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Particle-Size Dust Concentrations of Polybrominated Diphenyl Ethers (PBDEs) in Southern Taiwanese Houses and Assessment of the PBDE Daily Intakes in Toddlers and Adults

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ABSTRACT

Our goal was to determine PBDE levels in Taiwanese household dust and to examine the particle-size distribution of PBDEs in dust samples. Nine paired samples of floor and electronic dust were gathered from nine individual houses. Each sample was sieved with 100 (< 0.149 mm) and 200 (< 0.074 mm) meshes to obtain three fractionated samples (> 0.149, 0.074–0.149, and < 0.074 mm). Each house had 8 samples taken from it, including a pair of overall dust samples and 3 pairs of fractionated samples. Thirty PBDEs (BDE-7, 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 139, 140, 153, 154, 156, 183, 184, 191, 196, 197, 203, 206, 207, 208, and 209) in 72 dust samples were analyzed by high resolution gas chromatograph/ high resolution mass spectrometry (HRGC/HRMS). The levels of Σ_{28}PBDEs and PBDEs in electronic dust were not significantly higher than those in floor dust in the paired overall dust samples, or the three fractionated samples. In addition, the levels of Σ_{28}PBDEs and PBDEs were not significantly different for these three particle-sizes of floor and electronic dust. Based on the results of the factor analysis, the patterns of the PBDE distributions in floor and electronic dust were also very similar. A significant correlation of Σ_{28}PBDEs was shown in the paired samples of floor and electronic dust. For the three fractionated dust samples, we found that only the levels of PBDEs from di- to tetra- in floor dust were significantly correlated with those in electronic dust. This may be because the PBDE contaminants in floor and electronic dust might originate from the same exposure sources, particularly for lower brominated PBDE congeners. The toddlers (6–48 months) (median estimated PBDE daily intake from mean floor dust: 2.73 ng/kg b.w./day) possibly ingested more PBDEs in household dust compared to the adults (0.0414 ng/kg b.w./day) based on our results. Our sample size was too small to represent the level of PBDE contamination in house dust from southern Taiwan, and thus a larger scale study is encouraged. In conclusion, our findings suggest that it may be not necessary for household surveys of PBDEs to collect different meshes of dust.

Keywords: Polybrominated diphenyl ethers; House dust; Electronic dust; Daily intake; Toddlers; Adults.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a class of brominated fire retardants used to decrease the flammability of consumer products (Alaee *et al.*, 2003). PBDEs contamination is a rising global concern, due to their lipophilicity, persistence, and bioaccumulation. Several epidemiological studies have reported that human exposure to PBDEs causes the adverse health effects, including worse birth outcomes (Chao *et al.*, 2007), disruption of thyroid hormones (Lin *et al.*, 2011; Shy *et al.*, 2012), interference with reproduction (Chao *et al.*, 2010), and effects on

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neurodevelopment (Chao *et al.*, 2011; Shy *et al.*, 2011). PBDEs are ubiquitous in the indoor environment due to their persistence and widespread use in consumer products, such as electronics (e.g., air conditioners, TV sets, and cellular phones), furniture, and electrical wiring and textiles. PBDEs have thus been recognized as significant persistent organic pollutants (POPs) in the indoor environment, and various studies show that PBDE concentrations in indoor house dust are several fold higher than those in outdoor house dust (Huang *et al.*, 2010; Yu *et al.*, 2012). A Chinese study examined PBDEs in central air conditioner filters from a new office building, and found that human exposure to PBDEs through inhalation and ingestion of indoor dust in the new building was less than that in old ones (Ni *et al.*, 2011). House dust samples from urban areas showed significantly higher levels of Σ_{32} PBDEs and BDE-209 than those from rural areas in Western Australia (Stasinska *et al.*, 2013). In the indoor environment, PBDEs in maternal and umbilical cord blood are associated with the levels of PBDEs in house dust (Frederiksen *et al.*, 2010). In a recent American study, dust pentaBDE concentrations had significantly positive correlations with serum levels of free thyroxine (Free T4), total triiodothyronine (Total T3), estradiol (E2), and sex hormone binding globulin (SHBG), but a negative association with follicle stimulating hormone (FSH); while dust octaBDEs was positively related with serum of free T4, thyroid stimulating hormones (TSH), luteinizing hormone (LH), and testosterone (T) and dust decaBDE was negatively correlated with T (Johnson *et al.*, 2013). In addition, toddlers (6–48 months) are estimated to have higher body burden of PBDEs from dust ingestion compared with adults (de Wit *et al.*, 2012). PBDEs are readily detected in samples of house dust and handwipes from American families with young children (Stapleton *et al.*, 2012), and for both toddlers and infants the indoor environment can be a key exposure route, through dust ingestion due to hand-to-mouth behavior as well as inhalation, thus leading to the accumulation of PBDEs in their bodies (Jones-Otazo *et al.*, 2005; Stapleton *et al.*, 2008). Although previous PBDEs studies have concentrated on residential exposures, little is known about fractionated dust exposure in the home environment. In Taiwan, most PBDEs survey studies were focused on PBDEs in outdoor environment and internal doses of PBDEs in human. PBDE concentrations in indoor environment including house and work place were still unknown in Taiwan. However, few reports have examined PBDE levels in the Taiwanese indoor environment, and this is the first study to examine the PBDE concentrations in indoor household dust. To the best of our knowledge, this work is also the first to examine three fractionated concentrations of PBDEs in house dust, including both floor and electronic dust.

METHODS

Chemicals and Reagents

This work used thirty PBDE standards (BDE-7, 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 139, 140, 153, 154, 156, 183, 184, 191, 196, 197, 203, 206, 207, 208, and 209) from Cambridge Isotope Laboratories (Andover, MA, USA) and nine ^{13}C-labeled PBDEs (BDE-15, 28, 47, 99, 153, 183, 197, 207, and 209) from Wellington Laboratories (Guelph, Canada). Sodium sulfate, alumina oxide, potassium oxalate, and silica gel of the highest grade were obtained from Merck (Darmstadt, Germany).

