]	ND-0.322	0.00650	0.02806±0.05918	ND-2.236	0.01155	0.14767±0.37378	ND-2.306	0.03289	0.21310±0.44157	ND-5.800	0.10945	0.37375±0.97149	ND-0.075	0.00072	0.01005±0.02044
Σ_7PBDEs[a]	0.002–0.753	0.04700	0.10731±0.16371	0.003–4.830	0.05000	0.32173±0.75326	0.002–4.674	0.19100	0.42694±0.78583	0.002–3.411	0.17600	0.40645±0.61195	ND-0.590	0.01300	0.08216±0.15965
Σ_7PBDEs[b]	0.070–77.795	3.13800	7.36107±13.19378	0.004–8.074	0.12400	0.84708±1.64726	0.014–55.660	1.68700	6.33046±12.09620	0.028–50.491	1.80700	5.03756±9.07546	0.064–8.498	0.54200	2.05012±2.81046

Values below the detection limit were considered none detected (ND) and set to zero when means were calculated

[a] Levels in samples are expressed in ng/g wet weight

[b] Levels in samples are expressed in ng/g lipid weight

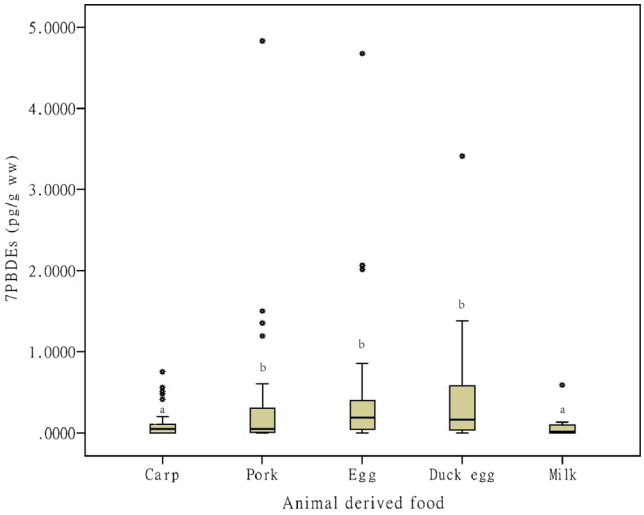

Fig. 2 Levels of Σ_7PBDEs in the five species of animal-derived foods

America (0.00082–0.032 ng/g ww, P=0.041) (Kiviranta et al., 2004; Bocio et al., 2003; Schecter et al. 2006a) and even higher than solid milk products like cheese (0.018–0.034 ng/g ww, P=0.039) in Europe (Darnerud et al. 2006; Kiviranta et al. 2004; Voorspoels et al. 2007). Commonly, ∑PBDE levels in solid milk products contain relatively higher amounts of PBDEs than liquid milk products (Frederiksen et al. 2009).

This difference of PBDE content in the five species of animal-derived foods with other studies may be partially due to variations in time of sampling and samples such as species, source, and size. A comprehensive review of recent literature suggested that factors like different sampling times, species, sampling sites, or tissues chosen for analyses can severely influence the PBDE content found in fish (Frederiksen et al. 2009).

Congener profiles of PBDEs

The congener compositions of animal-derived food samples are given in Fig. 3. Many studies indicated that BDE-47 was the most predominant congener in aquatic biota, eggs, fish, shellfish, meat, and milk production (Gao et al. 2009; Gomara et al. 2006; Labandeira et al. 2007; Schecter et al. 2006a; Su et al. 2012). Interestingly, this study found that the most abundant congener was BDE-183, accounted for about 40 % in carps, pork, and eggs averagely. The observed PBDE profiles were highly variable, most probably because of various contamination/exposure sources and because PBDE may transform very easily in the environment (Roszko et al. 2013). The finding was similar to the study of fish from South China Sea, in which BDE-183 was the most predominate, with relative proportions 33 % of the overall PBDEs (Liu et al. 2011). Gomara et al. (2006) also showed that non-fish samples contained relatively larger amounts of the higher brominated congeners (BDE-183, 196, 197, and 209). However, there

