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Indoor Level of Polybrominated Diphenyl Ethers in the Home Environment and Assessment of Human Health Risks

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ABSTRACT

It has been demonstrated that human exposure to polybrominated diphenyl ethers (PBDEs) might be associated with several adverse health effects. Dietary and microenvironmental sources are considered to be the main routes of PBDEs exposure. The study aimed to investigate PBDEs in residential indoor and outdoor air and further to assess the health risks in family members of different ages. Indoor and outdoor air samples from houses in residential areas were simultaneously collected for analysis of BDE-47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, and 209 by high-resolution gas chromatography/high-resolution mass spectrometry. PBDE concentrations were non-significantly higher indoors (81.1 pg/m^3) than outdoors (42.7 pg/m^3) (p = 0.513). For the outdoor air, the mean PBDE level was lower in air outside houses than in air from industrial and urban areas. Levels of Σ_{14}PBDEs and BDE-209 in house indoor air were no higher in Taiwan than other countries. The daily intake of non-dietary PBDEs from house air and dust in Taiwan was highest in the toddlers (1–2 years old; 8.22 ng/kg b.w./day) and lowest in the male adults (≥ 20 years old; 0.562 ng/kg b.w./day) among family members. For Taiwanese, the risks of non-cancer (hazard quotient: HQ) and cancer (cancer risk: R) with neurobehavioral effects of exposure to non-dietary PBDEs in the home environment were assessed to be lower than the critical values of 1.00 and 1.00×10^{-6} for HQs and Rs, respectively. In conclusion, levels of indoor PBDEs and non-dietary daily intake were found to be low in home environments in Taiwan. This result suggests that PBDEs in the home environment are not harmful to family members from the newborn to the elderly if we only consider the neurobehavioral effects.

Keywords: Polybrominated diphenyl ethers; Indoor air; House; Daily intake; Health risk.

INTRODUCTION

Staying indoors is associated with numerous negative effects and one of them is exposure to hazardous toxic substances in indoor air including polybrominated diphenyl ethers (PBDEs). PBDEs are a class of brominated fire retardants (BFRs) known to disrupt endocrine hormone functions. PBDE commercial formulations, namely PentaBDEs, OctaBDEs, and DecaBDE, have been widely utilized as BFRs in consumer products such as consumer electronics (i.e., television sets), textiles, carpets, building materials, and upholstered furniture during the last two decades. Toxicological data was achieved on the US EPA Integrated Risk Information System (IRIS) showing that reference doses for chronic oral exposure (RfDs) on neurobehavioral effects were 0.0001 (Benchmark Dose level (BMDL): 0.35 mg/kg), 0.0001 (BMDL: 0.29 mg/kg), 0.0002 (no-observe-adverse-effect level (NOAEL): 0.45 mg/kg) and 0.007 mg/kg/day (NOAEL: 2.22 mg/kg) in BDE-47, 99, 153, and 209, respectively (US EPA, 2008). PBDEs have continuously raised the public's and public officials' concern because of their ubiquity in the microenvironment and their association with several adverse health effects, including

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disruption of thyroid and growth hormone homeostasis, neurodevelopment, menstruation, pregnancy, and semen quality (Chao *et al.*, 2007; Chao *et al.*, 2010; Lin *et al.*, 2011; Shy *et al.*, 2012; Chao *et al.*, 2014a; Hsu *et al.*, 2014). Air levels of PBDEs are often lower outdoors than indoors (Hazrati and Harrad, 2006). According to the Swedish report, many BFRs were frequently found in indoor, ventilation, and outdoor air samples and PBDE levels increasing as outdoor air samples were obtained in more urban areas than rural areas (Newton *et al.*, 2015). In general, the higher PBDE concentrations in indoor air than outdoor air have been attributed to their extensive use in consumer products in the indoor environment (Wilford *et al.*, 2004; Besis and Samara, 2012) and higher PBDE levels of gaseous and particulate indoors than outdoors have been attributed to the abundance of indoor sources of PBDEs emission, smaller space, restricted air exchange rates, and weaker air circulation systems. PBDEs are dispersed through the ventilation system or by natural convection (Besis and Samara, 2012; Björklund *et al.*, 2012; Zhang *et al.*, 2011). Although commercial products containing pentaBDEs and octaBDEs mixtures have been phased out in most countries since 2005, and decaBDE mixtures are only banned by the European Union, PBDEs are continuously released from existing consumer products into the indoor environment and will be for many years to come (Frederiksen *et al.*, 2009). Different indoor environments have different airborne PBDE concentrations and different PBDEs emission characteristics. Several studies reported diverse air PBDE concentrations inside the passenger compartments of automobiles (Mandalakis *et al.*, 2008; Thuresson *et al.*, 2012), in-flight airplane cabins (Allen *et al.*, 2013), homes and offices (Zhang *et al.*, 2011), houses, offices, apartments, day care centers (Thuresson *et al.*, 2012), and e-waste recycling plants (Guo *et al.*, 2015).

