Research

Assessment of Human Exposure to Polybrominated Diphenyl Ethers in China via Fish Consumption and Inhalation

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This study examined human exposure to polybrominated diphenyl ethers (PBDEs) associated with fish consumption and inhalation in China. The median intake of Σ_7 PBDEs via human milk was 48.2 ng/day for nursing infants (0−1 years old) (a range of 23.4−99.1 ng/day). For all other age groups, the median intake of Σ_{11} PBDEs via fish consumption was 1.7−12.9 ng/day with a range of 0.59−56.3 ng/g. Additionally, human exposure to PBDEs via inhalation was 2.7−9.2 ng/day (a range of 0.72−108 ng/day). The median total Σ_{11} PBDEs intakes for nursing infants (6874 and 7372 pg/kg b.w./day for males and females, respectively) were much higher than other age groups (215−608 pg/kg b.w./day). No significant difference in the total PBDEs intakes was found between males and females. Of the 11 PBDEs congeners, BDE-47 was predominant in the total intake for nursing infants with a mean contribution of 38%, whereas BDE-209 was the dominant congener of total intake for other age groups, varying from 44 to 61%. Currently, the PBDEs levels in Chinese consumer fish and the total intakes of PBDEs via fish consumption were at the lower end of the global range. Compared with similar studies in other countries, however, human exposure to PBDEs via inhalation in China was relatively high. Overall, estimated daily intake of total PBDEs in the Chinese population was far below the LOAEL. However, studies are needed to further understand the fate and impact of PBDEs as PBDE-containing products are still used widely in large quantities in China.

Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of man-made chemicals widely used as flame retardants in electronics, furniture, automotives, textiles, foam, and household plastic products (*1*). For example, the concentration of PBDEs added into some polyurethane foam can range from 10 to 30% of the product by weight (*2*). These PBDEs are present in three commercial mixtures, primarily referred to as penta-, octa-, and deca-BDE. The global demand for PBDEs was estimated at ∼67 490 tons in 2001 (11, 6, and 83% were produced as penta-, octa-, and deca-BDE, respectively), of which 49% was consumed in North America, 37% in Asia, and 12% in Europe (*3*). As a result of the widespread use of PBDE-containing products, PBDEs are commonly detected in air, water, sediment/soil, biota, and human tissues (*4*−*7*). Structurally similar to PCBs and dioxins, PBDEs are considerably persistent, bioaccumulative in the fatty tissues of organisms, and biomagnifiable throughout food chains (*8*). PBDEs were also shown to move from local sources to polar regions through long-range atmospheric transport (*9*).

The concentrations of PBDEs in human samples have increased by a factor of ∼100 over the last 30 years (*10*). The presence of PBDEs in humans is of particular concern due to their potential ability to cause thyroid hormone disruption, neurodevelopment deficits, and cancer (*11*). Parallel to other persistent organic chemicals, diet is the main route for human exposure to PBDEs, although some researchers recently suggested that home dust might play an important role for infants and children (*12*−*14*). The contributions of diet and inhalation to PBDEs intake amounted to 93 and 7%, respectively, in the U.K. (*15*), and were about 96 and 4%, respectively, in Canada (*16*). Clearly, fish consumption is a main route for human exposure to PBDEs (*17*−*21*). Furthermore, some studies found a positive relationship between PBDE concentrations in human samples and dietary intake of fish and shellfish (*22*, *23*).

The domestic demand for brominated flame retardants (including PBDEs) in China has increased at an annual rate of 8% (*4*), which would inevitably result in continuously rising PBDE levels in the environmental media. On the other hand, China (mainland) is the largest fishery producer and exporter in the world (*24*). It is clear that systematic assessments of human exposure to PBDEs via fish consumption are a critical step toward the institution of effective measures to ensure food safety. Unfortunately, no human exposure assessment regarding PBDEs has been conducted in China. The present study aimed to estimate human daily intake of PBDEs and to assess potential health risk of PBDEs via fish consumption and inhalation for twelve groups of Chinese persons with different age/sex.