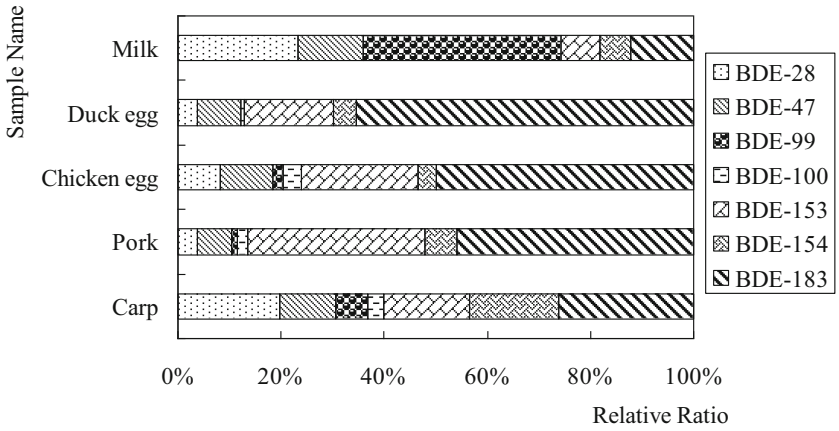

Fig. 3 Relative abundances of individual PBDE congeners in animal-derived foods of Hubei province

were different congener profiles in milk, and BDE-99 was the most predominant congener (Fig. 3). Similar observation was in the congener profiles of PBDEs in cow milk of USA (Schecter et al. 2006a).

In order to find out the potential differences and similarities among PBDE profile in samples, principal component analysis (PCA) was performed. It was found that the values of three factor variables were added up to nearly 91 %. Samples can be classified into two groups when considering PBDEs in animal-derived food samples of five species (Fig. 4). Overwhelming majority of samples was located in the big group, which can be explained that there were the same PBDE sources in Hubei. The congener profiles of PBDEs in the samples of the big group were BDE-183 (47.6 %), followed by BDE-153 (20.2 %), BDE-47 (11.7 %), −28, −154, −99, and −100. Meanwhile, congener profiles of PBDEs in the samples of the small group were BDE-183 (67.7 %), BDE-153 (31.1 %), BDE-47 (0.6 %), −154, −100, −99, and −28. Then, it was indicated that BDE-183, −153, and −47 were the most predominant congeners in animal-derived foods of Hubei. Considering BDE-183 is the major component (around 25 %) in the octa-BDE commercial mixtures, and BDE-153 and −47 are the major components in the penta-BDEs; this finding may suggest consumption history of octa-BDE and penta-BDE products in Hubei. Well, PBDE are in fact pretty unstable in the environment until incorporated in any kind of a fatty matrix. Observed PBDE levels/profiles (e.g., BDE-183) may be affected by (bio-)transformations of

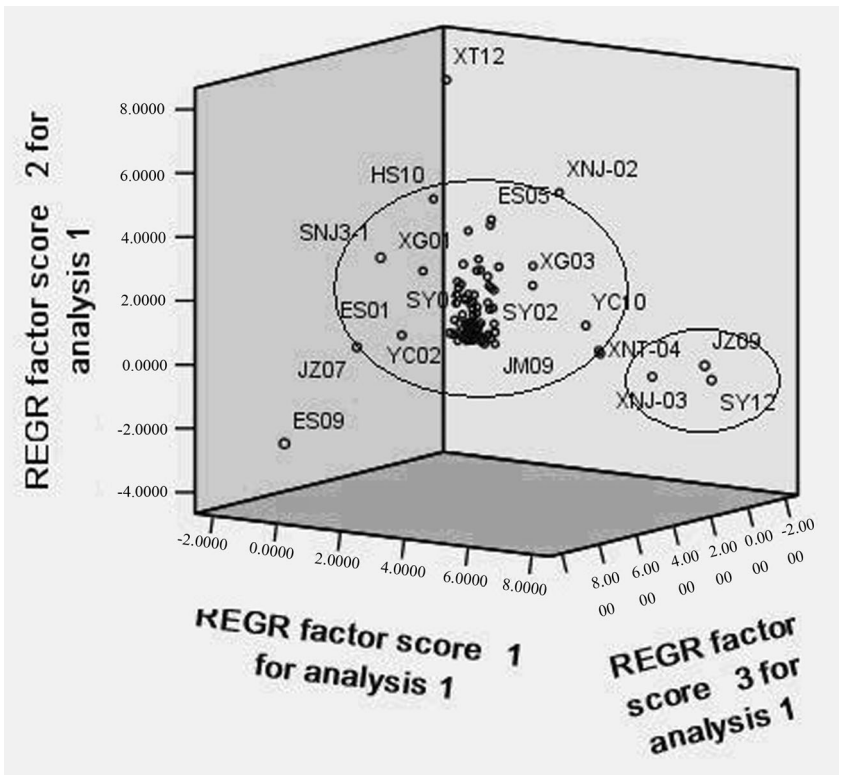

Fig. 4 Score plots of the principal component analysis (PCA) of Σ_7PBDE results in animal-derived food samples from Hubei province

