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ARTICLE *in* JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY · JUNE 2013

Impact Factor: 2.91 · DOI: 10.1021/jf4006565 · Source: PubMed

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JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

Article

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Polybrominated Diphenyl Ethers (PBDEs) and Indicator Polychlorinated Biphenyls (PCBs) in Foods from China: Levels, Dietary Intake, and Risk Assessment

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Ⓢ *Supporting Information*

ABSTRACT: A national survey of polybrominated diphenyl ethers (PBDEs) and indicator polychlorinated biphenyls (PCBs) congeners in various foodstuffs from the Chinese total diet study (TDS) performed in 2007 was conducted for the first time. Meats and aquatic foods had the highest average sum PBDEs (192.5 and 190.6 pg g^{-1} fresh weight, respectively). For indicator PCBs, the highest average concentration was found in aquatic foods (628.7 pg g^{-1} fresh weight). On the basis of measured PBDE and indicator PCB levels, the dietary intake estimate was subsequently calculated for the nonoccupationally exposed population in China. For adults, average estimated dietary intakes of PBDEs and indicator PCBs were 0.76 and 2.34 ng kg^{-1} bw day^{-1}, respectively. Health risk assessment of PBDEs using a MOE approach recommended by EFSA suggested unlikely health concern with respect to current dietary intake of PBDEs in China.

KEYWORDS: *PBDEs, indicator PCBs, dietary intake, risk assessment*

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are one class of brominated flame retardants (BFRs) used for several decades to protect people from fires, which have been widely used as nonreactive additives in textiles, polyurethane foams, thermoplastics, and electronic appliances, originating from three commercial PBDE mixtures: penta-, octa-, and deca-BDEs.[1,2] Due to their persistent characteristics and toxicological effects, there have been strict bans on the use of penta- and octa-BDEs for all products in the European Union, Japan, and United States.[2,3] Components of the penta- and octa-BDE commercial mixtures have been recently added to the list of the Stockholm Convention of the Persistent Organic Pollutants (POPs) to be eliminated from production and use.[4] Unlike PBDEs, polychlorinated biphenyls (PCBs) are the classic POPs, and the production and use of these pollutants have been banned all over the world since the 1980s.[5] These two classes of POPs are ubiquitous in environmental and biological samples worldwide because of persistence and long-range transport.[6] Therefore, the contamination status of these POPs and their potential health risk have been matters of great concern.

It is well-known that the exposure of nonoccupationally exposed population to polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and PCBs mainly originates from food, especially foods of animal origin. However, there are complicated pathways of human exposure to PBDEs, which occurs mainly via dietary intake, ingestion of indoor dust, inhalation of indoor air, and dermal absorption.[2,7−11] The relative contribution of these three pathways considerably varied among different populations. With regard to human exposure through food consumption, data on the concentration of PBDEs and indicator PCBs were relatively abundant in fish and meat, but much less information has been reported in other major food groups for which the contribution to total dietary exposure should be taken into account.[2,3,5]

Over the past 30 years, China has been rapidly industrializing, which has resulted in great release of some organic compounds into the environment.[12,13] There has been an increasing interest in dietary intake of PBDEs and PCBs of residents in China, but only a few regional studies have been conducted.[12,14] In this study, a nation-wide survey was conducted to investigate levels of PBDEs and indicator PCBs in various foodstuffs and evaluate the dietary exposure of the nonoccupational population in China. Seven so-called "indicator PCBs" were selected as suitable representatives for all PCBs.[15] At the end, risk assessments were performed to determine the potential health risk.

Received: February 28, 2013
Revised: May 30, 2013
Accepted: June 6, 2013
Published: June 6, 2013

Table 1. Levels of PBDEs and PCBs in Selected Food Groups in China (Picograms per Gram Fresh Weight, n^a = 12)