Materials and Methods

Sample Collection. Thirteen fish species were selected based on the fish-consuming habits in China, including seven freshwater farmed fish, i.e., tilapia (*Tilapia*), grass carp (*Ctenopharyngodon idellus*), bighead carp (*Aristichthys nobilis*), bluntsnout bream (*Megalobrama amblycephala*), largemouth bass (*Micropterus salmoides*), mandarin fish (*Siniperca chuatsi*), and northern snakehead (*Ophicephalus argus*); three seawater farmed fish, i.e., red drum (*Sciaenops Ocellatus*), snubnose pompano (*Trachinotus blochii*), and crimson snapper (*Lutjanus erythopterus*); and three wild marine fish, i.e., hairtail (*Trichiurus lepturu*), golden thread (*Nemipterus virgatus*), and common mullet (*Mugil cephalus*). In November 2004 and January 2005, approximately 30 individuals (a total of 390 samples) for each species were randomly acquired in local fish markets and supermarkets from 11 typical fishery-producing regions of Guangdong Province, China (Figure S1, "S" designates figure and tables in the Supporting Information), to ensure sufficient statistical power for data analysis (*25*). Upon shipping to the laboratory with ice, about 20 g (wet weight) of fish muscle without skin were taken for measurements of 11 PBDE congeners,

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including BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183, and -209.

Sample Preparation and Extraction. Detailed extraction and instrumental analytical procedures of PBDEs were described previously (*26*). Briefly, we spiked the surrogate standards (^{13}C−PCB-141 and PCB-209) to each sample before extraction with a mixture of acetone and hexane (1:1, v:v). Lipid contents were determined gravimetrically using 20% of each extract. The remaining extract was subject to a gel permeation chromatography column and eluted with a mixture of dichloromethane:hexane (1:1, v:v). The fraction from 90 to 280 mL were collected and concentrated to 4 mL. Two milliliters of the extract were further purified with a 10mm i.d. silica /alumina column. We eluted the column with 70 mL of dichloromethane:hexane mixture (1:1, v: v), and concentrated the effluent to 100 μL under a gentle stream of N_2. The internal standard (^{13}C−PCB-208) was added to the final extract for instrumental analysis. We quantified 11 PBDE congeners using a Shimadzu model 2010 gas chromatograph coupled with a model QP2010 mass spectrometer (Shimadzu, Japan), at the wet weight-based limits of detection (LOD) from 1.2 to 2.5 pg/g for all tri- to hepta-BDEs and 100 pg/g for BDE-209.

Quality Assurance/Quality Control. One procedural blank, one spiked blank, one matrix spiking sample (11 PBDE congeners spiked into preextracted fish sample), and one matrix spiking duplicate for each batch of 20 fish samples were processed, as part of a strict quality assurance/quality control program. The spiked samples contained the 11 target BDE congeners. Recoveries of the 11 PBDE congeners, ^{13}C−PCB-141, and PCB-209 were 87.2 ± 19.0, 81.8 ± 17.3, and 84.8 ± 18.4%, respectively. Because levels of BDE-47 (<1.7 pg/g wet weight) in the procedural blanks were lower than 5% of those in fish samples, we did not subtract the blank values from the sample measurements. We also did not correct reported concentrations based on the surrogate recovery data. Concentrations below the LOD were assumed to be 1/2 LOD in the calculation and assessment.