Health risk studies (Jones-Otazo *et al.*, 2005; Toms *et al.*, 2009; de Wit *et al.*, 2012; Hearn *et al.*, 2012) have found that human exposure to PBDEs is mainly from ingesting or inhaling indoor dust. Several reports have revealed a correlation between PBDEs adhering to house dust and PBDEs contaminating breast milk, maternal and cord blood, and hair (Wu *et al.*, 2007; Frederiksen *et al.*, 2010; Kang *et al.*, 2011). A Chinese study indicated that indoor PBDEs are released from the surface of consumer products into the air and accumulate in dust and air-conditioner filters (Ni *et al.*, 2011). Although current studies show that body PBDEs concentrations are not related to indoor air PBDE concentrations (possibly because of small sample size; Fromme *et al.*, 2009; Toms *et al.*, 2009), a pattern of non-dietary PBDEs exposure probably exists and the possible route of PBDE exposure is release from the surface of electronics and transfer to human bodies through accumulation of airborne PBDEs on indoor dust.

In Taiwan, PBDE levels have been investigated in urban, suburban, industrial, and rural ambient air, house dust in residences, and the oceanic atmosphere near the island of Formosa (Wang *et al.*, 2011; Lin *et al.*, 2012; Chao *et al.*, 2014b, c). A few environmental studies have focused on measurement of indoor PBDEs levels and the impact of PBDEs on human health in Taiwan. Our goal was to determine levels of PBDEs in the indoor and outdoor air of residences located in southern Taiwan. We used PBDE concentrations in house air in the present study and in house dust in our previous study (Chao *et al.*, 2014b) to assess the non-dietary exposure of the adults, adolescents, children, and toddlers to PBDEs via indoor air inhalation and dust ingestion.

METHODS

Reagents and Chemicals

The standard solutions of the 14 PBDE congeners (BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208 and 209) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA) and the 8 ^{13}C-labeled PBDEs (BDE-28, 47, 99, 153, 183, 197, 207, and 209) were obtained from Wellington Laboratories (Guelph, Canada). Sodium sulfate, alumina oxide, potassium oxalate, and silica gel of the highest grade were from Merck (Darmstadt, Germany).

Samples Collection, Extraction, and Cleanup

Samples in the present study were obtained from residential areas in southern Taiwan between October 2012 and March 2013. One was in Kaohsiung City and two were in Pingtung County. Indoor air in the houses and ambient air outside the houses were simultaneously sampled by two high-volume air samplers and, following US EPA Reference Method TO9A, each sample was collected for approximately 40 h (1–2 days, ~600 m^3) using model PS-1 air samplers (Graseby Andersen Inc., Smyrna, GA) each equipped with a quartz fiber filter followed by a glass cartridge containing polyurethane foam (PUF) to capture the particulates and gas in each sample. The quartz fiber filter and PUF of each air sample were combined before the chemical analysis of the 14 PBDEs.

The sampling, extraction, and cleanup procedures for air samples of PBDEs used in previous studies were followed with minor modification (Wang *et al.*, 2011; Chao *et al.*, 2014c). In brief, before the air sampling, pre-labelled isotopes and identifiable surrogate standards were spiked in the cartridges to evaluate the collection efficiency of the sampling process. After air samples were collected, internal standards were spiked into the samples, well mixed with toluene, and then extracted with toluene for 24 h in a Soxhlet extractor to monitor the extraction and cleanup process. The extracts were concentrated, treated with concentrated sulfuric acid, and passed through a multicolumn system of acid silica, alumina, and activated carbon columns. The eluate was collected, concentrated to near dryness using a nitrogen stream, and then transferred to a vial. A total of 50 μL of ^{13}C-labeled BDE-139 was added to each eluate as an internal recovery standard after the clean-up and prior to injection to minimize the possibility of loss. The final extract was reduced in volume to 0.2 mL under a stream of nitrogen.

PBDEs Analysis

The sample extracts were analyzed by high-resolution gas chromatography/high-resolution mass spectrometry

(Hewlett-Packard 6970 Series gas/Micromass Autospec Ultima) using a positive electron impact (EI+) source in the selected ion monitoring (SIM) mode with a resolving power of 10,000. The GC analysis of PBDEs was performed on a DB-5HT column (L = 15 m, i.d. = 0.25 mm, film thickness = 0.1 μm) (J&W Scientific, Folsom, CA) in splitless mode at 280°C with constant helium flow of 1 mL/min. The GC temperature program consisted of 100°C for 4 min, increase to 200°C at a rate of 40 °C/min between 100 and 200°C, 200°C for 3.5 min, increase to 325°C at a rate of 10 °C/min, and 325°C for 2.5 min in the final step.

The electron energy and source temperature were specified at 35 eV and 250°C, respectively. The two most abundant isotope masses were measured for each component. Quantification was performed using internal/external standard mixtures via the isotope-dilution method. The US EPA Method 1614A of analytical quality assurance and quality control (QA/QC) was followed. Prior to air sampling, PUF cartridges were spiked with PBDE surrogate standards pre-labeled with isotopes to obtain the recoveries of PBDEs surrogate standards within 82–121% of acceptable QA/QC limits (i.e., 70–130%). The limits of detection (LODs) and quantification (LOQs) were defined as the amount at which the signal-to-noise (S/N) ratios were higher than 3 and 10, respectively. The LODs for the 13 PBDE congeners (BDE-28 to -208) ranged from 0.288 to 49.0 pg/g and the LOD of BDE-209 was 314 pg/g. The analysis of the PBDE labeled internal, precision and recovery (PAR), and surrogate standards all met the relevant standards. Laboratory blanks were analyzed for each batch of 10–12 samples. The total amounts of PBDEs in the field and laboratory blanks were extremely low (mostly negligible) compared with those of the real samples. The isotopic ratios of at least two characteristic ions for each sample were consistent with theoretical values to within a deviation of 15%. Calibration mixtures with isotopically labeled internal standards were tested in the quantification of the target compounds.