Samples Collection

The paired samples of indoor floor and electronic dust were collected from nine different houses selected from our previous PBDEs cohort. This PBDEs cohort contains 138 healthy mother-infant paired participants that were randomly selected from four hospitals in southern Taiwan between April 2007 and April 2010 (Shy *et al.*, 2012). The first criteria for selecting participants from our PBDEs cohort was as follow: (1) subjects were voluntarily willing to join this study after we informed them, (2) each floor and electronic dust had to weight more than 10 g, and (3) participants had a willingness to strictly follow our protocol. More than 100 cohort parents were invited to join this program; of these, 65 agreed to answer the detailed questionnaire. A total of 42 housed were initially enrolled. Of these 42 families, thirty-three subjects who did not offer sufficient floor dust or electronic dust were excluded. Only nine house dust samples were obtained for further chemical analysis.

Paired samples of floor and electronic dust were collected by our researchers using a vacuum cleaner (Nilfisk Advance Euroclean UZ934 HEPA canister vacuum cleaner) and following a strictly protocol. This high efficiency particulate air filter (HEPA filter) vacuum is recommended by Housing and Urban Development (HUD) and Occupational Safety and Health Administration (OSHA) for capturing and controlling lead paint dust, and is an ideal vacuum for cleaning hospitals, hotels, office, and homes. This vacuum cleaner has four stages of filtration, including a dust bag and washable pre-filter, which collect regular to fine particulates before the HEPA filter, as well as an exhaust filter. Samples were taken from ground floors, including living rooms, studies, and bedrooms, and consumer products, including the surfaces and filters of electronic goods. Dust was collected from each house two or three times within a three-month period. The collecting instruments were cleaned thoroughly by washing them with deionized distilled water and soapy water, and using an n-hexane-impregnated disposable wipe (Ali *et al.*, 2011). Dust was collected in the participants' houses, and then transported to the laboratory in National Pingtung University of Science and Technology. Each dust sample was immediately sieved through 100 (< 0.149 mm) and 200 meshes (< 0.074 mm) by shaking for 10 minutes, then homogenized and stored in a glass bottle with a Teflon cap at –20°C prior to analysis. Three fractionated samples, including > 100 mesh (1st fraction, > 0.149 mm), 100–200 mesh (2nd fraction, 0.074–0.149 mm), and < 200 mesh (3rd fraction, < 0.074 mm) dust, were separated in the floor and electronic dust using industrial steel stainless sieves. Seventy-two dust samples, including 18 overall and 54 fractionated samples, were packaged in chemically clean containers and transported to Cheng Shiu University in southern Taiwan to examine the PBDE levels they contained.

Analytical Procedure for PBDEs

Fractionated and overall dust samples (1–3 grams) in cellulose tubes were extracted by a Soxhlet extractor with 200 mL of toluene for 16 hours; these were then concentrated to dryness by a rotary evaporator. $^{13}C_{12}$-labeled PBDE congeners of BDE-15, 28, 47, and 99 (100 ng/mL), and 153, 183, 197, 207, and 209 (200 ng/mL) were added to the samples before the extraction. The extract was cleaned up as follows. The first cleanup involved treatment with concentrated sulfuric acid. The next cleanup procedure used a multi-layered silica column. The final cleanup procedure involved an acid alumina column. The eluate was collected, concentrated to near dryness by using a nitrogen stream, and then transferred to a vial. A total of 50 µL of ^{13}C-labeled BDE-139 was added to a vial containing the eluate as an internal standard after the clean-up prior to injection. The final extract was reduced in volume to 0.2 mL under a stream of nitrogen. PBDE levels were analyzed using a high resolution gas chromatography (Hewlett-Packard 6970) and high resolution mass spectrometry (Micromass Autospec Ultima) with 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 gas chromatography oven temperature was programmed to be held at 100°C for 4 min, to increase from 100°C to 200°C at a rate of 40 °C/min, to stay 3.5 min at 200°C, to change to 325°C by 10 °C/min, and to maintain 325°C for 2.5 min. 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 limits of detection (LODs) and quantification (LOQs) were defined as a signal-to-noise (S/N) higher than 3 and 10, respectively, for each chromatographic peak. The LODs for the 29 PBDE congeners from BDE-7 to 208 range from 0.262 to 46.0 pg/g. The LOD of BDE-209 is 333 pg/g. A field blank was taken during the individual sampling events to evaluate contamination during sampling. Laboratory blanks were analyzed for each batch (10–12 samples) of analyses. The data in the chemical assay were not blank corrected in this study. The mean recoveries of PBDE internal and labelled cleanup standards for the dust samples were within the standard recovery criteria (70%–130%).

Assessment of PBDE Daily intake from House Dust

We calculated the estimated PBDE daily intake in toddlers and adults via house dust based on the equation of $Intake_{dust} = C_{dust} \times R_d$ that has been used in previous reports (Wilford *et al.*, 2005; Chen *et al.*, 2011). Owing to our consideration that adults' weight and toddlers' weight are extremely different, the final equation was modified as $Intake_{dust} = C_{dust} \times R_d$/body weight. The overall dust PBDE concentrations from our nine selected Taiwanese families were expressed as C_{dust}. Five PBDE concentrations were evaluated, including the mean, median, 1st quartile, 3rd quartile, and 95% percentile. Two exposure scenarios were assumed for mean and high dust ingestion rates (R_d) in toddlers (6 to 24 months) and adults (Wilford *et al.*, 2005). The mean and high R_d values are 0.00416 and 0.1 g/day, respectively, for adults and 0.055 and 0.2 g/day, respectively, for toddlers. We assumed that averaged weight for adults and toddlers were 65 and 13 kg, respectively, based on Department of Health, Taiwan.

