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Polybrominated Diphenyl Ethers in Foodstuffs from Taiwan: Level and Human Dietary Exposure Assessment

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) may contaminate food through bioconcentration and biomagnification. PBDEs often exist in the food chain and are consumed by humans. This study aims to determine the concentrations of PBDEs in food intake and to estimate the daily exposure of Taiwanese citizens to PBDEs. One hundred and eight food samples from nine types of commonly consumed foodstuffs were collected from northern, central, southern, and eastern regions of Taiwan. The samples were analyzed for PBDE level by gas chromatography mass spectrometry. Also, a daily dietary intake survey was conducted of 466 adults (153 men, 313 women) in these four regions of Taiwan. Taiwanese daily dietary intake of PBDE is calculated by means of food PBDEs level and daily dietary intake. The result of this study showed the highest concentration of ΣPBDE was found in butter (890.3 ± 309.0 pg/g wet weight), followed by egg and pork (553.0 ± 185.0 pg/g wet weight and 545.4 ± 181.0 pg/g wet weight). Deca-BDE was found the highest concentration among eight kinds PBDEs. The average daily intake of PBDEs for the 466 subjects was 67.95 ± 23.01 ng/day. There was a significant difference between the daily intake of ΣPBDE in different regions of Taiwan ($p<0.05$). The highest daily intake of ΣPBDE was in northern Taiwan, which is also the most urbanized area.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of structures with halogen-containing diphenyl ethers, with 209 types of congeners based on the different number of bromine atoms. Commercial PBDE products were widely used as brominated flame in plastics, foam, textiles, furniture, building materials, auto parts or electronics. Among them, penta-BDE, octa-BDE, and deca-BDE were the most commonly used. PBDEs could enter the environment during use, production, disposal, and recycling (Naert et al., 2007).

Fat-solublility and slow degradation of PBDEs often resulted in a lasting presence in the environment (Peng et al., 2007; Xiang et al., 2007); the less brominated substances had a longer half-life. PBDE content showed a rising trend year by year in sediments tested (Song et al., 2005; Li et al., 2006). Owing to PBDEs characteristic of bioconcentration and biomagnification, they often existed in the food chain, especially for BDE-209 congener (Kelly et al., 2007; Voorspoels et al., 2007a; Van den Steen et al., 2007). The half-life of PBDEs in the human body is several weeks or even months (Thuresson et al., 2006). In 2003, the European Parliament and Council banned the use of hazardous substances composed of PBDEs in electrical and electronic equipment (EUC, 2003). In 2009, parts of PBDEs were listed as persistent organic pollutants at the Stockholm Convention by the United Nations Environment Programme (Persistent Organic Pollutants Review Committee, POPRC, 2009).

As the structure of PBDEs is similar to thyroid hormones thyroxine and triiodothyronine, they interfere with thyroid secretion; animal exposed to PBDEs would have thyroid disruption with lower and free thyroxine concentrations in serum and neurodevelopmental toxicity in pubertal stage (Hallgren and Darnerud, 2002; Zhou et al., 2002; Stoker et al., 2004). It should be noted that exposure to one of the most persistent PBDE congeners, BDE-99, could cause renal and liver impairment in animal model (Huwe et al., 2008; Albina et al., 2010). Quite significantly, PBDEs have been found in the existence of human tissues of liver, breast, blood, breast milk, and placenta (Johnson-Restrepo et al., 2005; Frederiksen et al., 2009).

Humans were probably exposed PBDEs through the ingestion of food (Schecter et al., 2006; Guo et al., 2007), air inhalation (Wilford et al., 2004), and dust (Betts, 2008). The bioaccessible contaminant

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of PBDEs was mostly absorbed by the human gastrointestinal tract (Tao et al., 2009; Xing et al., 2008). Due to the fact that foods provided a much greater contribution of PBDE in humans (Bakker et al., 2008; Domingo et al., 2008), the contamination of food from PBDEs had received a great deal of attention (Schecter et al., 2010; Voorspoels et al., 2007b; Yu et al., 2011); therefore, food intake should be more closely mentioned so as to prevent the exposure of humans to PBDEs.

