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Indoor and Outdoor Concentrations of Polybrominated Diphenyl Ethers on Respirable Particulate in Central and Southern Taiwan

How-Ran Chao[1+], Danielle E. Que[2+], Yan-You Gou[1], Chun-Yu Chuang[3], To-Yao Chang[1], Yi-Chyun Hsu[4*]

[1] *Emerging Compounds Research Center, Department of Environmental Science and Engineering, National Pingtung University of Science and Technology, Pingtung County 912, Taiwan*
[2] *Department of Environmental Engineering, National Cheng Kung University, Tainan City 701, Taiwan*
[3] *Department of Biomedical Engineering and Environmental Sciences, National Tsing Hua University, Hsinchu City 300, Taiwan*
[4] *Department of Environmental Engineering, Kun Shan University, Tainan City 710, Taiwan*

ABSTRACT

High levels of fine particulate ($PM_{2.5}$) in indoor and outdoor air have globally threatened human health and environment. There are still few studies which concern on the emerging persistent organic pollutants like polybrominated diphenyl ethers (PBDEs) bound on $PM_{2.5}$. The aim of this study was to investigate PBDEs in $PM_{2.5}$ in various outdoor (metropolis, industrial, and rural areas) and indoor (library, rail station, hospital, supermarket, department store, and office) environments. $PM_{2.5}$-bound PBDEs was analyzed by high resolution gas chromatography/high resolution mass spectrometry after $PM_{2.5}$ was collected. Mean levels of $PM_{2.5}$-bound Σ_{14}PBDEs were 79.0 and 116 pg m^{-3} in outdoor and indoor air, respectively. Compared to other outdoor locations, the industrial sites, Taixi (169 pg m^{-3}) in particular, has the highest $PM_{2.5}$-bound PBDEs levels which might be attributed to nearby industrial activities and indoor to outdoor migration behaviors. For indoor air, $PM_{2.5}$-bound PBDEs mean concentrations (libraries, rail stations, department stores, offices, hospitals, and supermarkets) were found to be 357, 35.3, 50.2, 73.2, 59.2, and 124 pg m^{-3}, respectively. The high indoor $PM_{2.5}$-bound PBDEs levels found in libraries are heavily affected by the presence of indoor electronic equipment or other consumer products. Similarly, this is also true for supermarkets which merchandise electronic consumer products. Although the abundant congener of deca-BDE consisted of 74.7% and 48.03% of Σ_{14}PBDEs in the indoor and outdoor air, respectively, nona-BDEs predominantly contributed 11.6% in the indoors while tri- and tert-BDES contributed 11.3% and 16% in the outdoors. Higher brominated PBDEs are more likely due to their emission from electronic surfaces while lower brominated PBDEs are products of photochemical degradation. Other factors affecting both the indoor and outdoor air $PM_{2.5}$-bound PBDE homologue levels such as migration behaviors might also be considered.

Keywords: Polybrominated diphenyl ethers (PBDEs); $PM_{2.5}$; Fine particulate; Aerosol; Indoor; Outdoor.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a class of emerging persistent organic pollutants which ubiquitously exist in both the aerosol indoors and outdoors. They are also a common ingredient in brominated flame retardants (BFRs) which are widely used on a variety of products such as electronic appliances, textiles, paints, etc. (Wang *et al.*, 2007; Qi *et al.*, 2014). PBDEs have been regarded as a major concern due to their ubiquitous existence in the environment (e.g., form the source to the surrounding air) and their harmful health effects. They are also highly resistant to biological, chemical, and physical degradation, and tend to bio accumulate easily due to their lipophilic property (Cincinelli *et al.*, 2014; Tung *et al.*, 2014). PBDEs negatively affect the endocrine and reproductive functions of humans most especially in females (Hood, 2006; Shy *et al.*, 2012) and cause neurodevelopmental delays after neonatal ingestion of breast milk with high PBDE level (Lin *et al.*, 2010; Chao *et al.*, 2011; Shy *et al.*, 2011; Gascon *et al.*, 2012; Shy *et al.*, 2012). The use of commercial PBDE technical formulations such as penta-BDE, octa-BDE, and deca-BDE have been restricted and voluntarily discontinued in several parts of the world including Europe and in some

[+] These authors are equally contributed.

[*] Corresponding author.
Tel.: 886-6-272-7175 ext. 500; Fax: 886-6-2056732
E-mail address: ychsu22@yahoo.com.tw

locations in USA (Besis and Samara, 2012). However, the use of deca-BDE in Taiwan has not yet ceased as of present and remains available in the manufacture of its many consumer products (Chao *et al.*, 2014).

Due to fine particle deposition, PBDEs bind on particulate matters with aerodynamic diameter less than 2.5 μm, $PM_{2.5}$, respectively (Mandalakis *et al.*, 2009; Li *et al.*, 2015a). Numerous studies have been conducted to investigate $PM_{2.5}$ levels due to the possibility of its relatively small particles being deposited more deeply into the lungs causing respiratory ailments and diseases, and other negative health problems (Zhang *et al.*, 2013; Guo *et al*., 2015a; Li *et al.*, 2015b; Shi *et al.*, 2015; Gao *et al.*, 2016). People with known history of cardiopulmonary problems face a higher risk of mortality due to the increasing $PM_{2.5}$ concentration in the atmosphere and so is the prevalence of respiratory problems in normal people (Dominici *et al.*, 2006; de Oliveira *et al.*, 2012; Xing *et al.*, 2016).