	aquatic foods	meats	eggs	milk	cereals	legumes and nuts	potatoes	vegetables
BDE-28	19.6 ± 12.6[b]	3.2 ± 2.1	1.7 ± 1.3	0.7 ± 0.5	1.2 ± 1.4	2.0 ± 1.8	0.6 ± 0.6.	1.2 ± 0.4
BDE-47	95.0 ± 48.8	33.3 ± 39.2	11.5 ± 7.1	3.6 ± 2.3	3.0 ± 2.8	2.9 ± 3.1	2.4 ± 2.5	4.8 ± 4.6
BDE-99	11.9 ± 9.7	21.3 ± 25.7	7.5 ± 5.8	1.5 ± 1.1	3.2 ± 2.9	1.8 ± 1.3	2.2 ± 3.7	4.3 ± 5.8
BDE-100	16.4 ± 10.7	7.2 ± 11.0	2.5 ± 1.5	0.4 ± 0.5	0.6 ± 0.5	0.3 ± 0.3	0.6 ± 0.7	1.0 ± 1.0
BDE-153	10.6 ± 9.3	54.4 ± 89.8	11.5 ± 7.2	2.5 ± 2.6	1.3 ± 2.1	1.4 ± 1.7	2.4 ± 3.2	3.1 ± 2.8
BDE-154	29.0 ± 17.3	23.5 ± 41.9	3.9 ± 2.8	0.8 ± 0.8	0.4 ± 0.4	0.5 ± 0.5	1.0 ± 1.3	1.4 ± 1.5
BDE-183	8.0 ± 10.9	49.7 ± 90.5	17.0 ± 15.5	0.7 ± 1.0	1.6 ± 3.2	3.0 ± 2.8	3.8 ± 6.8	3.6 ± 5.0
$\sum_7$PBDEs	190.6 ± 102.7	192.5 ± 237.5	55.6 ± 27.1	10.2 ± 7.6	11.2 ± 10.6	11.9 ± 6.8	13.0 ± 12.3	19.5 ± 15.3
	(46.8−418.9)[c]	(35.2−763.3)	(12.4−104.1)	(3.2−27.2)	(0.2−37.1)	(1.3−21.4)	(0.7−33.5)	(3.0−47.0)
PCB-28	180.3 ± 109.5	96.5 ± 48.6	99.1 ± 66.7	17.6 ± 19.9	5.2 ± 4.8	21.1 ± 14.7	15.1 ± 17.0	25.7 ± 23.8
PCB-52	53.0 ± 55.0	13.8 ± 8.1	6.6 ± 3.4	3.8 ± 2.7	2.4 ± 2.2	5.9 ± 4.0	6.0 ± 7.1	5.9 ± 4.1
PCB-10	78.6 ± 45.2	20.8 ± 8.9	16.0 ± 14.0	4.7 ± 3.4	11.4 ± 12.6	10.2 ± 10.1	8.6 ± 9.2	5.5 ± 3.3
PCB-118	62.2 ± 37.1	23.9 ± 22.8	24.8 ± 15.4	11.1 ± 8.4	3.3 ± 4.1	2.6 ± 2.2	3.1 ± 2.6	2.8 ± 2.7
PCB-138	84.2 ± 54.0	36.6 ± 42.1	24.0 ± 14.3	11.5 ± 9.3	5.3 ± 6.5	5.6 ± 8.2	5.0 ± 3.6	2.5 ± 2.9
PCB-153	126.1 ± 75.3	52.0 ± 48.5	34.3 ± 17.4	14.8 ± 10.1	15.0 ± 20.9	12.3 ± 17.5	5.6 ± 4.2	3.4 ± 3.3
PCB-180	44.3 ± 33.5	18.1 ± 12.1	10.6 ± 7.9	5.2 ± 3.9	4.4 ± 5.8	3.1 ± 5.3	2.3 ± 1.9	1.0 ± 1.2
$\sum_7$PCBs	628.7 ± 337.1	261.6 ± 153.4	215.2 ± 93.9	68.7 ± 35.0	46.9 ± 49.6	60.9 ± 48.2	45.7 ± 39.0	46.7 ± 33.7
	(192.7−1215.4)	(133.1−693.8)	(92.6−346.9)	(20.6−134.6)	(2.4−174.8)	(23.4−159.6)	(18.2−165.0)	(7.2−114.6)

[a]Number of food composite samples of every food group. [b]Mean ± standard deviation. [c]Range of contaminants given in parentheses.

MATERIALS AND METHODS

Food Consumption Survey and Sampling. The Chinese Total Diet Study (TDS) is a continual national study monitoring the levels of various chemical pollutants in foods and estimating the dietary exposure to these chemicals of the general, nonoccupationally exposed, population in China, which has been detailed elsewhere.[16−18] The food consumption survey was organized by the Chinese Center for Disease Control and Prevention in 2000. A multistage random cluster sampling method was used in this study. First, all provinces were classified into northern area and southern area according to geographical attribute and resident dietary pattern. Then, six provinces were randomly sampled from each area. The 12 provinces were Heilongjiang, Liaoning, Hebei, Shanxi, Ningxia, Henan, Shanghai, Fujian, Jiangxi, Guangxi, Hubei, and Sichuan, covering about 50% of the total Chinese population. In each province, three sampling sites including one urban site and two rural sites were randomly selected, and then 30 households were randomly sampled from each site to conduct the food consumption survey by a 3-day household dietary survey that documented all of the food consumption by a weighing and recording method. All food items were aggregated into 12 groups, and then the average food consumption was calculated to present the pattern of food consumption. Of these 12 groups, 8 groups were selected for the determination of PBDEs and indicator PCBs, including aquatic foods, meats, eggs, milk, cereals, legumes and nuts, tubers, and vegetables. The average value of daily consumption of selected food groups for adults from various regions in China is shown in Supplementary Table 1 of the Supporting Information, and the food items comprising each food group are shown in Supplementary Table 2 of the Supporting Information.