Health Assessments

Non-dietary health assessments of PBDEs in Taiwanese houses including daily intake and neurobehavioral effects of non-cancer risk (Hazard quotient: HQ), and cancer risk (neurobehavioral effect) (R) assessments were carried out in the present study. Two non-dietary PBDEs exposure routes, such as house dust ingestion and airborne inhalation, were considered. Health risks were assessed with the Monte Carlo simulation. Airborne PBDE levels from the present study and house dust PBDE levels from our previous study (Chao *et al.*, 2014b) were used to calculate daily non-dietary PBDEs intakes for toddlers, preschool and elementary school children, adolescents, and adults living in Taiwanese houses. Daily intakes of non-dietary PBDEs for Taiwanese were calculated using the equation of $DI_{indoor\ air\ inhalation} = C_{indoor\ air} \times IR_{inhalation\ rate} \times 0.95_{absorption\ rate} \times IEF_{indoor\ exposure\ fraction}/BW_{body\ weight}$ and the equation $DI_{dust\ ingestion} = C_{indoor\ dust} \times IR_{dust\ ingestion\ rate} \times 0.95_{absorption\ rate} \times IEF_{indoor\ exposure\ fraction}/BW_{body\ weight}$. Inhalation rates ($IR_{inhalation\ rate}$) for Taiwanese were obtained from the report "Compilation of Exposure Factors" (Taiwanese DOH, 2008), and house dust ingestion rates ($IR_{dust\ ingestion\ rate}$) were obtained from the "Child-specific Exposure Factors Handbook" (US EPA, 2008). The absorption rate of PBDEs in human intestinal tract was set at 0.95. Indoor exposure fraction (IEF) was from the "Compilation of Exposure Factors" (Taiwanese DOH, 2008) for Taiwanese adults and "Child-specific Exposure Factors Handbook" for Taiwanese infants to adolescents (US EPA, 2008). Average body weights of different age groups in Taiwan were according to the report on the Nutrition and Health Survey in Taiwan (NAHSIT) produced by the Health Promotion Administration of the Ministry of Health and Welfare (MOHW) from 2005 to 2008 (National Health Research Institute, 2015). Oral reference doses (RfDs) of BDE-47, 99, 153, and 209 (0.0001, 0.0001, 0.0002, and 0.007 mg/Kg/day, respectively) to evaluate their neurobehavioral effects and non-cancer risks (HQs) were provided by the US EPA IRIS (US EPA, 2008). The equation of $HQ = DI_{dust\ ingestion} \times EF \times ED/(AT_{nc} \times 365)$ from a previous study (Lim *et al.*, 2014) was used to estimate non-cancer risk in Taiwan, where DI, EF, ED, and AT_{nc} are daily intake, exposure frequency per year (day/year), exposure duration (year), and average time for non-cancer effects during exposure duration (year), respectively. Quantitative estimates of carcinogenic risk with neurobehavioral effects via oral ingestion of BDE-209 (oral slope factor = 0.0007 per mg/Kg/day) was also obtained from IRIS (US EPA, 2008). We calculated cancer risk (R) in the Taiwanese population using the equation R = CDI (chronic daily intake) × SF (slope factor). CDI was defined as $C_{indoor\ dust} \times IR_{dust\ ingestion\ rate} \times 0.95_{absorption\ rate} \times IEF_{indoor\ exposure\ fraction} \times EF \times ED/(BW_{body\ weight} \times AT \times 365)$, where AT was the average lifespan in Taiwan in 2012 (76.43 and 82.82 years for men and women, respectively) obtained from the Nutrition and Health Survey in Taiwan (NAHSIT).

Statistical Analysis

Measurements of airborne PBDEs below the limits of detection (LODs) were set to zero. The difference in level of airborne PBDE between indoor and outdoor samples was examined by the Mann-Whitney *U* test. Differences were considered to be significant when the *p* value was less than 0.05 or at the 95% confidence level. The Statistical Product and Service Solutions (SPSS) software, version 12.0, was used in the present study.

RESULTS AND DISCUSSION

Levels of Airborne PBDEs inside and outside Residential Homes

Table 1 shows airborne PBDE concentrations inside and outside residential homes in Taiwan (n = 3). BDE-209 was the predominant congener among 14 BDE congeners and accounted for 70.0% of the total in both indoor and outdoor air. Although the mean Σ_{14}PBDE and BDE-209 concentrations were higher indoors than outdoors, the difference was not significant ($p = 0.513$ for Mann-Whitney *U* test and $p = 0.254$ for paired samples *t* test) probably due to the small sample size. Concentrations of the 14 BDE congeners did not differ between indoor and outdoor air

Table 1. Air PBDE concentrations in indoor and outdoor environment from the resident houses (pg/m^3) (n = 3).