Human Exposure Estimate. A questionnaire-based fish consumption survey was conducted in 12 cities of Guangdong Province in April 2006. A total of 1536 people participated in our survey, divided into age groups of 2−5 years (4 male and 5 female), 6−17 years (204 male and 194 female), 18−44 years (495 male and 490 female), and 45−59 years (48 male and 39 female), and ≥60 years (28 male and 29 female). Fish consumption quantity for each age group was shown in Table S1. Estimated daily intake of PBDEs (EDI_{PBDEs}) via fish consumption ($EDI^{fish}_{\Sigma_{11}PBDEs}$) was calculated by multiplying the fish PBDE concentrations (Σ_{11} PBDEs, sum of all target BDE congeners) and the amount of fish consumption from the survey. Human exposure to PBDEs via inhalation ($EDI^{air}_{\Sigma_{11}PBDEs}$) was estimated using PBDEs concentrations (Σ_{11}-PBDEs) reported previously (*6*), in combination with the inhalation rate data from the U.S. Environmental Protection Agency (*27*). In June 2004, Chen et al. (*6*) collected 32-pair samples from four sites, including two industry sites, one urban site, and one city background cite, in the city of Guangzhou, a typical urban center in South China to examine 11 PBDE congeners (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183, and -209). The LOD ranged from 0.14 to 0.58 pg/m^3 for all tri- to hepta-BDEs, and 14.3 pg/ m^3 for BDE-209 when an average air volume of 700 m^3 was collected. In addition, EDI_{PBDEs} via human milk ($EDI^{human\ milk}_{\Sigma_7PBDEs}$) was calculated on the basis of PBDEs levels in breast milk (*5*) and infant ingest (688 mL/day) (*27*), assuming that human milk is the only food source for nursing infants (0−1 years old). Bi et al. (*5*) collected 27 breast milk samples in South China to determine the occurrence of BDE-28, -47, -99, -100, -153, -154, and -183. BDE-209 was not quantified due to the high interference level in the blank samples. The LOD of individual PBDE congeners ranged from 0.01 to 0.07 ng/g. In the present study, human exposure to PBDEs via different routes was assumed as 100% absorption.

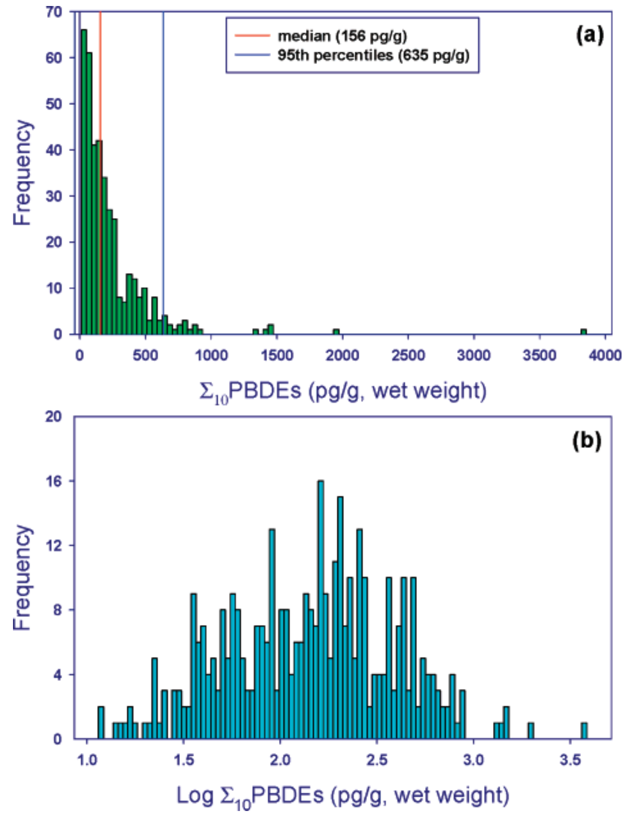

FIGURE 1. (a) Histogram showing the distribution of Σ_{10} PBDEs concentrations (pg/g, wet weight) in fish from China. The red line marks the median (156 pg/g), the blue line marks the 95th percentile concentration (635 pg/g); (b) Histogram showing the distribution of Log Σ_{10} PBDEs concentrations (pg/g, wet weight) in fish from China.