According to the pattern of food consumption, food samples were collected from local food markets, grocery stores, or rural households in each sampling site and then prepared using local practice and recipes. The composite of each food group was made by blending the prepared foods with weights proportional to the average daily consumption in each province. These provincial composites were shipped to the China National Center for Food Safety Risk Assessment and frozen at −20 °C until analysis.

Analytical Methods. Analysis of PBDEs and indicator PCBs was as described elsewhere,[19] with slight modification. Briefly, approximately 50 g of food sample was freeze-dried and spiked with ^{13}C-labeled internal standards of PBDEs (^{13}C-BDE-28, -47, -99, -100, -153, -154, and -183) and ^{13}C-labeled internal standards of indicator PCBs (^{13}C-PCB-28, -52, -101, -138, -153, and -180). The samples were extracted with a mixture of 50% hexane/dichloromethane (1:1) by using an accelerated solvent extractor (ASE300, Dionex, Sunnyvale, CA, USA) at 120 °C and 1500 psi. The bulk lipid was removed by shaking with acid-modified silica gel after solvent evaporation, and further cleanup was achieved using a Power Prep instrument (Fluid Management Systems, Waltham, MA, USA). The fraction containing the PBDEs and indicator PCBs was collected. After this fraction had been concentrated to approximately 50 μL, the ^{13}C-labeled injection standards (^{13}C-PCB-70, -111, and -170 and ^{13}C-BDE-77 and -138) were added prior to instrument analysis.

The identification and quantification were performed by high-resolution gas chromatography−high-resolution mass spectrometry (HRGC-HRMS, MAT95XP, ThermoFinnigan, Germany). DB-5MS capillary columns, 15 m × 0.25 mm i.d. × 0.1 μm, and 60 m × 0.25 mm i.d. × 0.25 μm, were applied for the analysis of PBDEs and indicator PCBs, respectively. The injector temperature was set at 270 °C, the interface temperature was 270 °C, and the injection volume was 1 μL. The temperature program for determination of PBDEs was as follows: initial temperature was set at 120 °C, held for 2 min, raised at 15 °C/min to 230 °C, at 5 °C/min to 270 °C, and at 9 °C/min to 325 °C, and held for 2 min. For indicator PCBs, the temperature program was as follows: initial temperature was set at 110 °C, held for 1 min, raised at 15 °C/min to 180 °C, held for 1 min, then raised at 3 °C/min to 300 °C, and held for 2 min.

Seven PBDE congeners and seven indicator PCB congeners were detected in most food composite samples. The total PBDE and total indicator PCB concentrations were calculated by summing all of the concentrations of the seven individual PBDE congeners and seven indicator PCBs congeners, respectively, and the concentration of the undetected congener was set to half of the limit of detection (LOD).

Quality Assurance and Quality Control. One test of procedural blank was carried out for every eight samples. The recoveries of internal standards were all in the range of 51−117%. The range of LOD was 0.01−0.50 pg g^{-1} fresh weight (fw) for PBDEs and 0.01−0.40 pg g^{-1} fw for indicator PCBs. The laboratory performance was validated by successfully participating interlaboratory comparison of PBDEs and indicator PCBs in food organized by the Norwegian Institute of Public Health in 2010−2012. Z scores for total PBDEs (u/BDE-209) and indicator PCBs were in the ranges from −0.41 to 0.70 and from −0.063 to 1.5, respectively.

Estimation of Dietary Intake and Risk Assessment. Estimated dietary intake of PBDEs and indicator PCBs was calculated by multiplying the concentration of PBDEs and indicator PCBs (pg g^{-1}, fw) in each food composite from each region by the consumption data, respectively. The mean and the 97.5th percentile of the daily exposure levels were used to represent the dietary exposure for the average and high consumer, respectively.

A margin of exposure (MOE) approach was used for the risk assessment of dietary intake of PBDEs by comparing estimated dietary exposure with benchmarker dose lower confidence limit 10% ($BMDL_{10}$).[20] Of the seven PBDE congeners determined in the present study, relevant toxicity data were available only for BDE-47, -99, and -153. Therefore, risk assessment could be carried out only for these three individual PBDEs congeners. On the basis of the effects on neurodevelopment as the critical end point and an estimation of chronic human dietary intake, $BMDL_{10}$ recommended by the European Food Safety Authority (EFSA) was 172 ng kg^{-1} bw day^{-1} for BDE-47, 4.2 ng kg^{-1} bw day^{-1} for BDE-99, and 9.6 ng kg^{-1} bw day^{-1} for BDE-153, respectively.[21]

Statistical Analysis. The data of dietary intake of PBDEs and indicator PCBs were log-transformed before statistical analysis was performed. Spearman correlation analyses were used to investigate correlations between dietary exposure and age, the amount of food consumption, and human body burden of these pollutants. A *t* test was applied to determine the difference of levels of PBDEs and indicator PCBs in foodstuffs as well as sexual difference of dietary exposure. All statistical analyses were performed with the SAS software package (version 8.2; SAS Institute Inc., Cary, NC, USA). All *p* values are two-tailed, and α was set at a significance level of 0.05.