PBDEs	Indoor residence		Outdoor residence		Mann-Whitney *U* test	Paired samples T test		
	Mean ± SD	Range	Mean ± SD	Range	*p* value	Paired samples correlations r	Paired samples correlations *p* value	*p* value [a]
BDE-28	7.76 ± 12.7	< LOD–22.4	0.210 ± 0.191	< LOD–0.373	0.376	0.759	0.451	0.242
BDE-47	4.97 ± 5.74	0.0770–11.3	0.595 ± 0.479	0.0750–1.02	0.275	0,924	0.250	0.256
BDE-99	0.742 ± 0.586	0.111–1.27	0.517 ± 0.369	0.172–0.861	0.827	0.999	0.029*	0.234
BDE-100	0.136 ± 0.101	0.0200–0.206	0.0887 ± 0.0758	0.0170–0.168	0.275	0.749	0.462	0.251
BDE-153	0.659 ± 0.574	0.196–1.30	0.758 ± 0.595	0.264–1.42	0.513	1.000	0.013*	< 0.001***
BDE-154	0.415 ± 0.453	0.071–0.928	0.509 ± 0.532	0.0950–1.11	0.513	1.000	0.013*	0.137
BDE-183	1.00 ± 0.789	0.371–1.77	1.18 ± 0.762	0.584–2.04	0.513	0.991	0.083	0.127
BDE-196	0.659 ± 0.596	0.256–1.34	0.772 ± 0.544	0.396–1.40	0.275	1.000	0.011*	0.242
BDE-197	0.796 ± 0.895	0.201–1.83	0.908 ± 0.881	0.309–1.92	0.513	1.000	0.010*	< 0.001***
BDE-203	0.809 ± 0.777	0.294–1.70	0.930 ± 0.766	0.371–1.80	0.513	0.997	0.046*	0.125
BDE-206	2.23 ± 1.55	0.478–3.84	2.17 ± 1.06	1.34–3.37	0.827	0.974	0.146	0.625
BDE-207	2.56 ± 2.05	0.836–4.23	2.54 ± 2.12	0.962–4.95	0.827	0.993	0.074	0.493
BDE-208	1.57 ± 1.19	0.698–2.93	1.65 ± 1.13	0.989–2.96	0.827	0.985	0.109	0.497
BDE-209	56.8 ± 45.8	9.24–101	29.9 ± 17.6	13.7–48.7	0.513	0.980	0.128	0.253
Σ_{14}PBDEs	81.1 ± 71.9	13.1–156	42.7 ± 26.9	19.3–72.0	0.513	0.994	0.067	0.254

[a] Bootstrap for paired samples tests was performed using 3000 samples.

using the nonparametric Mann-Whitney *U* test. Using parametric tests (paired samples *t* test with 3000 bootstrap samples), concentration differences between indoor and outdoor air were only significant for BDE-153 ($p < 0.001$) and BDE-203 ($p < 0.001$), with higher levels outdoors. Fig. 1 shows the concentration of BDE homologues of different bromine number. As expected, airborne Σ_{14}PBDE levels were higher indoors than outdoors, particularly for BDE-209. Bradman *et al.*, (2014) found a possible correlation of higher indoor air levels of various BFRs with volatilization of BFRs or re-suspension of contaminated indoor dust.

The ambient outdoor air levels of PBDEs in residential neighborhoods were higher in the present study than in previous studies in a Pingtung rural area (15.9 pg/m^3) (Chao *et al.*, 2014c) and Kaohsiung urban area (35.3 pg/m^3) (Wang *et al.*, 2011), but lower than in two industrial areas of Kaohsiung (area of heavy industry and steel production [165 pg/m^3] and area of metals production [93.3 pg/m^3]) (Wang *et al.*, 2011) and three areas of Tainan (industrial: 58.5, urban: 88, rural: 55.4 pg/Nm^3) (Lin *et al.*, 2012). Our value had the similar magnitude with ambient levels of PBDEs in the urban areas, but lower than those in the industrial areas of Florence, Italy (Cincinelli1 *et al.*, 2014). Compared with the other brominated POPs like polybrominated biphenyls (PBBs) and polybrominated dibenzo-*p*-dioxins and furans (PCDD/Fs), PBDEs are the most abundant pollutants in the flue gas and outdoor environment (Chao *et al.*, 2014c; Wang *et al.*, 2014). Although PBDEs have the structural similarity with PCBs and PCDD/Fs, their emission sources and transportation routes are quite different. Discharge from vehicles or industrial flue gases is directly associated with PCB and dioxin emission, but higher PBDE concentrations are mainly from the electric equipments and low PBDE levels are associated with emission from PUF furniture and carpets (Zhang *et al.*, 2011; Zhou *et al.*, 2014).

Current Levels of Airborne PBDEs in Indoor Microenvironments

Table 2 shows airborne PBDE concentrations in residential houses, apartments, offices, daycare centers, cars, public facilities, and schools according to currently published and the present data. The levels of airborne PBDEs in residences are lower in Taiwan than in Sweden (Thuresson *et al.*, 2012), Vietnam (in houses near backyard electronic (e)-waste recycling sites; Tue *et al.*, 2013), Korea (Lim *et al.*, 2014), and the United States (Allen *et al.*, 2007; Bennett *et al.*, 2014), and slightly higher in Taiwan than Japan (Takigami *et al.*, 2009), United Kingdom (Harrad *et al.*, 2006), Sweden (in apartments; Thuresson *et al.*, 2012), Vietnam (in houses in suburban and rural areas; Tue *et al.*, 2013), and Australia (Toms *et al.*, 2009). Compared to their levels in other microenvironments listed in Table 2, PBDE levels in the houses or apartments (except Vietnamese houses near backyard e-waste recycling sites) were lower. Although several studies did not detect BDE-209 (Harrad *et al.*, 2006; Zhang *et al.*, 2011; Watkins *et al.*, 2013; Bennett *et al.*, 2014), BDE-209 was at least compositing of 50% in the environmental samples and human specimens (Chao *et al.*, 2014a). BDE-209 was the predominant BDE congener