Data Analysis. The normality of the distribution was tested using nonparametric test (Kolmogorov−Smironov *Z*). Differences between estimated daily intakes of PBDEs in different groups were evaluated using analysis of variance (ANOVA) (SPSS v13.0).

Results and Discussion

Levels of PBDEs in Fish. Table S2 summaries the PBDEs concentrations in Chinese consumer fish presented on a wet weight basis (fat content provided), expressing as parts per trillion (pg/g). We detected PBDEs in 389 fish samples. Of the 390 fish samples analyzed, the median and mean concentrations of Σ_{10}PBDEs (sum of all target BDE congeners except BDE-209; 1/2 LOD was used for undetectable BDE congeners) were 156 pg/g and 231 pg/g, respectively. The PBDEs concentrations were slightly higher than those reported in our previous study (median and mean concentrations of Σ_{10} PBDEs were 153 and 226 pg/g, respectively), where undetectable concentrations were set as zero (*26*). When fish species were compared, snubnose pompano had the highest PBDEs levels with a median of 426 pg/g, followed by hairtail (272 pg/g), mandarin fish (254 pg/g), crimson snapper (228 pg/g), red drum (224 pg/g), largemouth bass (199 pg/g), bluntsnout bream (188 pg/g), northern snakehead (140 pg/g), grass carp (89.0 pg/g), tilapia (78.3 pg/g), golden thread (57.2 pg/g), and common mullet (38.5 pg/g). The PBDEs levels in fish from China were log-normally distributed as determined by one-sample Kolmogorov−Smirnov test (p >0.05; Figure 1b), although a large variation was observed

(Figure 1a). The highest PBDEs level was 3853 pg/g found in a hairtail fish, whereas only 5% of the samples contained Σ_{10}PBDEs higher than 635 pg/g.

When compared with similar studies conducted in other countries or regions recently, the median and mean PBDEs concentrations (156 and 231 pg/g wet weight, respectively) were lower in China. Schecter et al. (*28*) reported median and mean concentrations of 616 and 1120 pg/g wet weight, respectively, in fish from three large supermarkets in Dallas, Texas. In 2005, Domingo et al. (*29*) determined the PBDEs contents in 14 edible marine species widely consumed by the population of Catalonia (Spain), and the mean total (tetra- to octa-BDE) PBDEs was 564 pg/g wet weight. Darnerud et al. (*19*) reported the sum of five PBDEs in market fish collected from four major Swedish cities with a mean concentration of 634 pg/g wet weight. Voorspoels et al. (*21*) obtained the low bound (not detected was replaced by zero) of average PBDEs levels at 450 pg/g wet weight in fish from Belgium. Tittlemier et al. (*30*) analyzed retail samples of fish and shellfish ($n = 122$) from three Canadian cities and reported that the geometric means of total PBDEs concentrations in trout, salmon, char, and tilapia were 1600, 1500, 620, and 180 pg/g wet weight, respectively. The results from the present study were relatively low compared to these previous results, but similar to the results from another survey of PBDEs levels in Spanish commercial fish samples (with a median value of 189 pg/g wet weight)(*31*).

The reasons that we observed relatively lower PBDEs levels in fish from China than those in other countries or regions may be due to the usage patterns of PBDEs-containing commercial products. The consumption of penta-BDE in Asia was 150 tons in 2001, accounting for 2.0% of the global demand, far less than the amount used in North America (7100 tons) (*3*). Interestingly, as a major congener (98%) in commercial deca-BDE products (*11*), BDE-209 was only found in 14 of the 390 fish samples with a range from <100 to 565 pg/g wet weight. The detection ratio of BDE-209 was only 3.6% of the total fish samples although deca-BDE commercial products have been the main brominated fire retardants used in China (*4*, *3*). However, it must be noted that the LOD of BDE-209 (100 pg/g wet weight) was much higher than those of other PBDE congeners (ranging from 1.2 to 2.5 pg/g wet weight). On the other hand, one possible explanation is the lower bioavailability of BDE-209 compared to other PBDE congeners due to its relatively higher molecular weight and larger octanol−water partition coefficient (log K_{ow} is approximately 10) (*32*). Another plausible explanation is the debromination of BDE-209 within fish tissue. Stapleton et al. (*33*) exposed juvenile carp to BDE-209 via diet and detected hexa- and hepta-BDE, metabolites of BDE-209. In a more recent study, several hepta-, octa-, and nona-BDE congeners were found in juvenile rainbow trout tissues as a result of BDE-209 debromination (*34*). In addition, many studies showed that BDE-209 could be degraded into lower brominated congeners under various conditions, such as photolytic breakdown under ultraviolet light or natural sunlight (*35*−*36*), reductive debromination by zero-valence iron sulfide and sodium sulfide (*37*), and microbial debromination (*38*).