RESULTS

Concentrations of PBDEs and indicator PCBs in various food groups are summarized in Table 1. Relatively high levels of $\sum$PBDEs were found in meat samples and aquatic samples with concentrations of 192.5 ± 237.5 (mean ± standard deviation) and 190.6 ± 102.7 pg g^{-1} fw, respectively. PBDE concentration in milk was very low and comparable to that in plant origin food, including cereals, legumes and nuts, potatoes, and vegetables. For indicator PCBs, the highest concentrations were found in aquatic foods with the concentration of 628.7 ± 337.1 pg g^{-1} fw.

Dietary intake of PBDEs and indicator PCBs for average and high consumers based on body weight results by age/sex groups are listed in Table 2. The dietary exposures to PBDEs for average and high consumers varied from 0.7 to 1.5 ng kg^{-1} bw day^{-1} and from 2.0 to 4.2 ng kg^{-1} bw day^{-1}, respectively.

Table 2. Estimated Dietary Exposure to PBDEs and Indicator PCBs for Various Age/Sex Groups in China

			$\sum_7$PBDEs (ng kg^{-1} bw day^{-1}, ND = 1/2LOD)		$\sum_7$PCBs (ng kg^{-1} bw day^{-1}, ND = 1/2LOD)	
age group	(years)	gender	mean	97.5th percentile	mean	97.5th percentile
children	3−6	male	1.5	4.0	4.5	13.4
		female	1.5	4.5	4.6	14.0
	7−12	male	1.3	3.9	3.5	10.1
		female	1.3	4.2	3.7	11.3
	13−17	male	0.9	2.6	2.5	8.6
		female	0.9	2.8	2.5	8.3
adults	18−44	male	0.8	2.7	2.4	8.2
		female	0.8	2.3	2.4	7.9
	45−59	male	0.7	2.3	2.3	8.4
		female	0.7	2.0	2.2	7.7
	≥60	male	0.7	2.3	2.3	6.9
		female	0.7	2.0	2.3	7.0

For indicator PCBs, the dietary exposures for average and high consumers varied from 2.2 to 4.6 ng kg^{-1} bw day^{-1} and from 6.9 to 14.0 ng kg^{-1} bw day^{-1}, respectively. No sexual difference was found. Average estimated dietary intakes of PBDEs and indicator PCBs for the adult Chinese population were 0.76 and 2.34 ng kg^{-1} bw day^{-1}, respectively. A strong correlation between dietary intake of PBDEs and that of PCBs was found in this study (r = 0.73, p < 0.01), indicating coexposure to PBDEs and indicator PCBs through the diet in China.

The average estimated daily intakes (EDI) of PBDEs and PCBs for adults from various regions by various food groups are depicted in Figures 1 and 2, respectively. Among various regions in China, the minimum dietary intake of PBDEs was 0.35 ng kg^{-1} bw day^{-1} found in Shanxi, and the maximum was 1.24 ng kg^{-1} bw day^{-1} found in Guangxi. The highest percentage contributor to average dietary intake in China corresponded to meats (32%), followed by cereals (27%), vegetables (16%), and aquatic foods (15%). For PCBs, the dietary intake varied by a factor approximately 7 from the minimum of 0.7 ng kg^{-1} bw day^{-1} in Jiangxi to the maximum of 4.5 ng kg^{-1} bw day^{-1} in Fujian. Aquatic foods, cereals, and vegetables made comparable contributions to average dietary intake of PCBs in China. The percentage contributions of aquatic foods, cereals, and vegetables were 28, 24, and 25%, respectively.