PBDE contamination in food usually came from external sources, such as environment exposures or food additives. As PBDE contamination impaired human health, a careful study on this subject was necessary. We needed to know what degree our food was contaminated with PBDE, and how much of our daily food intake was contaminated. This study analyzed the concentrations of PBDEs in daily food intake and the estimates of PBDEs in total daily intake to determine exposure to PBDE. The findings of the study would be useful for evaluating current policy regarding the control and management of PBDE usage.

2. Methodology

2.1. Foodstuff samples and targeted compounds

This study sampled nine types of food regularly consumed by Taiwanese. Milkfish, salmon, chicken, beef, pork, butter, egg, rice and milk were selected for sampling. Each kind of food was randomly selected as three replicates for a total of 108 food collected from four regions of Taiwan. Foods were purchased in Dec. 2009 at supermarkets or general markets in northern, central, southern and eastern Taiwan. Samples were refrigerated in their original packaging at −20 °C and analyzed within a week after purchase. Most samples were analyzed by using the whole food, except the salmon, beef and pork, which were analyzed by using parts normally consumed.

The eight PBDE-congeners were chosen for analysis including BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, and BDE-209, based on their profusion and toxicity (de wit, 2002; Voorspoels, et al., 2007b), which were frequently contained in commercial products in daily life. Panel on Contaminants in the Food Chain also suggested these eight PBDE congeners would be primaries on PBDEs in food contamination (EFSA, 2011).

2.2. Method of analysis

The method of analysis was adopted by the reference method for polybrominated diphenyl ethers determination of the Taiwan Environment Protection Agency, NIEA M802.00B method (Taiwan EPA, 2007). For the eight PBDE analysis, the standard reference materials (SRM) were purchased from AccStandard Company, USA, BDE-CSM and BDE-EPA-SET, BDE in isooctane. ^{13}C labeled BDE-209 were obtained from Wellington Laboratories (Andover, MA, USA), and used as internal standard.

2.2.1. Foodstuff sample pretreatment

2.2.1.1. Remove water for food sample. Food samples including milkfish, salmon, chicken, beef, pork, butter, egg and rice were placed in a −20 °C refrigerator for at least 24 h. The next day, the samples were lyophilized in a lyophilizer to remove water. The water content in food was determined by gravimetric method. Milk was put into a microwave and heated with 720 W, 2450 MHz for 4 min twice to dry and remove water.

2.2.1.2. Lipid extract for sample and lipid weight determination. For the lipid extraction, 15 g of homogenized dried sample were ground to powder, and extracted with 200 mL acetone/n-hexane (2:1 v/v) for 8 min with stirred and filtration, repeat this procedure again with 200 mL acetone/hexane solution, afterword, the extract was placed in a 60 °C water bath to decompress and concentrate to dry, and the content of lipid that performed on an aliquot of the exact, could be determined by weight method.

2.2.1.3. Remove lipid from the extract and clean up. In order to remove the lipid from 2. 2. 1. 2 treated extract, the extract was dissolved with 100 mL of n-hexane, and slowly adding 30 g of acidic silica gel (silica gel/ sulfuric acid = 60/40) and stirred for 10 min to collect the hexane filtrate, then another 30 mL hexane was added and stirred 10 min. The two hexane filtrates were combined, and concentrated by a vacuum concentration device, and then make up to 15 mL with n-hexane. The extract was further cleaned up by acidic alumina tube, the extract was put into the tube, and then washed by 15 mL of n-hexane and discharged the eluate. The tube was then washed by 40 mL dichloromethane/ n-hexane (50/50, v/ v), and the eluate was collected, and then make up to 100 μL by dichloromethane for GC/MS analysis.