Estimated Daily Intake of PBDEs via Fish Consumption. As shown in Table S1, the median $EDI^{fish}_{\Sigma_{11}PBDEs}$ was 1.7−12.9 ng/day for all groups, ranging from 0.59 to 56.3 ng/day. Due to the limited size of the population under investigation (e.g., only four male and five female samples for the 2−5 year old group), the $EDI^{fish}_{\Sigma_{11}PBDEs}$ results should be interpreted with caution. Except for the nursing infants (the 0−1 years old group), $EDI^{fish}_{\Sigma_{11}PBDEs}$ (10.1−12.9 ng/day) for older people (≥60 years old) were slightly higher than those for adults (18−44 years old and 45−59 years old; 5.1−12.9 ng/day), adolescents (6−17 years old, 5.1−5.2 ng/day), and children (2−5 years

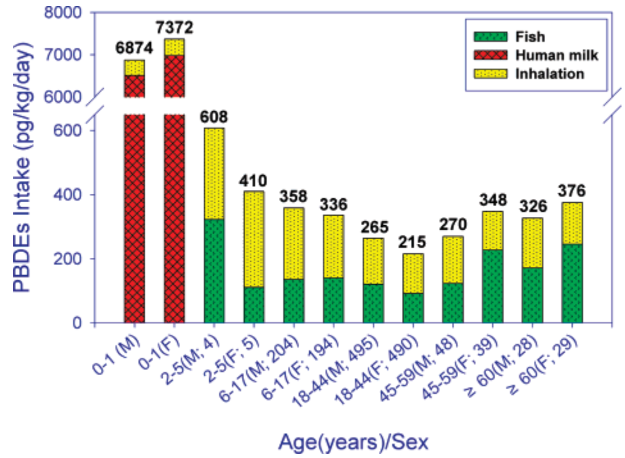

FIGURE 2. The total PBDEs intake of China population by different age/sex groups (pg/kg b.w./day). The numbers on the bar shows the values of total PBDEs intakes, and the numbers in the parentheses are the sample sizes of participants in the survey.

old; 1.7−5.2 ng/day). This may be attributed to the difference in dietary habits that older people consume more fish daily than people in other age groups. However, no significant difference in $EDI^{fish}_{\Sigma_{11}PBDEs}$ ($F = 0.027$, $p > 0.05$) was found between male and female for all age groups (Table S1). When compared with human intake of PBDEs via fish consumption in other countries, our median $EDI^{fish}_{\Sigma_{11}PBDEs}$ for adults (5.1−12.9 ng/day) was slightly lower than those reported in Sweden (mean = 23.1 ng/day) (*19*), Catalonia of Spain (mean = 20.8 ng/day) (*29*), Belgium (mean = 13 ng/day) (*21*), Finland (mean = 23 ng/day) (*18*), the U.S. (median = 4.5−15.7 ng/day) (*28*), and The Netherlands (mean = 29.6 and 15.9 ng/day for 2001/2002 and 2003/2004, respectively) (*20*), but higher than that reported in Canada (mean = 1.3 ng/day) (*39*).