DISCUSSION

PBDEs and Indicator PCBs in Foods. Previous studies have investigated levels of PBDEs and indicator PCBs in various foods, showing that the most heavily contaminated foods generally correspond to fish and shellfish or other aquatic foods.[2,22−25] However, the highest PBDE contamination corresponded to meats in the present study, and the mean level of PBDEs in aquatic foods was comparable to that in meats. The levels of PBDEs in aquatic foods in this study were relatively lower than that from some European countries[22,24−28] and North America.[29,30] This difference might be partly because the aquatic food group was mainly composed of various freshwater-farmed fishes that generally have markedly lower contamination levels than seawater fishes in China.[12,14,31] Like previous studies, the most abundant congener found in aquatic food composites was BDE-47, which accounted for half of the total PBDEs concentration, but the contribution of BDE-99 was negligible in the present study.[2]

A recent study has detected relatively high levels of PBDEs in meat from Nanjing, China.[12] In the present study, high PBDE levels were also found in meat composite samples, which were higher than that from the United States and European countries, suggesting relatively high PBDE contamination in meats from China.[24−30] For milk products, the very low PBDE contamination level, which was comparable to that of plant origin food, was mainly because the Chinese used to consume liquid milk rather than solid dairy products. Some studies have revealed relatively high PBDE levels in solid dairy products such as butter and cheese but very low contamination levels in liquid milk products such as whole milk and yogurt.[2,22,29,30] BDE-47 was the most abundant congener, followed by BDE-153 and BDE-99, and these congeners accounted for about 74% of the total concentration of PBDEs in milk. Food of plant origin is usually expected to have considerably low PBDE levels due to larger water content, low lipid content, and the primary position in the ecosystem, which was confirmed by the present

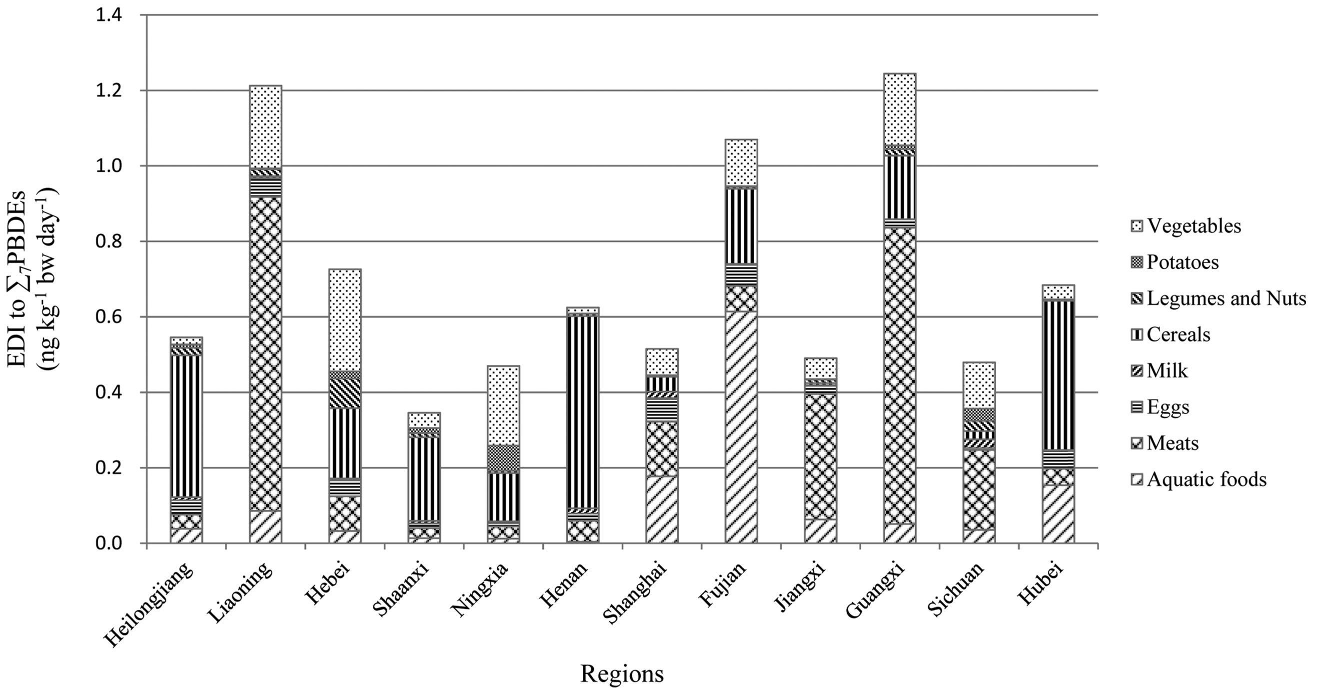

Figure 1. Average estimated daily intake (EDI) of $\sum_7$PBDEs for adults from various regions in China.