2.2.2. Gas chromatography mass spectrometry (GC/MS) analysis

Eight PBDEs were analyzed by GC/MS. GC is of Trace GC ultra II and MS is of DSQ (Thermo Fisher Scientific, Waltham, MA, USA). The parameters were set as the following: column: J&WDB-5HT capillary column,15 m × 0.25 mm ID × 0.1 μm (Agilent, Wilmington, DE, USA); injection volume: 1 μL; splitless mode; carrier gas: He 99.999%; carrier gas volume:1.0 mL/min; temperature program: started at 120 °C, hold for 1 min, then increased at 25 °C/min to 330 °C, hold for 10 min; ion source: 280 °C; transfer line: 310 °C; ionization: electron impact/selected ion monitoring mode; mass: BDE-28: m/z 406, 408, BDE-47: m/z 484/486, BDE-99/100: m/z 564/566, BDE-153, 154: m/z 642/644, BDE-183: m/z 721/723, BDE-209: m/z 957/959.

2.3. Quality Control: Calibration curves, recovery, precision and method detection limits

A daily check of calibration curves were conducted. Regular standard solution including BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209 were checked by one in each batch of 10 samples. Calibration curves of concentration range of BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 were from 10 to 2000 ng/L, and BDE-209 was from 100 to 2000 ng/L. The correlation coefficients of each analyzer were above the 0.994.

Spike analysis was conducted, adding 100 μL of 0.02 μg/mL, 0.20 μg/mL and 0.50 μg/mL standard solution of PBDEs (SRM) into the food samples and carrying out the same extraction procedure of food samples to calculate the recoveries of the pretreatment and analysis methods. Recoveries of BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209 of samples were 87.19 ± 25.42%, 96.69 ± 19.71%, 96.79 ± 23.13%, 93.59 ± 19.92%, 83.71 ± 18.71%, 82.47 ± 14.95%, 106.31 ± 28.28%, 130.91 ± 27.86%, respectively.

The precision of the eight PBEs were conducted, and CV % of eight PBDEs were ranged from 0.88% to13.90% (as shown in Table 1).

A blank sample was conducted to avoid background contamination in the laboratory by a daily check. We found BDE-209 congener in blank solution significantly, the level of BDE-209 was ranged 18 ~ 55 ng/mL blank, but relative steady; the concentration was around 20 ~ 28 ng/mL, and mean ± SD was 28.78 ± 12.28 ng/mL. We analyzed the blank solutions each day, and 14 blank solutions were analyzed in the experiment. This procedural blank value was used for subtraction, after blank subtraction, the limit of detection was set at three times of the standard deviation of the blank. Method detection limit (MDL) of each PBDE was shown in Table 1.

2.4. Food intake survey and estimation of daily food intake of PBDEs

For estimates of the daily intake of PBDEs in a typical Taiwanese, a food intake survey was conducted in northern, central and southern

Table 1
PBDEs analysis of the quality assurance and quality control.

Name of PBDEs	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209
Retention time (min)	4.82	5.65	6.25	6.42	6.89	7.11	7.77	10.3
R	0.998	0.999	0.999	0.998	0.998	0.998	0.999	0.994
Recovery (%)	87.19 ± 25.42	96.69 ± 19.71	96.79 ± 23.13	93.59 ± 19.92	83.71 ± 18.71	82.47 ± 14.95	106.31 ± 28.28	130.91 ± 27.86
CV (%)	7.93	6.11	0.88	9.78	7.10	10.52	10.95	13.39
MDL (pg/g dry weight)[a]	5.6	8.0	1.5	6.0	2.6	6.8	1.4	102.0
MDL for milk (pg/mL)	0.9	1.3	0.3	1.0	0.4	1.1	0.2	17.0

MDL: method detection limit.
[a] Milk samples excluded.

Taiwan. Participants included 466 persons (153 men, 313 women) between 35 and 60 years of age, who were randomly selected in each region. Participants would be interviewed face-to-face by an interviewer asking amount and frequency of milkfish, salmon, chicken, beef, pork, butter, egg, rice and milk they consume everyday. We calculated the quantity these foods taken each day, then multiplied with the level of PBDE in each food. The formula was as follows: daily intake of PBDE (μg/day) = Σ (daily quantity of each kind of food consumed by participants (g/day) × PBDE levels of each foodstuff (ng/g)/1000.

2.5. Statistical Analysis

All statistics were calculated using the software SPSS 17.0. ANOVA was applied to test Taiwan four area daily PBDEs intake differences. The daily PBDEs intake was compared between male and female with student *T* test.