Estimated Daily Intake of PBDEs via Human Milk. The median $EDI^{human\ milk}_{\Sigma_{7}PBDEs}$ was 48.2 ng/day for nursing infants (0−1 years old) with a range of 23.4−99.1 ng/day (Table S1). Compared with exposure of PBDEs via inhalation, human milk was the main route and attributed to 95% of the total intake (Figure 2). This result supported the viewpoint by other researchers that babies accumulated PBDEs by placental transfer from nursing on breast milk (*40*). Our median $EDI^{human\ milk}_{\Sigma_{7}PBDEs}$ (48.2 ng/day) was lower than newborn PBDEs intakes in Sweden (96 ng/day), the United Kingdom (U.K.) (210 ng/day), and the United States (1770 ng/day) reported by Stapleton and co-workers (*12*).

Exposure via Inhalation. The median $EDI^{air}_{\Sigma_{11}PBDEs}$ was 2.7−9.2 ng/day, ranging from 0.72 to 108 ng/day for all groups (Table S1). Also, the median $EDI^{air}_{\Sigma_{11}PBDEs}$ (including males and females) were 0.76, 1.3, 2.0−2.4, 1.9−2.6, and 1.9−2.6 ng/day for the nursing infants, children, adolescents, adults, and older people, respectively. In addition, the median $EDI^{air}_{\Sigma_{11}PBDEs}$ were relatively constant for all ≥6 year old age groups (6.8−9.2 ng/day) with slightly higher values for males than for females. Currently, only two publications are available in the literature dealing with human intake of PBDEs via inhalation. During 2003 and 2004, Wilford et al. (*16*) collected air samples randomly in 74 selected homes and seven outdoor sites in Ottawa, Canada and estimated the human exposure to PBDEs via inhalation (median values of 2.0 ng/day and 1.9 ng/day for male and female, respectively). Harrad et al. (*15*) collected air samples from a range of office and home indoor microenvironments in the U.K. and obtained a median human intake of PBDEs of 6.9 ng/day. Clearly, our median $EDI^{air}_{\Sigma_{11}PBDEs}$ was slightly higher than those in Canada and the U.K., which may be attributed to

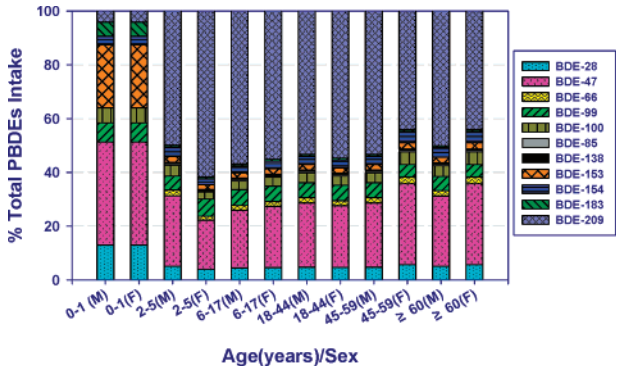

FIGURE 3. Congener-specific contributions of total PBDEs (including 11 PBDE congeners) intake by age and sex.

the different PBDE analytes used. In the present study, 11 PBDE congeners (including BDE-209) were examined, and BDE-209 was the dominant congener, with an average relative abundance of 72, 79, 48, and 70% in four sampling sites (*6*). On the other hand, no BDE-209 was included in the studies conducted in Canada and the U.K. In this study, the PBDEs levels in ourdoor air samples were used the calculation the estimated daily intake via inhalation due to the lack of PBDEs data for indoor air. As a result, the daily intake of PBDEs via inhalation by the Chinese population may have been underestimated because the PBDEs levels in these two environments may remain somewhat different based on the previous studies (*15*, *16*). However, the difference might be ignored because air exchange is typically adequate between the indoor and outdoor environments in China. Furthermore, human exposure to PBDEs via inhalation is rather complicated, which is dictated by many factors, such as gas−particle partitioning (*6*, *14*) and exposure duration (*41*). Therefore, this study should be regarded a preliminary evaluation of human exposure to PBDEs via inhalation.