Statistical Analysis

The concentrations of PBDEs were not normally distributed using the Kolmogorov-Smirnov method. Spearmen's rank correlation coefficients were initially tested to examine the relations of PBDE homologues in paired samples of floor and electronic dust in overall and fractionated samples. The differences in PBDEs between floor and electronic dust or among three fractionated samples were examined by paired-samples T tests with 2,000 bootstrap samples. We used the bootsrap method to avoid statistical inaccuracy because our sampling size was small. We also used factor analysis to determine the associated characteristics of PBDE homologues from diBDEs to decaBDE. Analyses were carried out using the Statistical Package for Social Science (SPSS) version 12 (SPSS Inc, Chicago, IL, USA).

RESULTS AND DISCUSSION

PBDE Levels in the Whole House Dust

Table 1 shows the results of the descriptive analysis of 28 PBDE congeners for the overall house dust, including floor and electronic dust, from 9 selected houses. Two congeners of BDE-126 and 156 are not included, as in most measurements they are below their LODs. The means and standard deviations (SD) of Σ_{28}PBDE levels for the overall house dust were 2630 ± 3820 ng/g dry weight (d.w.) in floor dust and 4530 ± 4550 ng/g d.w. in electronic dust for the nine houses. The predominant congener in both floor and electronic dust was BDE-209, accounting for 41.1 and 62.7%, respectively, of all the PBDEs by weight. This finding is consistent with the results of several previous studies (Huang *et al.*, 2010; Chen *et al.*, 2011; Yu *et al.*, 2012; Johnson *et al.*, 2013; Stasinska *et al.*, 2013). A paired sample of the overall house dust (i.e., both floor and electronic dust) from one of the houses shows extremely higher levels of Σ_{28}PBDEs and 28 PBDE congeners compared to the results for the other eight homes, particularly for BDE-209 (data not shown). Compared with the indoor dust PBDE levels in different countries listed in a review article (Besis and Samara, 2012), the median PBDE levels in floor dust from our study were lower than those reported in the studies from America, Canada, the United Kingdom, Sweden, China, and Australia, but higher than those from Kuwait, Thailand, Belgium, and Portugal. Although the Σ_{28}PBDE levels in electronic dust were significantly higher than those in floor dust, there were no statistically significant differences in the paired overall dust levels of the 28 PBDE congeners and Σ_{28}PBDEs in the present study ($p < 0.05$) after we used paired *t* tests. These results are inconsistent with those in studies carried out in Japan and China (Takigami *et al.*, 2008; Huang *et al.*, 2010).

Table 1. Descriptive analysis of PBDE levels without fractionation in the floor and electronic dust of Taiwanese homes in southern Taiwan (ng/g d.w.).

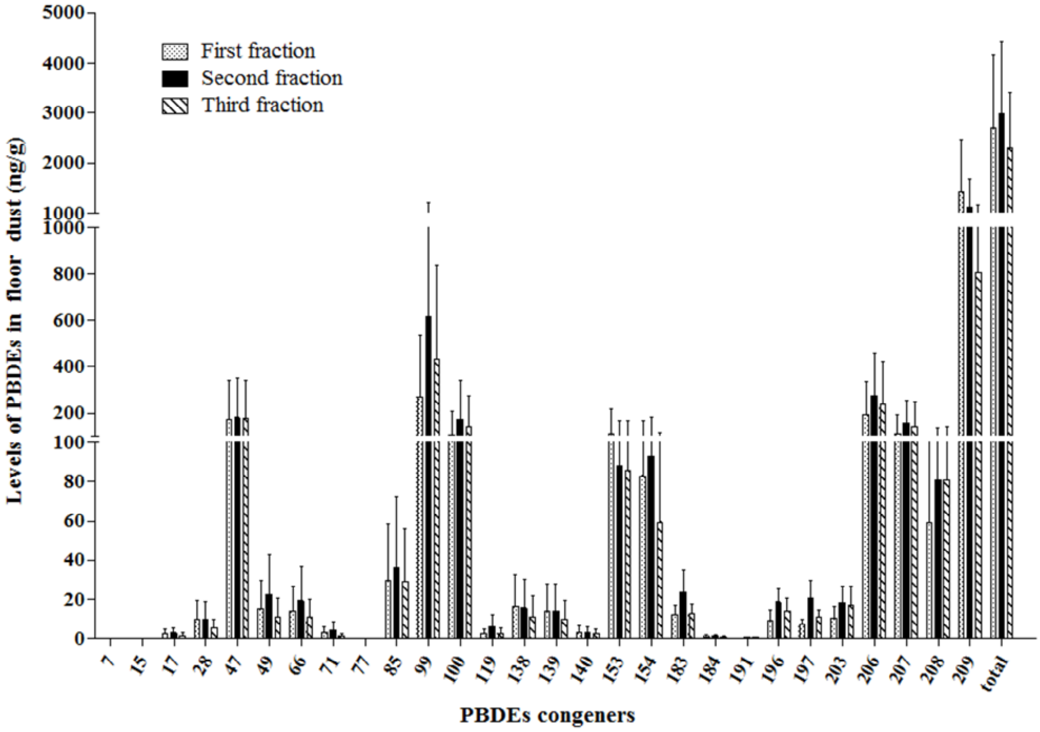
Congeners	Floor dust		Electronic dust		*p* value
	Mean ± SD	Median	Mean ± SD	Median	
7	0.034 ± 0.034	0.012	0.016 ± 0.017	0.030	0.666
15	0.105 ± 0.144	0.430	0.103 ± 0.123	0.100	0.222
17	2.35 ± 6.04	0.147	1.67 ± 2.61	0.330	0.340
28	8.00 ± 22.7	0.386	4.12 ± 7.17	0.500	0.863
47	178 ± 474	6.24	161 ± 407	22.3	0.605
49	15.8 ± 44.3	0.633	10.4 ± 19.0	1.23	0.931
66	14.3 ± 39.5	0.443	11.0 ± 20.6	1.12	0.546
71	2.84 ± 8.70	0.100	1.29 ± 2.19	0.200	0.666
77	0.238 ± 0.366	0.040	0.326 ± 0.479	0.130	0.190
85	31.4 ± 88.1	0.420	29.8 ± 82.8	1.56	0.546
99	445 ± 1276	9.95	370 ± 993	34.6	0.730
100	141 ± 392	1.66	91.0 ± 252	6.09	0.605
119	3.821 ± 11.5	0.152	3.61 ± 9.58	0.460	0.222
138	14.04 ± 39.8	0.456	14.8 ± 37.3	1.93	0.190
139	12.4 ± 35.1	0.267	12.3 ± 32.3	0.920	0.436
140	2.97 ± 8.28	0.113	3.31 ± 8.01	0.160	0.340
153	93.2 ± 257	2.662	110 ± 234	30.2	0.387
154	76.3 ± 221	1.26	80.0 ± 205	7.45	0.436
183	16.0 ± 22.5	6.26	46.0 ± 60.9	65.4	0.161
184	1.07 ± 1.99	0.323	1.59 ± 1.93	1.27	0.297
191	0.558 ± 0.553	0.449	0.903 ± 1.13	1.03	0.077
196	14.2 ± 18.01	8.98	24.9 ± 24.3	29.9	0.258
197	13.0 ± 17.8	8.79	25.8 ± 24.1	24.3	0.019*
203	15.5 ± 17.8	8.24	28.6 ± 34.0	20.9	0.258
206	236 ± 491	57.7	349 ± 466	204	0.340
207	138 ± 271	33.7	196 ± 252	127	0.387
208	75.0 ± 152	16.6	118.7 ± 165	66.0	0.190
209	1080 ± 2050	426	2840 ± 3800	1690	0.190
Σ_{28}PBDEs[a]	2630 ± 3820	645	4530 ± 4550	3180	0.387