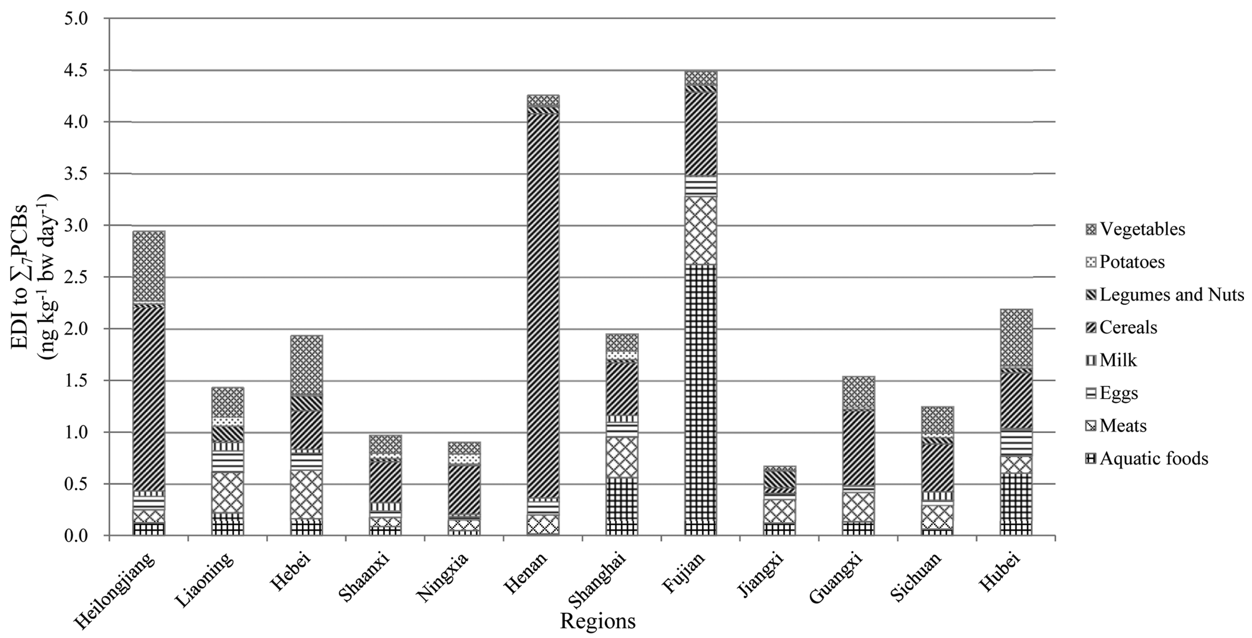

Figure 2. Average estimated daily intake (EDI) of $\sum_7$PCBs for adults from various regions in China.

study.[2] However, there was some overlap of the PBDE levels in plant origin foods with that in animal origin foods.

For indicator PCBs, the highest contamination also corresponded to aquatic food in this study. The indicator PCB levels in aquatic foods were much lower than those in other studies as well as the maximum levels for non-dioxin-like PCBs in foodstuffs proposed by the European Union, indicating relatively low indicator PCB contamination levels in common aquatic foods from China.[5,15,32,33] Moreover, indicator PCB levels were significantly higher than PBDE levels in these foods from China (paired t test, $p < 0.01$), which was consistent with our previous study on the determination of PBDEs and indicator PCBs in marine fish from China.[34]

Estimated Dietary Exposures. Many studies have been conducted in some countries or regions to systematically estimate dietary PBDE exposure for residents, and an overview of these studies is listed in Table 3. Because of different methodologies applied in various studies as well as different sampling years, the interpretation of these results should be cautious. The average dietary intake of PBDEs for adult Chinese was lower than that in Germany,[9] Norway,[28] Spain,[25] and the United Kingdom[22] and comparable to or slightly higher than that in Belgium,[26] Finland,[35] Sweden,[24] The Netherlands,[27] and Japan.[36] Although the values of dietary exposure to PBDEs in the United States[29] and South Korea[37] were higher

Table 3. Overview of Average Dietary Intake of PBDEs for Adult Population in Recent Studies

		dietary intake				
country/region	sampling year	ng day^{-1}	ng kg^{-1} bw day^{-1}	study approach	food groups	ref
Finland[a]	1997	44		market basket study/individual samples/ND = LOQ	fish, meat, and eggs, liquid milk products, solid milk products, fats, cereal products, potato products, vegetables, fruits and berries, and beverages, spices, and sweets	39
Norway[b]	2002−2006		1.47	market basket study/individual samples/ND = 0	fish, fish products, shell fish and marine oils, meat, dairy products, eggs, and some plant origin foods	29
Sweden[c]	2005		0.7	market basket study/individual samples/ND = 1/2LOD	fish and fish products, meat and meat products, dairy products, egg, fats	25
Belgium[b]	2005	35		market basket study/individual samples/ND = 1/2LOD	fish and seafood, meat products, eggs and dairy products, and fast food	31
The Netherlands[a]	2003−2004		0.79	market basket study/individual samples/ND = 1/2LOD	fish, meat, dairy, eggs, oils and fats, bakery products, bread and cereals, vegetables, sweets, complex dishes	30
Catalonia, Spain[d]	2006	75.4	1.1	market basket study/individual samples/ND = 1/2LOD	fish and shellfish, meat and meat products, eggs, milk, dairy products, vegetables, tubers, fruits, cereals, pulses, oils and fats, and bakery products	26
Germany[b]	2005		1.2	duplicate diet study		9
UK[b]	2003−2004		1.25	total diet study/composites samples /ND = LOD	fish, meat, poultry, eggs, milk, dairy products, and food groups of plant origin	23
USA[e]	2008		0.8−1.3	market basket study/individual samples/ND = 1/2LOD	fish, meat, dairy, and vegetable-based foods	27
Osaka, Japan[f]	2006	46		total diet study/composites samples/ND = 0	14 food groups, including animal origin foods and plant origin foods as well as water and beverages	40
South Korea[c]	NA	72.3		market basket study/individual samples/ND = 1/2LOD	fish and shellfish, meats, eggs, and cereals	37
China[b]	2007	48.0[g]	0.76	total diet study/composites samples/ND = 1/2LOD	fish, meats, eggs, milk, cereals, legumes and nuts, tubers, and vegetables	this study