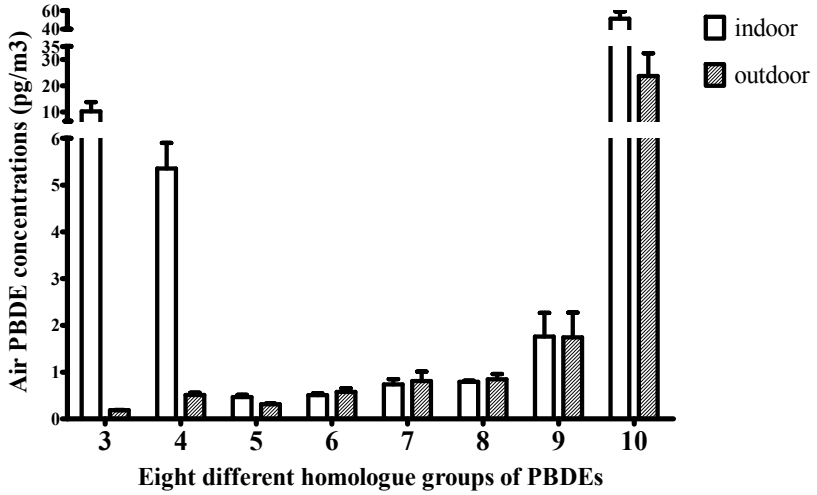

Fig. 1. The distribution of air-sample PBDE homologues indoors and outdoors.

in indoor air of various microenvironments except Korean elementary schools where the concentration of airborne BDE-47 was highest among 8 airborne BDE congeners (Lim *et al.*, 2014). Indoor air levels of PBDEs were lower in residences including houses and apartments than in other microenvironments (Table 2). This may be due to compliance with current restrictions placed by the Restriction of Hazardous Substances Directive 2002/95/EC (RoHS) on the use of PBDEs in BFR building materials for homes.

Non-Dietary PBDEs Daily Intake in Taiwan

Table 3 estimates the daily intake of non-dietary PBDEs by age groups of family members via house dust and indoor air in their living quarters. The estimates for house dust PBDE were taken from our previous report (Chao *et al.*, 2014b). In Table 3, the PBDE daily intake was highest (11,800 pg/Kg b.w./day or 162 ng/day) in toddlers (1–2 years old) and lowest in adult males (males: 808 pg/Kg b.w./day or 55.8 ng/day, females: 1050 pg/Kg b.w./day or 58.8 ng/day). In all the age groups, approximately 90% of exposure to PBDEs was non-dietary from house dust and BDE-209 was major. According to previous studies (Zhang *et al.*, 2011; Bennett *et al.*, 2014; Bradman *et al.*, 2014; Król *et al.*, 2014), PBDE levels in indoor air are weakly or slightly correlated with those in indoor dust. Although we cannot show correlations and the significances of differences in PBDE levels between house dust and indoor air in the present study, indoor air and dust levels of PBDEs can be used to evaluate daily intakes of non-dietary PBDEs for different age groups. House dust is the main source of non-dietary PBDEs exposure for humans especially toddlers and young children in residential indoor environments, and may be associated with several adverse health effects (Chao *et al.*, 2014a). Several reports propose that the indoor home environment is a major source of exposure to PBDEs in humans, especially the very young who spend a lot of time at home, play on the floor, and have frequent hand-to-mouth contact (Stapleton *et al.*, 2008; Stapleton *et al.*, 2012; Buttke *et al.*, 2013). Lim *et al.* (2014) indicated that the home among indoor environments was recognized as the largest contributor to daily exposure and health risk among Korean school children (daily dose: 80%, 16%, 3%, and 1% for home, elementary school, private academy, and public facility, respectively). In addition to dietary PBDE daily intake (67.95 ng/day for Taiwanese adults) (Chen *et al.*, 2012), non-dietary PBDE daily intake (adult males: 39.2 ng/day, adult females: 41.2 ng/day) estimated in the present study is also an important source of PBDEs exposure in Taiwan.

The estimated mean daily intake of indoor PBDEs by different age groups (infants, toddlers, children, teenagers, and adults) in the present study was comparable to those in studies from Heilongjiang, China (Zhu *et al.*, 2013), all China (Zhu *et al.*, 2015), and Busan, Korea (for adults and toddlers; Lee *et al.*, 2013); lower than the daily intake in studies from southern China (for house dust in homes near e-waste recycling sites; Zheng *et al.*, 2015) and northern Vietnam (Tue *et al.*, 2013), but higher than the daily intake in a study from northern Poland (for adults and toddlers; Król *et al.*, 2014). The primary sources of human exposure to PBDEs appear to be dietary, dust, air, handwipes, handkerchiefs, and direct skin contact. Bennett's (Bennett *et al.*, 2014) and Lim's (Lim *et al.*, 2014) studies found that PBDE levels in indoor dust are significantly correlated with levels in other environmental media. House dust is convenient to collect, easy to collect in large amounts, and PBDEs level in house dust is well correlated with that in other indoor media. Moreover, risk is easier to calculate from house dust data than from house indoor air and other environmental media data. House dust may be a good indicator of non-dietary PBDE exposure indoors.

Assessment of the Risks of Non-Dietary PBDE Exposure

According to the toxicological assessment data from the IRIS of the US EPA, chronic oral exposure to reference doses (RfDs) associated with neurobehavioral effects were only shown for four PBDE congeners (BDE-47, 99, 153, and 209). In Fig. 2, the HQs for human exposure to BDE-47, 99, 153, and 209 were calculated based on assumption of individual PBDE concentrations in house dust, house

Table 2. Current data of PBDE concentrations in indoor air (pg/m^3).