[a] Σ_{28}PBDEs: Two congeners of BDE-126 and 156 are not included, as most measurements were less than the limits of detection.
* $p < 0.05$.

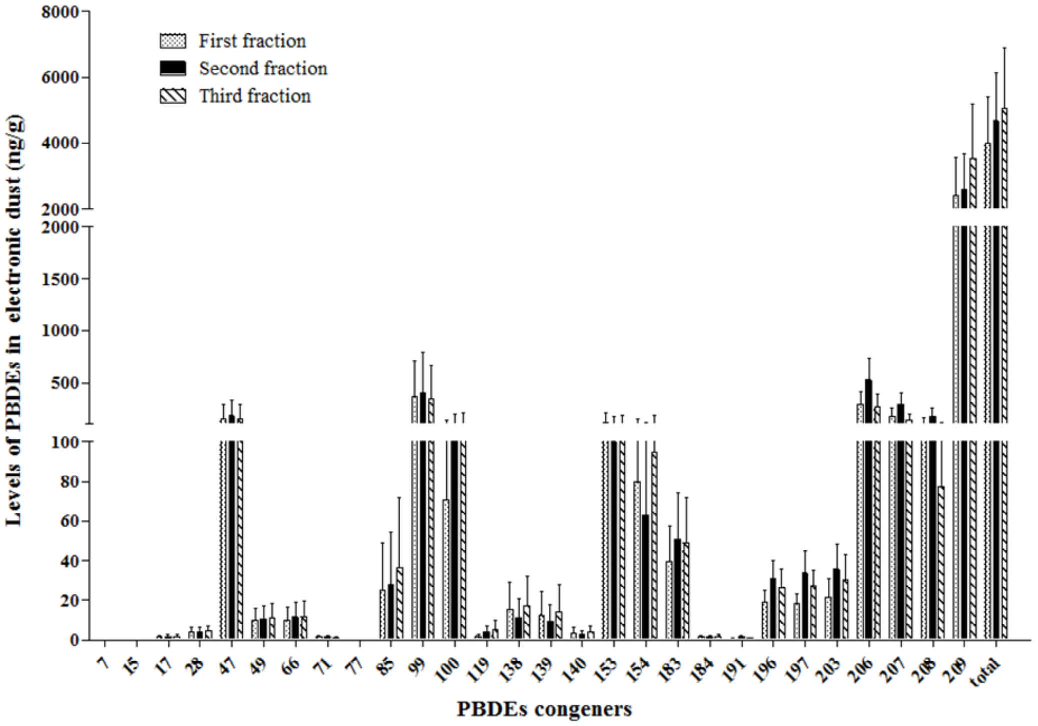
The differences were described as follows. Extremely high PBDE levels were found in dust from inside television sets, which suggested that PBDEs may be transferred from television components to indoor dust (Takigami *et al.*, 2008). The Chinese study also found significantly higher levels of ΣPBDEs in television and computer dust in comparison to house dust, and concluded that PBDEs-containing products used in the indoor environment were potentially important emission sources for PBDEs in indoor dusts (Huang *et al.*, 2010). The different results in the present study and these two previous reports are probably due to the small sample sizes used in all these works (five television sets in Takigami *et al.* (2008), two TV sets and two computers in Huang *et al.* (2010), and electronic dust from nine houses in the present study), as well as the fact that the present study collected dust from each house two or three times, and these homes contained many electronic goods, such as air conditioners, refrigerators, fans, computers, printers, television sets, microwaves, and ovens.