3. Results

3.1. PBDE concentrations in food samples

Nine types of typical food consumed for people including 108 food samples were analyzed. ΣPBDE concentrations in each food sample on the basis of wet weight are shown in Table 2. Out of all the food samples, butter has the highest ΣPBDE concentrations, 890.3 ± 309.0 pg/g wet weight, followed by egg and pork, 553.0 ± 185.0 pg/g wet weight and 545.4 ± 181.0 pg/g wet weight respectively. We found that some foods in different region have distinct PBDE congeners; for milkfish, BDE-47 had the highest concentration in northern Taiwan, and BDE-153 had the highest concentration in central, southern, and eastern Taiwan. For milk, BDE-209 had the highest concentration in central Taiwan. However, some foods had the same PBDE congener which existed in whole area of Taiwan. For salmon, BDE-47 had the highest concentration in the whole Taiwan. For chicken, pork, beef, and milk, BDE-209 was detected to have the highest PBDE congener concentration in whole areas of Taiwan. The lipid contents of food samples with PBDE congeners are also shown in Table 2. Out of all the food samples, chicken had the highest PBDE concentration, 13017.9 ± 4072.0 pg/g lipid, followed by pork, 11180.0 ± 3775.0 pg/g lipid.

3.2. Daily intake of PBDEs in Taiwanese people

The daily average PBDE intake from food was estimated to be 67.95 ± 23.01 ng/day in the whole of Taiwan, and it was estimated to be 95.43 ± 16.58 ng/day, 41.02 ± 7.23 ng/day, 56.40 ± 8.18 ng/day and 77.75 ± 9.54 ng/day in northern, central, southern, and eastern Taiwan respectively ($F = 28.8$, $p < 0.05$). The daily PBDEs intake in males was significantly higher than in females ($F = 8.2$, $p < 0.05$). Pork was the greatest source of daily PBDEs intake in Taiwan residents, followed by milk and egg (Table 3).

4. Discussion

PBDEs were widely used in various Taiwan industries. In this study, PBDE congeners detected in food samples from Taiwan was similar to other countries (Schecter et al., 2010; Voorspoels et al., 2007b; Yu et al., 2011). Through the estimate of daily intake of PBDEs in Taiwanese, we knew residents living in different regions of Taiwan were exposed to varying degrees of PBDE pollution and the

Table 2
Concentrations of PBDE congeners in food samples in Taiwan determined in wet weight and lipid weight (n = 108).