PBDE in the environment including debromination (Roszko et al. 2014). The results of several studies indicate that BDE 209 degrade relatively easily, forming a wide spectrum of lower-brominated analogs (tri- to octa-substituted) (Shih and Wang 2009).

Dietary exposure to PBDEs

The basis for the animal-derived food intake estimation is the occurrence data of PBDEs as well as the dietary survey of animal-derived foods on local residents of Hubei province. In the present study, the consumption data of the five species of animal-derived food were based on TDS of Hubei in 2010 (Table 1), and the PBDE levels were shown in Table 2 and Fig. 2. Then, the medium and 95th percentile of dietary PBDE exposures of the Hubei adults were 157.5 and 1960.3 pg/kg bw/day, respectively. However, it should be emphasized that although it is well known that BDE-209 is a great contributor to the dietary intake and BDE-209 dietary exposure in the EU does not raise a health concern (EFSA 2011), it has not been analyzed in this work and therefore it has not been taken into account in the estimated daily intake of the sum of PBDEs.

Estimated dietary exposures to PBDEs expressed as MOE

The Joint FAO/WHO Expert Committee on Food Additives (JECFA) stated that the available toxicity data are inadequate to derive a TDI for PBDEs (JECFA 2006). Therefore, a MOE approach was used to determine the health risk of dietary PBDE exposure of the population by the European Food Safety Authority (EFSA) (EFSA 2011). The panel on contaminants in the food chain (CONTAM) set a $BMDL_{10}$ (lower 95 % confidence limit for a benchmark response of 10 %) for BDE-47, BDE-99, and BDE-153 (EFSA 2011). EFSA calculated the values called "chronic human dietary intakes" for BDE-47, BDE-99, and BDE-153, which were derived by taking into account the body burden in the experimental animals at $BMDL_{10}$, The chronic human dietary intake are 172, 4.2, and 9.6 ng/kg bw/day for BDE-47, −99, and −153, respectively. The MOE can be calculated by comparing the chronic human dietary intake/$BMDL_{10}$ with the estimated dietary exposure of the population. EFSA considered that an MOE larger than 2.5 might indicate that there is no health concern (EFSA 2011). Estimated MOE for average and high consumers for adults were calculated by comparing the chronic human dietary intake with the estimated medium and 95th percentile dietary exposures of the Hubei's adults in Table 3. The results show that all the MOEs for the three congeners were greater than 2.5. In conclusion, the MOEs did not indicate a health concern with respect to current dietary exposure in the Hubei province.

Table 3 Margins of exposure (MOEs) for BDE-47, −99, and −153 of the medium and 95th percentile of exposures of dietary PBDEs for the Hubei people

PBDE congener	Chronic human dietary intake (pg/kg bw/day)	Medium		95th percentile	
		Dietary exposure (pg/kg bw/day)	MOE	Dietary exposure (pg/kg bw/day)	MOE
BDE-47	172000	3.24	53086	199.21	863
BDE-99	4200	0.41	10244	61.19	69
BDE-153	9600	6.58	1459	641.85	15

Comparison of PBDE dietary intake

Compared with the results of recent studies, the medium value of dietary intake of ∑PBDE in Hubei (9.4 ng/d) was in line with those from seafood in South China (1.7–18.5 ng/d) (Guo et al. 2010; Meng et al. 2007) and those from animal-based foods in Nanjing City (9.9 ng/d) (Su et al. 2012) and Shanghai City (13.67 ng/d) (Yu et al. 2012). The dietary intake in Hubei was lower than those reported from basket surveys of European food (23–55.3 ng/d, P=0.022) (Bakker et al. 2008; Dirtu and Covaci 2010; Gomara et al. 2006; Kiviranta et al. 2004; Törnkvist et al. 2011) and much lower than those reported from foods of animal origin in Hong Kong (222–2170 ng/d) (Chen et al. 2010; Cheung et al. 2008), foodstuffs in the USA (72.2 ng/d) (Johnson-Restrepo and Kannan 2009), and marine foodstuffs in Australia (63.0–89.6 ng/d) (Shanmuganathan et al. 2011). The notable differences of dietary intake values observed from other countries and regions could be expected the different food items and the number of individual PBDE congeners (Domingo 2012).