[a]Sum of BDE-47, -99, -100, -153, and -154. [b]Sum of BDE-28, -47, -99, -100, -153, -154, and -183. [c]Sum of BDE-15, -17, -28, -33, -47, -49, -66, -71, -85, -99, -100, -119, -126, -138, -153, -154, -183, and -209. [d]Sum of BDE-47, -99, -100, -153, -154, and -183. [e]Sum of BDE-28, -47, -49, -66, -85, -99, -100, -138, -153, -154, -183, -203, and -209. [f]Sum of BDE-17, -25, --28, -30, -32, -33, -35, -37, -47, -48, -49, -66, -71, -75, -77, -85, -99, -100, -116, -118, -119, -126, -138, -153, -154, -155, and -166. [g]For an adult of 63 kg bw.

Table 4. Overview of Margins of Exposure (MOEs) for BDE-47, -99, and -153 for Different Population Groups

	BDE-47				BDE-99				BDE-153			
	estimated intake (ng kg^{-1} bw day^{-1})		MOE		estimated intake (ng kg^{-1} bw day^{-1})		MOE		estimated intake (ng kg^{-1} bw day^{-1})		MOE	
age group (years)	AC[a]	HC[b]	AC	HC	AC	HC	AC	HC	AC	HC	AC	HC
3−6	0.4	1.5	384	116	0.3	0.7	16	6	0.3	1.4	38	7
7−12	0.3	0.9	518	182	0.2	0.5	20	8	0.2	1.2	42	8
13−17	0.2	0.7	733	258	0.1	0.4	28	11	0.2	0.9	59	11
18−44	0.2	0.7	821	247	0.1	0.4	31	10	0.1	0.7	69	14
45−59	0.2	0.6	916	304	0.1	0.4	34	11	0.1	0.6	82	16
≥60	0.2	0.5	922	337	0.1	0.3	35	12	0.1	0.7	78	14

[a]Average consumers. [b]High consumers.

than those of the present study, it was difficult to draw conclusions because BDE-209 was involved.

The dietary exposure to PBDEs of residents is usually affected by some factors such as age, sex, catering culture, and so on. Some studies have shown a decreasing trend of dietary intake of PBDEs per kilogram of body weight with age.[38,39] In the present study, there was a significantly negative correlation between dietary intake of PBDEs and age because of higher food consumption per kilogram of body weight for young individuals. Moreover, although no sexual difference of dietary PBDE exposure per kilogram of body weight was found, the amount of dietary intake of PBDEs per day for males was significantly higher than that for females because of higher food consumption for the males (t test, $p < 0.01$). Moreover, cooking processes such as broiling could reduce the amount of PBDEs in certain animal origin foods due to dripping fat, suggesting cooking might reduce the dietary intake of PBDEs.[26,40] However, a recent Spanish study has shown that cooking does not always lead to the reduction of PBDE levels in foods, and the influence of cooking on PBDE levels depended not only on the particular cooking process but also on the specific food item.[41] Thus, it is necessary to take into consideration cooking process on contamination levels in foods for the actual estimation of dietary exposure.

The relative contribution of different food groups to total dietary PBDE intake might vary considerably mainly due to various food habits and culture. In European countries, fish is usually the main contributor to the dietary PBDE exposure, but other food groups, such as dairy products and meats, hav also shown high contribution to the dietary intake.[22,24−28,35] In Japan, fish is the predominant contributor to the dietary PBDEs because of the consumption of large quantities of fish.[36,42] In the United States, the highest contributor to dietary PBDE intake corresponded to meats rather than fish because of the tendency to eat less fish in North America.[29,30,38] In the present study, large variations in relative contribution of various food groups to dietary PBDE intake were also found among regions in China. In general, meats would make higher contribution than aquatic foods in most regions. Aquatic foods made the highest contribution only in Shanghai and Fujian, where relatively large amounts of aquatic foods were consumed. The contribution of milk to dietary PBDE intake could be negligible due to lower consumption of milk as well as very low contamination level in China. Moreover, impressively high contribution to total dietary PBDE intake was made by plant origin foods, which would result from considerable consumption of these foods in China, although very low levels of PBDEs were detected. Thus, plant origin foods should not automatically be omitted from the evaluation of dietary exposure.