	n	ΣPBDE	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209
		Unit: pg/g Wet Weight								
Milkfish	12	260.0 ± 60.0	ND[a] (1.8)	36.0 ± 34.2[b,c]	6.3 ± 13.0	5.7 ± 17.0	26.0 ± 46.0	5.0 ± 8.1	2.7 ± 5.6	117.4 ± 337.6
Salmon	12	503.7 ± 65.0	45.0 ± 87.0	170.6 ± 147.3	35.0 ± 48.0	63.0 ± 31.4	30.0 ± 26.5	11.8 ± 20.0	3.3 ± 10.0	145.0 ± 360.0
Chicken	12	191.2 ± 60.0	2.1 ± 7.2	3.6 ± 7.1	ND (0.32)	2.3 ± 7.0	ND (0.6)	7.3 ± 12.4	4.4 ± 9.0	171.0 ± 248.0
Beef	12	196.3 ± 54.0	ND (1.7)	1.4 ± 2.1	4.0 ± 1.2	ND (1.8)	31.4 ± 26.0	ND (2.0)	1.1 ± 3.2	156.0 ± 250.0
Pork	12	545.4 ± 181.0	ND (1.7)	6.3 ± 5.0	ND (0.4)	19.4 ± 39.1	ND (0.7)	ND (1.9)	1.2 ± 2.3	516.0 ± 512.0
Butter	12	890.3 ± 309.0	1.2 ± 1.1	5.4 ± 3.0	ND (1.3)	2.3 ± 4.0	ND (2.2)	4.0 ± 10.4	ND (1.2)	875.0 ± 610.0
Egg	12	553.0 ± 185.0	ND (3.0)	6.4 ± 6.2	1.2 ± 2.2	ND (3.0)	ND (1.3)	ND (3.4)	15.0 ± 22.3	525.0 ± 492.0
Rice	12	ND	ND (<0.5)	ND (<0.5)	ND (<0.5)	ND (<0.5)	ND (<0.5)	ND (<0.5)	ND (<0.5)	ND (<0.5)
Milk	12	137.5 ± 39.0	2.2 ± 5.3	4.3 ± 8.0	4.5 ± 10.0	7.0 ± 1.3	2.4 ± 6.0	ND (6.8)	ND (1.4)	113.0 ± 47.0
		Unit: pg/g Lipid Weight								
Milkfish	12	3824.3 ± 676.0	ND (26.5)	625.0 ± 543.0	104.0 ± 250.0	170.0 ± 565.0	772.0 ± 1575.0	110.0 ± 235.0	36.0 ± 100.0	1994.0 ± 3624.0
Salmon	12	4145.8 ± 645.0	390.0 ± 782.0	1823.0 ± 1288.0	313.0 ± 410.0	178.0 ± 271.0	122.0 ± 220.0	93.4 ± 174.0	29.4 ± 103.3	1197.0 ± 2843.0
Chicken	12	13017.9 ± 4072.0	153.0 ± 523.0	216.0 ± 524.0	ND (21.7)	96.2 ± 31.2	ND (37.6)	564.0 ± 1108.0	267.0 ± 662.0	11692.0 ± 12325.0
Beef	12	6513.4 ± 2002.0	ND (58.4)	62.3 ± 167.0	62.1 ± 167.0	ND (58.4)	539.0 ± 1868.0	ND (66.2)	24.5 ± 60.2	5734.0 ± 11450.0
Pork	12	11180.0 ± 3775.0	ND (34.3)	88.5 ± 81.3	ND (8.6)	291.0 ± 541.0	ND (14.8)	ND (38.8)	21.2 ± 48.0	10731.0 ± 11435.0
Butter	12	2455.0 ± 850.0	3.4 ± 8.0	19.7 ± 13.4	ND (1.8)	6.2 ± 12.0	ND (2.8)	13.6 ± 24.3	ND (1.5)	2409.0 ± 2407.0
Egg	12	8607.0 ± 2894.0	ND (46.5)	88.2 ± 95.1	6.7 ± 21.3	ND (46.5)	ND (20.2)	ND (52.7)	205.0 ± 337.0	8224.0 ± 7921.0
Rice	12	ND	ND	ND	ND	ND	ND	ND	ND	ND

Calculated assuming that non-detected congener concentrations are equal to half of the limit of detection.
[a] ND: not detected, numbers in parenthese denote limit of detection (LOD).
[b] Concentrations: mean ± standard deviation.
[c] Calculated by concentration on dry weight base in individual foodstuff for wet and lipid weight.

Table 3
Concentrations (mean ± standard deviation) of daily PBDEs intake for Taiwan adult residents (ng/day).