Major food contributors

It was found that fish was not the largest contributor to the dietary intake of PBDEs through animal-derived foods in Hubei province, which was not consistent with the results of fish products as the main contamination route for humans (Chen et al. 2010; Darnerud et al. 2006; Gomara et al. 2006; Törnkvist et al. 2011). The medium value of $\sum_7$PBDEs in chicken eggs was the highest and much higher than that in pork (Fig. 2). Considering the similar daily consumption of chicken eggs and pork (Table 1), the dietary exposure of $\sum_7$PBDEs through chicken eggs was 103.7 pg/kg bw/day and nearly three times higher than that through pork. In a word, chicken eggs (65.9 %) and pork (23.4 %) were the largest contributors to $\sum_7$PBDEs among the different animal-derived food types (Fig. 5). The contamination of eggs with PBDEs appears to be of low concern for public health and the contribution of eggs to the total dietary intake of

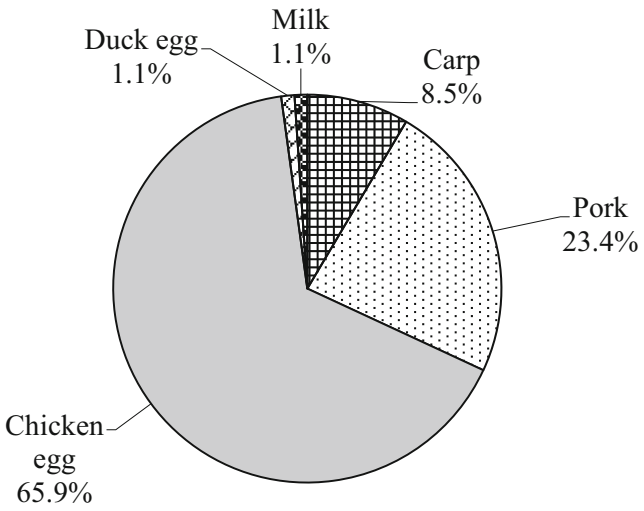

Fig. 5 Relative abundances of Σ_7PBDE daily intake through animal-derived foods of Hubei province

PBDEs appears to be limited (Covaci et al. 2009). For pork, a similar observation by the study in Nanjing was that meat products were a more important source of PBDEs than fish (Su et al. 2012). In the USA, meat and meat products tended to account for most of the exposure to PBDEs (Schecter et al. 2006a; Schecter et al. 2010). The reason could be that the different dietary habits between regions and countries, and the people in Hubei consume animal-derived foods based mainly on pork and chicken eggs.

It is also important to note that we analyzed fresh foods, whereas preparation and different cooking methods can influence the levels of contaminants and thereby also consumers' exposure (Törnkvist et al. 2011). Cooking processes have resulted in the loss of PBDEs via the loss of fat (Schecter et al. 2006b), and PBDE losses were even relatively greater than other POPs (Perelló et al. 2009).

Conclusion

To our best knowledge, this is the first study to report concentrations of PBDEs in foods of animal origin, such as pork, egg, and cow milk samples from interior areas of China. It was presumed that there were the same PBDE sources in Hubei province through principal component analysis (PCA). The particular congener profile in samples of animal-derived foods revealed that the commercial PBDE formulation of octa-BDEs and penta-BDEs might have exposure history in Hubei province. Chicken eggs and pork accounting for 90 % were predominant contributors to the dietary intake of $\sum_7$PBDEs through animal-derived foods in Hubei. In relation to risk analysis, margins of exposure (MOEs) for the BDE-47, −99, and −153 congeners for adults were calculated, and it did not indicate a health concern with respect to current dietary exposure in Hubei. Well, it should draw attention that for the total PBDEs, daily intake must be taken into account other external exposure routes as dust, air, and other exposures via diet (dairy products and oils, vegetables, etc.). On the other hand, the analysis of BDE-209 should be considered in future works to avoid the underestimation of the PBDEs intake, since it was has been described by EFSA as a great contributor to the dietary intake.

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