The human body burden of PBDEs for the general population was estimated by determination of PBDEs in human breast milk samples in China.[19] There was a weak but significant correlation between levels of total PBDEs in breast milk and dietary PBDE exposure per kilogram of body weight ($r = 0.69$, $p < 0.05$), indicating that dietary intake should be a major route of PBDE exposure in China. The weak correlation might be caused by other exposure routes. Besides dietary intake, inhalation of indoor air is a credible pathway of exposure, which accounted for approximately 12% of the average human daily exposure to PBDEs in the United Kingdom.[7] Ingestion of dust would be a more important exposure pathway in North America, and exposures to PBDEs in dust accounted for approximately 82% of the overall PBDEs intake.[8,10,43,44] However, the diet was proved to be the main source of PBDE exposure in Europe, which accounted for >90% of the total exposure.[9,45] This difference in the contribution of exposure pathways to total PBDEs is the probable cause of the human body burden of PBDEs detected in North America being 1−2 orders higher than that in Europe, although dietary exposures were comparable between Europe and North America.[46] The human body burden of PBDEs and dietary exposure in China were all comparable or slightly lower than that in Europe, suggesting the pattern of exposure pathways might be similar between China and European. A recent study has indicated that dietary intake could account for 98% of overall PBDE intake in Shenzhen, China.[14] However, the ratio of levels of PBDEs in breast milk (data were shown in our previous study[19]) to dietary PBDE exposure per kilogram of body weight varied from 1.6 to 4.7, suggesting that relative contribution of various PBDE exposure pathways to overall human exposure might be different among regions in China.

For indicator PCBs, dietary exposure of the adult has been evaluated in France,[5] Italy,[33] and The Netherlands,[15] with average estimated dietary exposures of 7.7, 5.6, and 10.9 ng kg^{-1} bw day^{-1}, respectively. By comparison, dietary intake of indicator PCBs was much lower in China, probably because of scarce commercial produce and usage of PCB in China. Moreover, fish and meat were the main contributors to the total intake of indicator PCBs in these European countries. However, unlike these studies, plant origin foods such as cereals and vegetables made the highest contribution to total dietary intake in almost all regions in China, which might stem from the considerable consumption of these foods.

Risk Assessment. The calculated margins of exposure (MOEs) of average and high consumers shown in Table 4 ranged from 16 (BDE-99) to 922 (BDE-47) and from 6 (BDE-

99) to 337 (BDE47), respectively. According to the conclusions of EFSA, a MOE >2.5 might indicate that there is no health concern.[21] Therefore, these data might suggest that there was unlikely a health concern with respect to current dietary exposure to PBDEs in China. However, a recent study by EFSA has indicated that younger children (1−3 years) might have higher potential health risk than other population groups through dietary intake of PBDEs.[21] Given that children would be more sensitive to neurobehavioral toxicity, it behooves us to pay attention to the potential health risk of dietary PBDEs of children. Thus, further studies must be done to recognize the health risk of dietary exposure to PBDEs of small children in China. On the other hand, a reference threshold of BDE-99, ranging from 0.23 to 0.30 ng kg^{-1} bw day^{-1}, was established on the basis of the reproductive toxicity in a Dutch study.[27] The average adult consumer of child-bearing (18−44 years) reached this threshold, and high consumers were much higher than the threshold. It is notable that the toxicological values presented in this study present a source of uncertainty because of possible cumulative effects between congeners as well as limited toxicological information of many individual congeners of PBDEs. In addition, there are also some uncertainties associated with the methodology used in this study, including intake calculations, consumption statistics, and sample representativeness. Nevertheless, further studies and strict legislation must be done to reduce POPs contamination in foods and levels of dietary exposure for the protection of health in China.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Values of average consumption of selected food groups for adults from various regions and main food items comprising various selected food groups. This material is available free of charge via the Internet at http://pubs.acs.org.

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Funding

This study was supported by the National Science and Technology Support Program of China (Grants 2007BAC27B02 and 2011BAK10B07), Science and Technology Projects of the Ministry of Health (Grant 200902009), National Nature Science Foundation of China (20907048), and the Special Foundation for Young Scientists of Department of Health of Hubei Province (QJX2010-33).

Notes

The authors declare no competing financial interest.

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