	Northern Taiwan			Central Taiwan			Southern Taiwan			Eastern Taiwan		
	Total (n = 213)	Male (n = 91)	Female (n = 122)	Total (n = 56)	Male (n = 14)	Female (n = 42)	Total (n = 98)	Male (n = 26)	Female (n = 72)	Total (n = 99)	Male (n = 22)	Female (n = 77)
Total [a]	95.5 ± 16.6	112.3 ± 20.4	82.9 ± 13.7	41.0 ± 7.2	39.8 ± 5.5	41.4 ± 7.8	56.5 ± 8.2	62.5 ± 9.7	54.3 ± 7.6	77.8 ± 9.5	114.0 ± 13.9	67.4 ± 8.3
Fresh fish	0.3 ± 0.5	0.3 ± 0.5	0.3 ± 0.5	1.3 ± 1.3	1.0 ± 1.0	1.4 ± 1.3	2.5 ± 1.0	2.3 ± 0.9	2.5 ± 1.0	8.8 ± 2.9	13.9 ± 4.6	7.4 ± 2.5
Salt fish	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	3.1 ± 1.8	5.3 ± 3.2	2.3 ± 1.4	4.6 ± 4.8	2.8 ± 2.9	5.2 ± 5.5	22.6 ± 11.3	18.3 ± 9.2	23.8 ± 11.9
Chicken	10.8 ± 11.5	11.2 ± 11.9	10.5 ± 11.2	1.1 ± 0.6	1.2 ± 0.6	1.1 ± 0.6	3.6 ± 5.8	3.6 ± 5.8	3.6 ± 5.8	3.0 ± 0.6	4.0 ± 0.8	2.7 ± 0.6
Beef	ND	ND	ND	1.8 ± 1.0	2.2 ± 1.2	1.6 ± 0.9	0.9 ± 1.5	1.8 ± 3.2	0.6 ± 1.0	2.8 ± 2.5	5.3 ± 4.6	2.1 ± 1.9
Pork	30.1 ± 21.7	33.9 ± 24.5	27.2 ± 19.6	5.6 ± 9.2	6.0 ± 9.7	5.5 ± 9.0	24.6 ± 28.0	28.5 ± 32.3	23.2 ± 26.3	19.8 ± 11.0	39.0 ± 21.8	14.3 ± 8.0
Egg	42.2 ± 30.8	53.5 ± 39.0	33.8 ± 24.6	6.8 ± 7.1	7.3 ± 7.0	6.6 ± 6.9	9.7 ± 5.0	12.2 ± 8.8	8.8 ± 6.4	2.5 ± 0.5	3.4 ± 0.7	2.3 ± 0.4
Rice	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Milk	12.0 ± 6.5	13.3 ± 7.2	11.0 ± 6.0	21.5 ± 4.9	16.8 ± 3.9	23.0 ± 5.3	10.7 ± 0.0	11.3 ± 0.0	10.5 ± 0.0	18.2 ± 3.3	30.1 ± 5.4	14.8 ± 2.7

ND: not dectected.
[a] Tested difference by one-way ANOVA, $p<0.05$.

highest daily intake of ΣPBDE was in northern Taiwan, which was the most urbanized area.

4.1. PBDEs in food samples

Out of a pool of eight kinds of food contaminated with PBDEs, BDE-209 congener was found in the highest concentration. BDE-209 congener was a major component of flame retardants in Taiwan, which were commonly used on furniture, cabinets made of plastic resin, and other utilities. BDE-209 congener was also used as a polyester fiber additive, in automobile fabrics and in mixtures added to rubber. BDE-209 congener seriously damaged the environment and human health because of its characteristics of accumulation, tissue-specific distribution and debromination to different congeners in exposed organisms (Kierkegaard et al., 1999; Stapleton et al., 2004; Van den Steen et al., 2007), and for its potential toxicity (Darnerud, 2003). In Taiwan, BDE-209 congener had been banned for commercial usage by the government; however, it still contaminated the Taiwan environment. According to the Panel on Contaminants in the Food Chain, the highest dietary contaminants are BDE-47 and BDE-209 (EFSA, 2011). This study showed similar results. Peak concentrations of BDE-209 were observed in chicken, pork, beef, and milk. Highest levels of BDE-47 were observed in salmon. In addition, BDE-47 and BDE-153 were higher in milkfish, which was similar to the results of other studies (Geyer et al., 2004; Trudel et al., 2011). Low bromine number PBDE congeners with a longer half-life could be easily absorbed, especially by fish (Bocio et al., 2003). In this study, the average concentration of PBDEs in milkfish was 0.625 ± 0.543 ng/ g lipid, while in salmon, it was as high as 1.823 ± 1.288 ng/ g lipid. However, salmon was not a Taiwan domestic cultured fish, and instead was mainly imported from the Atlantic coast.