PBDE Levels in Fractionated Floor and Electronic Dust

The distributions of PBDEs in three particle-size dust samples, including floor and electronic dust, are shown in Fig. 1. The means of the Σ_{28}PBDE levels in the second fraction (0.074–0.149 mm) of floor dust and in the third fraction (< 0.074 mm) of electronic dust were highest among these three fractionated dust samples, but no statistically significant differences in levels of Σ_{28}PBDEs and 28 individual PBDE congeners were found among them ($p < 0.05$). Based on our findings, it is proposed that the particle size distribution of indoor dust PBDE concentrations may not be correlated with the particle sizes obtained by using industrial stainless steel sieves. We suggest that further studies examining PBDEs in house dust should use a sieve with mesh micron ≥ 149 µm (e.g., 50 mesh approximately to 0.297 mm) to remove coarse particles, human hair and dander, mold, pet, and cockroach waste. PBDEs were not consistently distributed in the three fractionated dust samples from the nine houses examined in this work, even though the third fraction (< 0.074 µm) accounted for 50–76% of the overall dust by weight, and the of PBDE levels in this fraction were not the highest among three fractionated dusts obtained from the nine homes. BDE-209 concentrations were the highest among



(A) PBDE levels in three fractions of floor dust



(B) PBDE levels in three fractions of electronic dust

Fig. 1. The distribution of 28 individual PBDE and Σ_{28}PBDE levels in three fractions of house dust, including floor (A) and electronic (B) dust.

those of all 28 PBDE congeners in all fractionated dust samples. This is not surprising, as BDE-209 is still used in Taiwan, while the mixtures of pentaBDEs and octaBDEs have been banned since 2005 (Koh *et al.*, 2010). The PBDE particle-size distributed patterns in floor and electronic dust were similar in all the houses examined in this work, except for seventh one (7 H) (Fig. 2.). BDE-209 accounted for 70–90% of the Σ_{28}PBDEs in most sampling houses, but BDE-209 accounted for 5–20% of Σ_{28}PBDEs in 7 H house. The main PBDE homologues were tetraBDEs, pentaBDEs, and hexaBDEs in 7 H house (data not shown). It should be noted that this was rarely cleaned and had a dirty indoor environment when the dust samples were collected in 7 H house. While the levels of PBDEs in floor or electronic dust may be influenced by numbers of electronics products in the indoor environment, how frequently these are used, and how often cleaning is done, although it remains unknown whether the latter factor affects the PBDEs size distribution in house dust. To the best of our knowledge, few studies of PBDEs have examined different particle sizes of indoor dust to explore their associations with human health, especially with regard to infants and toddlers, who

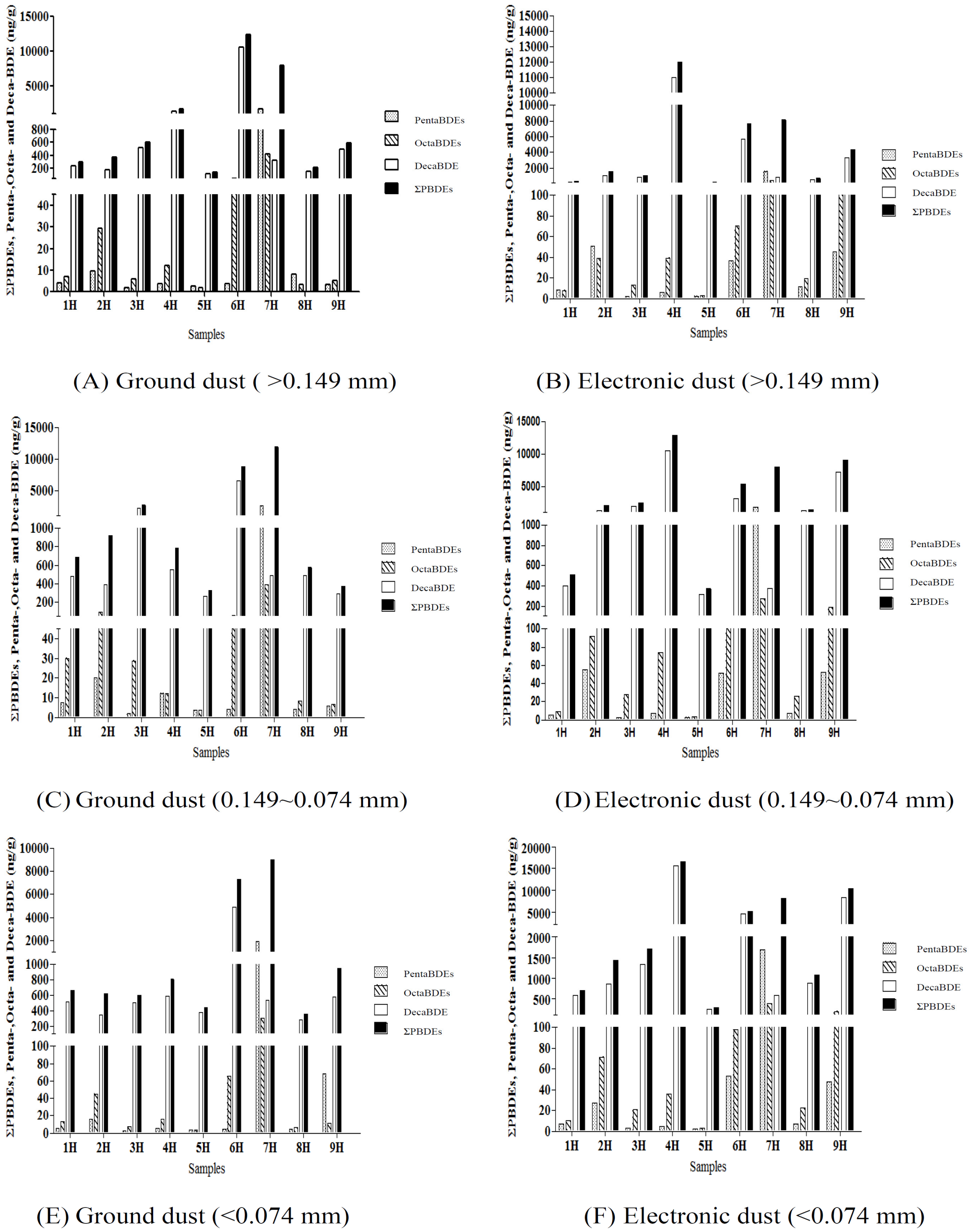


Fig. 2. Distribution of pentaBDEs, octaBDEs, decaBDE, and Σ_{28}PBDEs in fractionated floor and electronic dust from nine homes.