Indoor environment	BDE-47	BDE-99	BDE-100	BDE-153	BDE-209	ΣPBDEs	References
Japan							
Houses (n = 2) [a]	1.75	0.81	0.215	< LOD [b]	24	36; BDE-28,47,66,99,100,153,154,209	Takigami *et al.* (2009)
Sweden							
Houses (n = 10) [c]	23	8	NA [d]	1.2	290	330; BDE-28,47,99,153,183,197,206,207,208,209	Thuresson *et al.* (2012)
Apartment (n = 44) [c]	11	2.7	NA	0.74	24	58; BDE-28,47,99,153,183,197,206,207,208,209	Thuresson *et al.* (2012)
Offices (n = 10) [c]	210	320	NA	32	2000	4000; BDE-28,47,99,153,183,197,206,207,208,209	Thuresson *et al.* (2012)
Day care centers (n = 10) [c]	110	26	NA	1.2	1100	1400; BDE-28,47,99,153,183,197,206,207,208,209	Thuresson *et al.* (2012)
Cars (n = 24) [c]	36	44	NA	4.1	250	510; BDE-28,47,99,153,183,197,206,207,208,209	Thuresson *et al.* (2012)
United Kingdom							
Homes (n = 31) [a]	18.4	12.5	3.5	1.7	NA	52; BDE-28,47,49,66,85,99,100,153,154	Harrd *et al.* (2006)
Offices (n = 33) [a]	77.0	59.4	12.8	1.3	NA	166; BDE-28,47,49,66,85,99,100,153,154	Harrd *et al.* (2006)
Cars (n = 25) [a]	383	174	36.9	3.3	NA	709; BDE-28,47,49,66,85,99,100,153,154	Harrd *et al.* (2006)
Public (n = 3) [a]	3.3	21.0	4.2	< LOD	NA	112; BDE-28,47,49,66,85,99,100,153,154	Harrd *et al.* (2006)
Vietnam [e]							
Houses-suburban or rural areas (n = 5) [a]	1.86	0.58	0.88	0.144	5.18	12.0; Σ_{40}PBDEs	Tue *et al*. (2013)
Houses-EWB (n = 4) [a]	106	65.5	12.5	15	265	670; Σ_{40}PBDEs	Tue *et al*. (2013)
Korea							
Elementary school (n = 30) [a]	399	279	135	8	208	1060; BDE-28,47,99,100,153,154,183,209	Lim *et al.* (2014)
Private academics (n = 31) [a]	17	6	2	11	148	190; BDE-28,47,99,100,153,154,183,209	Lim *et al.* (2014)
Homes (n = 12) [a]	33	18	6	11	412	490; BDE-28,47,99,100,153,154,183,209	Lim *et al.* (2014)
Public facilities (n = 8) [a]	17	6	2	3	500	530; BDE-28,47,99,100,153,154,183,209	Lim *et al.* (2014)
Elementary schools CL-1 (n = 23) [a, g]	NS [h]	NS	NS	NS	NS	21.6-1440; BDE-28,47,99,100,153,154,183,209	Wu *et al.* (2010)
Elementary schools CL-2 (n = 13) [a, g]	NS	NS	NS	NS	NS	0.659-1660; BDE-28,47,99,100,153,154,183,209	Wu *et al.* (2010)
Computer rooms in elementary schools (n = 5) [a]	NS	NS	NS	NS	NS	134-220; BDE-28,47,99,100,153,154,183,209	Wu *et al.* (2010)

[a] Mean;
[b] Below the limit of detection;
[c] Median;
[d] Not analyzed;
[e] House-EWB means houses with backyard e-waste recycling sites. The authors did not list 40 BDE congeners in their published article;
[f] Geometric mean;
[g] CL-1: normal classrooms were dominated by higher-brominated congeners. CL-2: normal classrooms were moderately correlated with pentaBDE mixture;
[h] Not shown;
[i] Σ_{20}PBDEs is the sum of BDE-7,15,17,28,47,49,66,77,85,99,100, 119/120,126,138,153,154,183,184,191;
[j] Σ_{26}PBDEs is the sum of BDE-17,28,33,47,49,66,71,77,85,99,100, 119,126,138,153,154,156,166,183,184,191,196,197,206,207,209.

Table 2. (*continued*).