On the other hand, the mean contributions of $EDI^{fish}_{\Sigma_{11}PBDEs}$ and $EDI^{air}_{\Sigma_{11}PBDEs}$ to the total PBDEs intake ($EDI^{total}_{\Sigma_{11}PBDEs}$, sum of $EDI^{fish}_{\Sigma_{11}PBDEs}$ and $EDI^{air}_{\Sigma_{11}PBDEs}$) were 48 and 52%, respectively, for all age groups except for nursing infants (Figure 2). This result indicated that fish consumption and inhalation were two significant routes for human exposure to PBDEs. However, it must be noted that BDE-209, although dominating in air samples, was present mainly in the particulate phase (*6*). Thus, the bioavailability of BDE-209 for humans via inhalation could be relatively low. Furthermore, no significant difference in the total PBDEs intakes was found between males and females ($F = 0.002$, $p > 0.05$). Several previous studies suggested that intake of PBDEs via fish consumption was dominant in overall intake through diet, as 47% of the total PBDEs intake from diet was attributed to fish consumption in Sweden, 30% in Spain, 38% in The Netherlands, 39% in Belgium, and 52% in Finland (*17*−*21*). Herein, we estimated human intake of PBDEs through diet ($EDI^{diet}_{\Sigma_{11}PBDEs}$) in China assuming that the contribution of $EDI^{fish}_{\Sigma_{11}PBDEs}$ to $EDI^{diet}_{\Sigma_{11}PBDEs}$ ranged from 20 to 60%. As a result, the mean $EDI^{diet}_{\Sigma_{11}PBDEs}$ for adults was 24.5 ng/day with a range of 2.8−64.5 ng/day. Furthermore, the contributions of diet and inhalation were 79 and 21% of the overall PBDEs intake (sum of $EDI^{diet}_{\Sigma_{11}PBDEs}$ and $EDI^{air}_{\Sigma_{11}PBDEs}$), which were slightly different to the results in the U.K. (93 and 7%) reported by Harrad et al. (*15*) and in Canada (around 96 and 4%) described by Wilford et al. (*16*).

Congener-Specific Distribution of PBDEs in Daily Intake. Figure 3 depicts the PBDE congener patterns observed in total PBDEs intakes via fish consumption and inhalation for Chinese population. Clearly, BDE-47 was the most abundant congener (38%) of the total PBDEs intake for nursing infants, followed by BDE-153 (23%), BDE-28 (13%), BDE-99 (7%), BDE-100 (6%), BDE-183 (5%), BDE-209 (4%), BDE-154 (3%), BDE-66 (0.05%), BDE-85 (0.03%), and BDE-138 (0.02%). However, for other age groups, BDE-209 was the predominant congener with a range from 44 to 61%. BDE-47 was the second most abundant congeners, accounting for 18−30% (both male and female). The remaining congeners followed the descending abundance of BDE-99 (5%), BDE-28 (5%), BDE-100 (4%), BDE-154 (3%), BDE-153 (2%), BDE-66 (2%), BDE-183 (0.7%), BDE-85 (0.6%), and BDE-138 (0.5%).