are more highly exposed to PBDEs than adults. According to our findings, it does not appear to be necessary to separate three or more different mesh dust samples using industrial stainless steel sieves for household dust in a survey of PBDEs. Although the levels of Σ_{28}PBDEs and 28 PBDEs were not associated with specific particle sizes of house dust in the present study, the PBDE levels in fine particles (e.g. particle size ≤ 2.5 µm) remain unknown, as do their effects on human health. Therefore, more studies are needed, especially longitudinal ones.

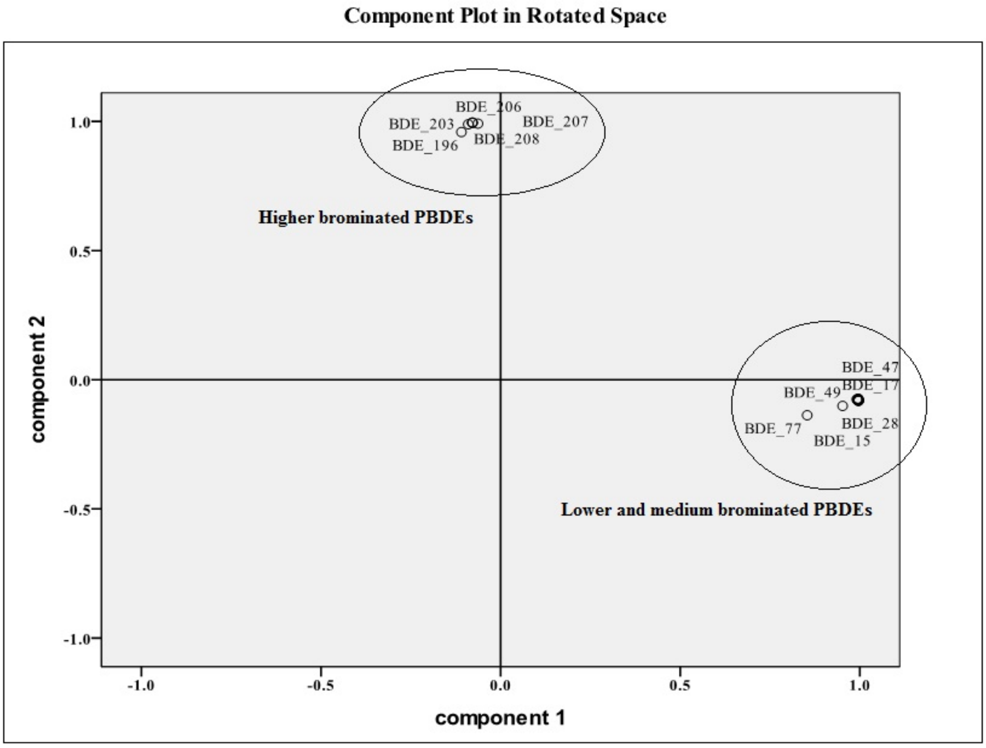
Correlations of Σ_{28}PBDEs and 28 PBDE Congeners between Floor Dust and Electronic Dust

The characteristics of 28 PBDE congeners were examined by factor analysis to reveal their exposure patterns in the overall floor and electronic dust, with the results shown in Fig. 3. In the floor dust, component 1 consisted of lower and higher brominated PBDEs from di to hepta (BDE-15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 138, 139, 140, 153, 154, and 184), and component 2 was composed of the highest brominated PBDEs, including octaBDEs, NonaBDEs, and decaBDE (BDE-196, 203, 207, 208, and 209). In electronic dust, component 1 and 2 comprised PBDEs from BDE-15 to 154 (BDE-15, 17, 28, 47, 49, 66, 71, 85, 99, 100, 119, 138, 139, 140, 153, and 154) and from 196 to 209 (BDE-196, 203, 207, 208, and 209), respectively. The PBDE patterns of three different particle sizes of dust are similar to those in the overall dust (data not shown). A previous report from China found a significant correlation between PBDE concentrations and air-conditioner filter dust and computer dust in a new building (Ni *et al.*, 2011). Ni *et al.* (2011) reported similarly distributed patterns of dust PBDEs in air-conditioner filter dust and computer dust, and speculated that PBDE contamination on the air-conditioner filter might be transferred from the surface PBDEs on computer dust. Furthermore, the factor analysis carried out in the present study shows that the contamination of PBDEs in floor dust and electronic dust may originate from the same exposure source.