Indoor environment	BDE-47	BDE-99	BDE-100	BDE-153	BDE-209	ΣPBDEs	References
United States							
Houses with the young residents (n = 88)[f]	380	100	20	10	NA	NS[g]; BDE-47,99,100,153,154	Bennett *et al.* (2014)
Houses with the elderly residents (n = 48)[f]	730	120	20	5	NA	NS; BDE-47,99,100,153,154	Bennett *et al.* (2014)
Early childhood education facilities (n = 40)[a]	520	190	10	330	1630	1050; BDE-47,99,100,153,154	Bradman *et al.* (2014)
Offices (n = 31)[a]	263	82	19	5.8	NS	472; BDE-28/33,47,49,85,99,100,153,154,155	Watkins *et al.* (2013)
Homes-Personal air (n = 20)[f]	227	111	22.2	8.6	174	766; BDE-17,28/33,47,49,66,85/155,99,100,153,154,209	Allen *et al.* (2007)
Homes-Bedroom (n = 20)[f]	157	66.9	14.4	4	94.8	460; BDE-17,28/33,47,49,66,85/155,99,100,153,154,209	Allen *et al.* (2007)
Homes-Main living area (n = 20)[f]	145	60.3	12.0	3.5	94.2	453; BDE-17,28/33,47,49,66,85/155,99,100,153,154,209	Allen *et al.* (2007)
Canada							
Homes (10)[a]	25.5	6.75	2.18	6.12	NA	59; Σ_{20}PBDEs[i]	Zhang *et al.* (2011)
Offices (5)[a]	97.6	119	6.46	4.78	NA	173; Σ_{20}PBDEs	Zhang *et al.* (2011)
Australia							
Homes (n = 5)[a]	12.2	< LOD	< LOD	1.63	23.0	50.2; Σ_{20}PBDEs[j]	Toms *et al.* (2009)
Offices (n = 3)[a]	119	6.67	1.53	0.83	8.06	173; Σ_{20}PBDEs	Toms *et al.* (2009)
Taiwan							
Houses (n = 3)[a]	4.97	0.742	0.136	0.659	56.8	81.1; Σ_{14}PBDEs	The present study

[a] Mean;
[b] Below the limit of detection;
[c] Median;
[d] Not analyzed;
[e] House-EWB means houses with backyard e-waste recycling sites. The authors did not list 40 BDE congeners in their published article;
[f] Geometric mean;
[g] CL-1: normal classrooms were dominated by higher-brominated congeners. CL-2: normal classrooms were moderately correlated with pentaBDE mixture;
[h] Not shown;
[i] Σ_{20}PBDEs is the sum of BDE-7,15,17,28,47,49,66,77,85,99,100, 119/120,126,138,153,154,183,184,191;
[j] Σ_{26}PBDEs is the sum of BDE-17,28,33,47,49,66,71,77,85,99,100, 119,126,138,153,154,156,166,183,184,191,196,197,206,207,209.

Table 3. PBDEs daily intake by Taiwanese adults and toddlers via inhalation and dust ingestion in the residential environment (pg/Kg b.w./day).

PBDEs	Indoor air inhalation [a]		Indoor dust ingestion [b]		Non-dietary intake [c]	
	Mean	SD	Mean	SD	Mean	SD
Male adults (≥ 20 yr)						
Σ_{14}PBDEs	10.7	9.34	557	798	568	808
BDE-209	7.49	5.95	238	452	245	458
Female adults (≥ 20 yr)						
Σ_{14}PBDEs	9.51	8.42	727	1040	736	1050
BDE-209	6.66	5.37	310	589	317	595
Male adolescents (15–19 yr)						
Σ_{14}PBDEs	14.4	11.7	1370	1970	1387	1980
BDE-209	10.1	7.44	586	1110	596	1120
Female adolescents (15–19 yr)						
Σ_{14}PBDEs	11.3	9.98	1670	2390	1680	2400
BDE-209	7.89	6.36	713	1350	720	1360
Male adolescents (12–14 yr)						
Σ_{14}PBDEs	14.1	9.52	1750	2510	1760	2520
BDE-209	9.91	6.07	747	1420	757	1430
Female adolescents (12–14 yr)						
Σ_{14}PBDEs	11.7	10.4	1985	2840	1200	2850
BDE-209	8.20	6.61	848	1609	856	1616
Elementary school children (8–9 yr)						
Σ_{14}PBDEs	26.3	23.3	2800	4010	2820	4030
BDE-209	18.4	14.8	1200	2270	1210	2280
Preschool children (5–6 yr)						
Σ_{14}PBDEs	33.1	29.3	4280	6130	4310	6160
BDE-209	23.2	18.7	1830	3470	1850	3480
Toddlers (1–2 yr)						
Σ_{14}PBDEs	41.5	36.8	8180	11700	8220	11800
BDE-209	29.1	23.5	3490	6630	3520	6650

[a] PBDEs intake via indoor inhalation was based on Hearn *et al.* (2012) with the equation of $DI_{indoor\ air\ inhalation} = C_{indoor\ air} \times IR_{inhalation\ rate} \times 0.95_{absorption\ rate} \times IEF_{indoor\ exposure\ fraction}/BW_{body\ weight}$.

[b] PBDE levels in house dust were from our previous report (Chao *et al.*, 2014b). PBDEs intake via indoor dust digestion was modified from the Greek study (Besis *et al.*, 2014) with the equation of $DI_{dust\ ingestion} = C_{indoor\ dust} \times IR_{ingestion\ rate} \times 0.95_{absorption\ rate} \times IEF_{indoor\ exposure\ fraction}/BW_{body\ weight}$.

[c] Non-dietary intake was the sum of indoor air inhalation and indoor dust ingestion.

dust daily intake, exposure time, body weight, and average life time and were highest between ages 1 and 2 years and the lowest at age ≥ 20 years. All HQs were lower than the critical value of 1.00, indicating all age groups did not have a risk for neurobehavioral effects in the present study. The risks (HQs) of neurobehavioral effects from residential indoor air inhalation were not calculated for two reasons. One was that reference concentrations for chronic inhalation exposure (RfC) have not been estimated from 2008 to the present by US EPA and are still under review. The other was that the percentage of non-dietary PBDE daily intake via indoor inhalation is low. Our HQs were consistent with those reported in previous reports (Lim *et al.*, 2014; Li *et al.*, 2015; Zhu *et al.*, 2015), i.e., below 1.00, indicating that PBDEs in the indoor environment are not harmful to human health.