Assessment of Human Exposure to PBDEs. We obtained the total PBDEs intake per body weight using $EDI^{fish}_{\Sigma_{11}PBDEs}$, $EDI^{air}_{\Sigma_{11}PBDEs}$, $EDI^{human\ milk}_{\Sigma_{7}PBDEs}$, and body weight. The median total PBDEs intakes for nursing infants were the highest with 6874 pg/kg b.w./day for male and 7372 pg/kg b.w./day for female (Figure 2). For other groups (≥2 year groups), the median total PBDEs intake was 215−608 pg/kg b.w./day. Our total daily intake of PBDEs via fish consumption and inhalation for adults (≥18 year groups) was estimated at only 215−376 pg/kg b.w./day, lower than those from dietary sources reported in other countries, such as in the U.S. (0.9−1.2 ng/kg b.w./day) (*28*), Spain (1.2−1.4 ng/kg b.w./day) (*17*), Sweden (0.58−0.63 ng/kg b.w./day) (*42*), and The Netherlands (1.7 ng/kg b.w./day) (*20*). However, the present study was the first assessment of human exposure to PBDE via fish consumption and inhalation in China to our knowledge. Further research is definitely needed to examine other food sources, such as meat, dairy products, eggs, vegetables, cereals, fruits, fats, and oils. For example, meat products accounted for around 30% of the total dietary intake of PBDE in Belgium (*21*). In Sweden, the contributions of meat, diary products, egg, fats and oils, and pastry to the total daily PBDE intake were 14, 17, 2, 15, and 5%, respectively (*19*). Sjödin et al. (*43*) found that around 50% of the total dietary PBDE intake in Sweden originated from fish, while the rest was from meat and other dietary sources. In addition, home dust has been a concern in recent years due to the high PBDEs levels and may be regarded as the most important exposure route of PBDEs for children (*12*−*14*).

To assess the potential health risk caused by PBDEs, a LOAEL value (lowest observed adverse effect level) of 1 mg/kg b.w./day was recommended by Darnerud et al. (*44*) when the most sensitive end points are chosen based on thyroid hormone effects in rats. Clearly, the median total intake of PBDEs via fish consumption and inhalation for adults in China (215−608 pg/kg b.w./day) was far below the recommended LOAEL value. In recent years, however, many studies have shown that the half-lives of tetra-, penta-, and hexa-BDE between rats and humans are different (*20*, *45*). The half-life of BDE-99 (2.9 years) in humans is longer than those of BDE-47 (1.8 years) and BDE-100 (1.6 years) (*20*). Thuresson et al. (*45*) found that the half-lives of three nona-BDEs, four octa-BDE congeners, and BDE-209 were 18−39, 37−91, and 15 days, respectively, in blood donated by rubber workers and electronics dismantlers. In consequence, using daily intake of PBDEs to evaluate potential health risk may lead to imprecise toxic risk assessment. Therefore, DeWinter-Sorkina et al. calculated a maximal allowed intake level of 0.26 ng BDE-99/kg b.w./day based on the bioaccumulation properties of BDE-99 and similar assessment procedure to 2,3,7,8-TCDD (*20*). In present study, the total median intakes of BDE-99 was 12−30 pg/kg b.w./day for ≥2 year old age groups, which much lower than the maximal allowed intake level (0.26 ng /kg b.w./day) (Table S1). For nursing infants, however, the total median intakes of BDE-99 (455 and 488 pg/kg b.w./day for male and female, respectively) higher than the maximal allowed intake level with a factor ∼2, indicating possible health risk for Chinese nursing infants with exposure to PBDEs.

Although the toxicity and pharmacokinetic data of PBDE congeners to humans are limited, previous studies have observed an exponential increase of the PBDEs levels in environmental media and human samples over the last 30 years (*10*, *45*, *3*). In China, Mai et al. (*4*) examined the distribution of PBDEs in sediments of the Pearl River Delta and adjacent South China Sea and found that BDE-209 concentrations were at the high end of the worldwide figures. Chen et al. (*6*) also found that the arithmetic mean atmospheric concentrations of BDE-209 were higher than those in North America and Europe. Because BDE-209 could be degraded into lower brominated congeners with higher bioavailability and toxicity (*35*−*36*) and PBDE-containing products are still used in large quantities in China, more studies are needed to fully understand the fate and impact of PBDEs.

Acknowledgments

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Supporting Information Available

Two tables show the estimated daily intake of PBDEs and the median level of PBDEs in fish. A figure shows a map of the study area and sampling sites. This material is available free of charge via the Internet at http://pubs.acs.org.

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