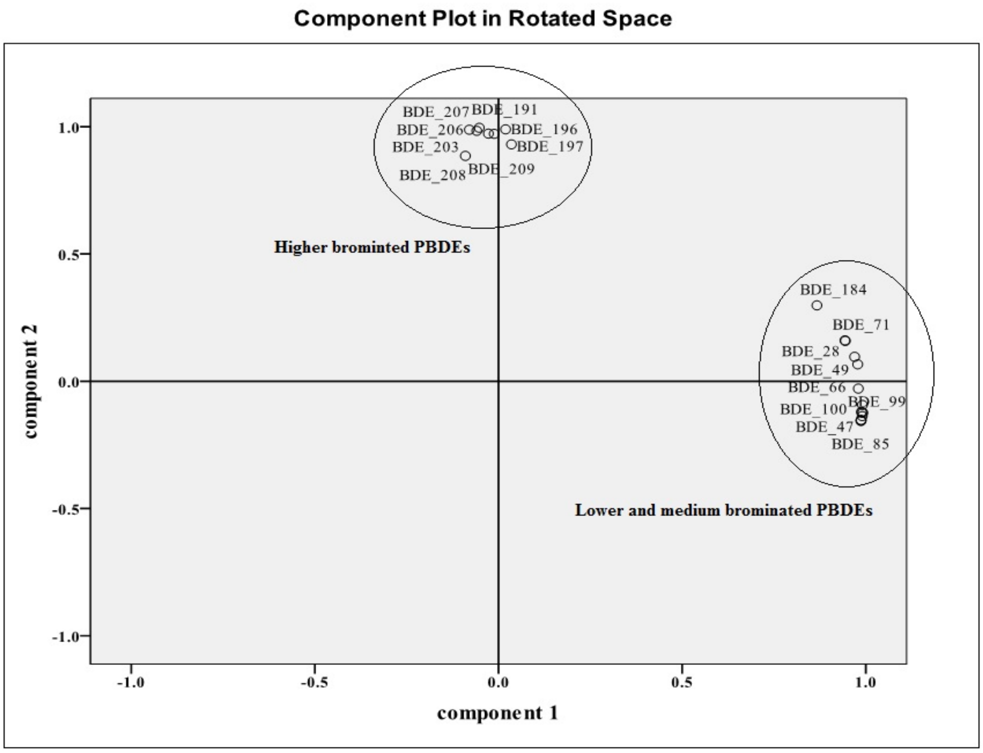
Significantly high correlation coefficients of Σ_{28}PBDEs between floor and electronic dust are found for the overall dust and first and second fractions (r = 0.694–0.738, p = 0.01–0.043), but a marginally significant correlation is found for the second fraction (r = 0.622, p = 0.074) (data not shown). The correlations of PBDE homologues in floor-electronic dust pairs are shown in Fig. 4. High Spearman's rho correlation coefficients of the low brominated PBDEs, from di to tetra, are found in floor and electronic paired dust samples in all three fractionated dusts (r = 0.667–0.933, $p < 0.05$). In the homologues of pentaBDEs (r = 0.683, p = 0.042) and hexaBDEs (r = 0.867, p = 0.002), significant associations in the paired dust are only observed in the third fraction. In the homologues of the highest brominated PBDEs, from nona to deca, significant associations appear in the first fraction of the paired dust samples (r = 0.733–0.967, $p < 0.05$). The paired correlations of octaBDEs, nonaBDEs, and decaBDE inconsistently vary in the three particle sizes of dust taken from the nine houses, and these three highest brominated PBDE homologues account for 70–95% of the total. Compared with diBDEs, triBDEs, and tetraBDEs, the lower correlation coefficients (r = 0.47–0.75, $p > 0.05$) of certain higher brominated PBDEs in floor-electronic dust pairs are not statistically significant, possibly due to following three reasons. First, our sampling size (n = 9) was too small to reach statistical significance, even though we used the bootstrap method (n = 2000) to increase our numbers. Second, high correlation coefficients (r > 0.75, $p < 0.05$) for the dust pairs were found in diBDEs, triBDEs, and teraBDEs. Stationary levels of PBDEs from di to tetra may exist in a Taiwanese indoor environment after debromination of the commercial decaBDEs mixture, since commercial products containing pentaBDEs and octaBDEs have been banned since 2005. High brominated PBDEs, especially BDE-209, are continuously released from the surfaces of electronic products kept indoors. Third, floor and electronic dust reflect the temporal and spatial concentrations of PBDEs, respectively. A recent British study that investigated PBDEs in indoor dust from two houses for eight consecutive months showed the existence of within-room temporal and within-building spatial variations in PBDE concentrations (Muenhor and Harrad, 2012). The PBDE levels in our study from nine homes might be associated with the number of the electronic products in use in these locations, the frequency with which they are used, and our sampling frequency. These associated factors may play the important roles in the PBDE levels in house dust.

Assessment of Estimated PBDE Daily Intake for Adults and Toddlers via House Dust

Table 2 shows the estimated PBDE daily intakes for adults and toddlers through ingestion of house dust in the Taiwanese general population. The daily intake for high and mean dust ingestion were obtained following the suggestions of previous studies (Wilford *et al.*, 2005; Chen *et al.*, 2011). Although five PBDE concentrations, i.e., mean, median, 1st quartile, 3rd quartile, and 95 percentile, are listed in Table 2, we only choose the median of PBDEs for further discussion, because the PBDE levels did not follow a normal distribution. The estimated median PBDE daily intakes via floor (2.73 ng/kg b.w./day) and electronic (13.5 ng/kg b.w./day) dust in toddlers for mean dust ingestion were 65.9 and 66.5 times compared to those in adults. The estimated daily PBDEs intakes in adults and toddlers from electronic dust were 4.9 times higher than those from floor dust. Recently, several studies have shown that the indoor environment to play an important role in human exposure to PBDEs, particularly for infants, toddlers, and young children, who spend a lot of time indoor and often engage in hand-to-mouth behavior (Jones-Otazo *et al.*, 2005; Wilford *et al.*, 2005; Stapleton *et al.*, 2008; Chen *et al.*, 2011; Stapleton *et al.*, 2012). According to our previous report (Chao *et al.*, 2011), BDE-209 was inversely associated with infants' cognitive scale. Toddlers will be ingested 10–66 fold dust BDE-209 compared to adults after our calculation. Parents and Department of Health have to concern about BDE-209 exposure in house dust. Our estimated median PBDEs daily intakes for adults and toddlers are comparable to



(A) Factor analysis of 28 PBDE congeners in the floor dust



(B) Factor analysis of 28 PBDE congeners in the electronic dust

Fig. 3. Spearman correlation coefficients of PBDE homologues for floor and electronic dust, $*p < 0.05$ and $**p < 0.01$.

those reported in both Canadian and Chinese studies (Wilford *et al.*, 2005; Chen *et al.*, 2011), but higher than those in studies from Singapore and Japan (Tan *et al.*, 2007; Takigami *et al.*, 2009).