4.2. Human exposure to PBDEs

In order to understand the PBDE pollution situation in Taiwan, we compared the PBDE concentrations in the neighborhood countries and areas of Japan, China and Taiwan. In Osaka, Japan, PBDE concentrations ranged between 0.02 and 1.65 ng/ g fresh weight in fish. Out of all the fish, the domestic cultured fish, young yellowtail, had an average concentration of PBDEs of 1.65 ng/ g wet weight, which had the highest PBDE concentration. Other than milkfish, the average pork concentration of PBDEs was 0.68 ng/g wet weight, which had the highest PBDE concentration among meat (Ohta et al., 2002). In Shanghai, China, PBDE concentrations ranged between 3.3 and 679.1 ng/ g wet weight in fish; of all fish, the river fish, large yellow croaker, had an average concentration of PBDEs of 515.7 ng/ g wet weight, which was the highest PBDE concentration. The average concentration of PBDEs in pork was 32.2 ng/g, and the average concentration of PBDEs in duck was 61.9 ng/g wet weight, which was the highest PBDE concentration among meat (Yu et al., 2011). In Taiwan, PBDE concentrations ranged between 260 and 443 pg/ g wet weight in fish; the domestic cultured fish, milkfish had an average concentration of PBDEs of 260 pg/ g wet weight. Taiwan had PBDE contamination similar to Japan but much less contamination than China. However, Shanghai, China, which was a bustling metropolitan area with industrial factories, PBDE pollution was a constant and a serious threat.

From the survey results of the PBDE estimate in the daily intake of Taiwan residents, we have found that people who lived in northern Taiwan had the highest levels of PBDE. Due to the fact that northern Taiwan was composed of mostly metropolitan and industrial areas, similar to Shanghai, China, food exposure to PBDEs in this area was more serious than that of other areas in Taiwan. We suspected that the contamination of the food chain by PBDEs was also highly connected with air pollution in the area. The route of air pollution and its role in contamination of the food chain by PBDEs will be a research topic of the near future. The estimates of PBDEs in the daily intake of Taiwanese was medium high in comparison to other countries in recent studies, e.g., 23–48 ng/day in Belgium (Voorspoels et al., 2007b), 34.2-66.2 ng/day in China (Miyake et al., 2008), 21–46 ng/day in Japan (Akutsu et al., 2008), 75 ng/day in Spain (Domingo and Bocio, 2007), 68 ng/day in China (Luo et al., 2009), 40 ng/day in Romania (Dirtu and Covaci, 2010), 50 ng/day in America (Schecter et al., 2010), 72 ng/day for men and 83 ng/day for women in Australia (Shanmuganathan et al., 2011), and 49 ng/day in Sweden (Törnkvist et al., 2011) (Table 4). Since Taiwan is an island nation, the chance to study PBDE contamination there was unique because the contamination was not influenced by other countries. The most serious PBDE contamination issue was in domestic meats such as pork and chicken. Previously, particularly in nations with a developing economy, PBDE's were not rigorously regulated. Although regulations are stricter now, industry is still producing enough PBDEs to pollute our environment.

The daily intake of PBDEs differs was due to the difference of dietary habits and different types of food in distinct areas and countries (Domingo, 2012). The critical issue was on how to reduce PBDE contamination in food. PBDE contamination in food in Taiwan was still

Table 4
Comparison of estimated dietary intake of PBDEs in Taiwan and other countries.

Country	Detected foods	PBDE intake (ng/day)	References
Belgium	Fish, meat, sea food, dairy, egg	23–48	Voorspoels et al., 2007a, 2007b
China	Fish, sea food	34–66	Miyake et al., 2008
Japan	Fish, meat, dairy, egg, vegetable based food, fat/oil	21–46	Akutsu et al., 2008
Spain	Fish, meat, dairy, egg, vegetable based food, fat/oil, bakery	75	Domingo et al., 2008
China	Chicken, duck	68	Luo et al., 2009
Romania	Meat, dairy, egg, oil	40	Dirtu and Covaci, 2010
America	Fish, meat, dairy, egg, vegetable based food	50	Schecter et al., 2010
Australia	Fish, sea food	72 (men), 83 (women)	Shanmuganathan et al., 2011
Sweden	Fish, meat, dairy, egg, fat/oil	49	Törnkvist et al., 2011
Taiwan	Fish, meat, dairy, egg, rice	68	This study

high. Evidently, regulations on the restriction of PBDE use must be strictly enforced by the Taiwanese authorities.

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