For the BDE-209 which is the predominant PBDE congener in the indoor environment and human bodies, life-time cancer risks (Rs) with neurobehavioral effects in Taiwan were assessed as 3.65×10^{-10} for men and 4.04×10^{-10} for women. Our R values were below the critical value of 1.00×10^{-6} with statistical significance and indicated that PBDEs in residential environments will not cause cancer associated with neurobehavioral effects in Taiwan. Li *et al.* (2015) revealed that potential health risk of PBDEs in the office environment is not associated with life time cancer risk (as suggested by extremely low Rs from 1.34×10^{-22} to 7.16×10^{-22}). After assessing health effects for the family members in the present study, the current levels of PBDEs in house dust and indoor air are extremely lower compared to the critical values. On the basis of our findings, we announced that PBDEs in indoor environment did not cause the negative impact on the family members if only non-cancer and cancer effects on neurological behavior were considered.

CONCLUSIONS

Non-dietary exposure to PBDEs and airborne levels of PBDEs are greater inside than outside the home. Among age groups, the toddlers had the highest daily intake and non-

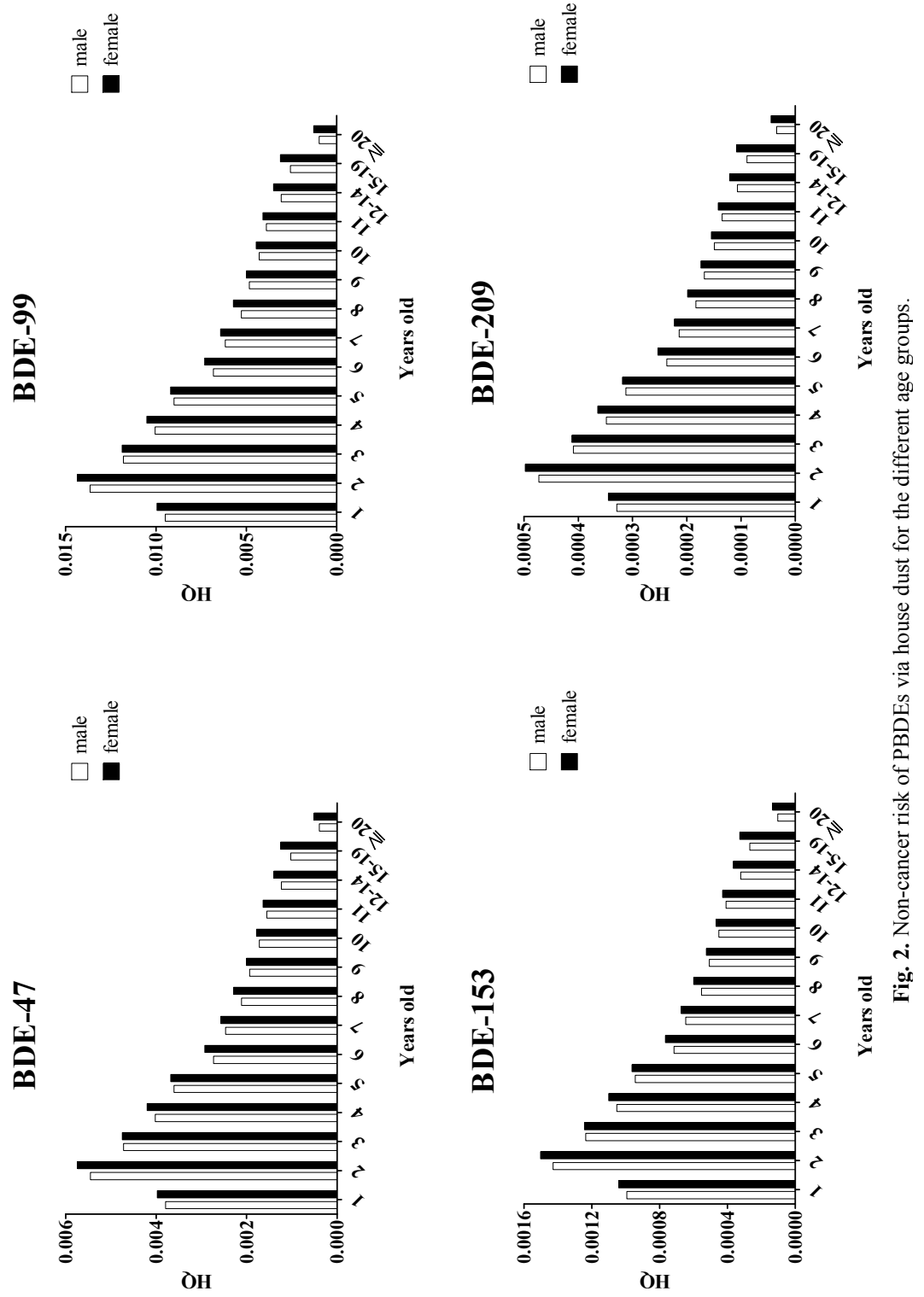

Fig. 2. Non-cancer risk of PBDEs via house dust for the different age groups.

cancer and cancer risks related with neurobehavioral effects from non-dietary PBDEs exposure in home environment. The estimated values for the toddlers were distinctly below the reference dose and threshold values for risk of cancer and non-cancer effects with neurobehavioral disorders, indicating that PBDEs levels inside houses are not harmful to family members.

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DISCLAIMER

The authors declare no conflicts of interest with regard to this study.

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