CONCLUSIONS

This article is the first report to present PBDE levels in fractionated floor and electronic dust. There were no

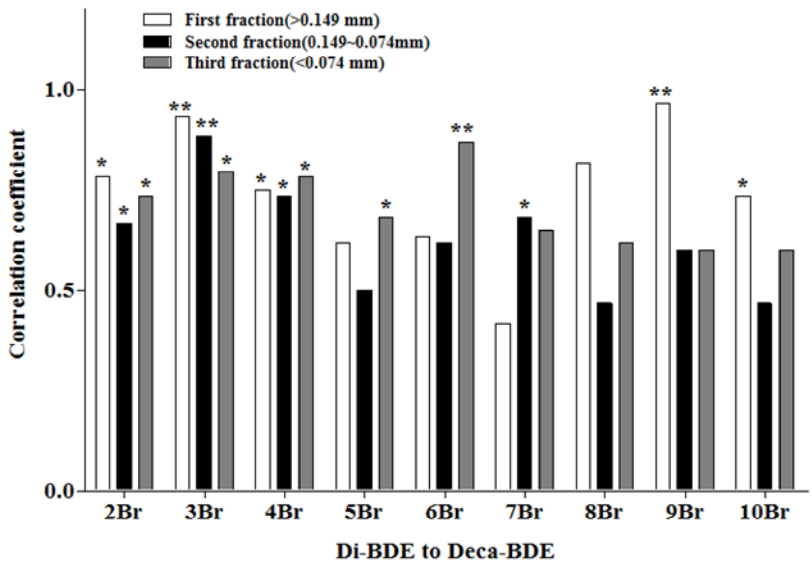


Fig. 4. Factor analysis of 28 PBDE congener concentrations in floor and electronic dust.

Table 2. Estimated daily intake of PBDEs without fractionation through house dust, including floor and electronic dust, for adults and toddlers in southern Taiwan (ng/kg b.w./day).

	PBDE daily intake via house dust[a]				
	Mean	Median	1st quartile	3rd quartile	95% percentile
Mean floor dust ingestion[b, c]					
Adults					
Σ_{28}PBDEs ingestion	0.171	0.0414	0.0314	0.345	0.565
BDE-209 ingestion	0.0720	0.0272	0.0172	0.0554	0.371
Toddlers					
Σ_{28}PBDEs ingestion	11.3	2.73	2.08	22.2	37.3
BDE-209 ingestion	4.75	1.80	1.15	3.66	24.5
High floor dust ingestion[d]					
Adults					
Σ_{28}PBDEs ingestion	4.11	0.992	0.755	8.31	13.6
BDE-209 ingestion	1.73	0.655	0.415	1.33	8.89
Toddlers					
Σ_{28}PBDEs ingestion	41.7	9.92	7.55	83.0	136
BDE-209 ingestion	17.3	6.55	4.15	13.3	88.9
Mean electronic dust ingestion					
Adults					
Σ_{28}PBDEs ingestion	0.617	0.203	0.0758	1.34	1.44
BDE-209 ingestion	0.338	0.154	0.0505	0.742	1.03
Toddlers					
Σ_{28}PBDEs ingestion	40.7	13.5	5.02	88.3	95.5
BDE-209 ingestion	22.4	7.14	3.33	49.0	68.3
High electronic dust ingestion					
Adults					
Σ_{28}PBDEs ingestion	14.8	4.89	1.83	32.1	34.7
BDE-209 ingestion	8.15	2.60	1.21	17.8	24.8
Toddlers					
Σ_{28}PBDEs ingestion	148	48.9	18.2	321	347
BDE-209 ingestion	81.5	25.9	12.2	101	248

[a] PBDE exposure via dust ingestion is evaluated using the equation of "$Intake_{dust} = C_{dust} \times R_d$" by Wilford *et al.* (2005). C_{dust} is PBDE levels in house dust (ng/g). R_d is dust ingestion rate (g/day).
[b] Mean R_d values are 0.00416 g/day for adults and 0.055 g/day for toddlers, respectively.
[c] We assumed that averaged adults' weight was 65 kg and averaged toddlers' weight was 13 kg, respectively.
[d] High R_d values are 0.1 g/day for adults and 0.2 g/day for toddlers, respectively.

significant differences in Σ_{28}PBDEs and certain PBDEs between the paired samples of floor and electronic dust overall, or in the three particle size dust samples. The levels of Σ_{28}PBDEs and 28 individual PBDE congeners were not significantly different among three fractionated floor or electronic dust samples (> 0.149, 0.074–0.149, and < 0.074 mm). The PBDEs in floor and electronic dust probably originated from the same source, due to the significantly high correlation coefficient of Σ_{28}PBDEs in the floor and electronic dust paired samples, and the similar PBDEs distribution patterns in floor and electronic dust in the factor analysis results. Compared with Taiwanese adults, it is proposed that toddlers may ingest more PBDE contaminants from floor and electronic dust after the estimated PBDE daily intakes of these two groups were assessed. In future works, longitudinal and long-term studies that examine toddlers' exposure to PBDEs via household dust, as well as the related health affects, are urgently needed in Taiwan.

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DISCLAIMER

The authors declare no conflicts of interest.

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