Particle-bound PBDEs are difficult to eliminate in the atmosphere and tend to have a longer residence time and are easily transported even at a longer distance (Xu *et al.*, 2016). PBDEs easily diffuse from the surface of BFR-coated products present in the indoor environment and unto $PM_{2.5}$ particles via volatilization or abrasion (Rauert and Harrad, 2015). The Σ_{15}PBDEs mean concentrations in indoor $PM_{2.5}$ in the office environment in Shanghai, China was found to be 51.8 pg m^{-3} (Xu *et al.*, 2016). In five Hong Kong kindergartens, PBDEs concentrations in indoor $PM_{2.5}$ are 0.10–0.64 ng m^{-3}, respectively (Deng *et al.*, 2016). PBDEs can exist in indoor $PM_{2.5}$ and outdoor $PM_{2.5}$ with its transport relative to both. The presence of $PM_{2.5}$ in the indoor environment is mainly attributed to both outdoor and indoor sources, through ventilation systems and infiltration (Weschler, 2004; Meng *et al.*, 2005; Lim *et al.*, 2011). In general, indoor air PBDE concentrations have been found to be much higher compared to the outdoor air PBDE concentrations due to the presence of home electronic devices and a higher risk of exposure due to majority of the people preferring the indoor environment (Wilford *et al.*, 2004; Hazrati and Harrad, 2006). Indoor occupational exposure of PBDE is also prominent in human adults especially those working in the e-waste recycling and dismantling factories (Julander *et al.*, 2005; Ma *et al.*, 2009; Muenhor *et al.*, 2010; Gou *et al.*, 2016a). If only fine particles are considered, PBDE concentrations are also observed to be higher in indoor $PM_{2.5}$ compared to the outdoor $PM_{2.5}$ possibly due to several factors such as weak indoor air circulation, slow degradation, low ventilation rate from indoors to outdoors, and high pollutant emission from consumer products (Stapleton *et al.*, 2008; Lv *et al.*, 2015; Gou *et al.*, 2016a; Xu *et al.*, 2016).

Exposure routes for PBDEs include dust ingestion, dietary intake and inhalation of particulates for both adults and young children. Previous studies showed that humans, particularly the younger population, are more likely to be exposed in the indoor environment than in the outdoor environment (Chao *et al.*, 2014; Shy *et al.*, 2015; Gou *et al.*, 2016a). A risk assessment study conducted by Gou *et al.* (2016b) showed that school-age children are more likely to be exposed to PBDEs from the indoor environment particularly in their homes compared to their indoor classrooms. The study on the effects of airborne PBDEs on human health through exposure in the indoor environment is still limited considering that the inhalation of these particulates is just a minor exposure route compared to dietary intake and indoor dust digestion as observed in most of PBDEs risk-assessment studies. $PM_{2.5}$ is known to cause adverse health effects especially on the human cardiorespiratory system but $PM_{2.5}$-bound PBDEs health effects are still unknown and yet to be confirmed. In the present study, levels of PBDEs in $PM_{2.5}$ were investigated in various indoor and outdoor environments located in central and southern regions of Taiwan.

METHODS

Chemicals and Reagents

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

Air Sampling and Collection

The study design in the present study was focused on the screened survey of PBDE concentrations on $PM_{2.5}$ particulate in outdoor air from the different types of the areas (i.e., industrial, metropolis, and rural areas) and in indoor air from various types of the microenvironments in Taiwan. The regulated air pollutants were collected in the indoor and outdoor air based on the standard methods from Environmental Protection Administration in Taiwan (TEPA). Twenty-eight outdoor air samples were gathered from seven TEPA air monitoring sites between May 5 and May 8 or between October 2 and October 5. Indoor air samples were obtained from 24 indoor environments including 3 liberies, 3 rail stations, 3 department stores, 3 offices, 6 hospitals, and 6 supermarkets from May to October in 2013. The sampling processes and analytical methods were followed by announcement of TEPA standard methods. The TEPA standard methods used in the present study were as follows: $PM_{2.5}$ (NIEA A205.11C), PM_{10} (NIEA A206.10C), carbon dioxide (CO_2) (NIEA A448.11C), carbon monoxide (CO) (NIEA A421.12C), ozone (O_3) (NIEA A420.11C), total volatile organic compounds (TVOCs) (NIEA A732.10C), formaldehyde (HCHO) (NIEA A705.11C), total bacterial count (TBC) (NIEA E301.11C), and total fungal count (TFC) (NIEA E401.11C). The $PM_{2.5}$ outdoor samplers were PQ200 Ambient Air Particulate Samplers form BGI by Mesa Labs (New Jersey, USA). The $PM_{2.5}$ indoor samplers, Model 200-Personal Environmental Monitor^{TM} (PEM^{TM}), were purchased from MSP Corporation (Minnesota, USA). To avoid the amount of $PM_{2.5}$-bound PBDEs below detection limits, the 4 outdoor air samples were pooled from the

continuous 4-day sampling samples and the 3 indoor air samples in the same category were pooled together. Prior to PBDEs analysis, the $PM_{2.5}$ samples were kept at –20°C after the condition and weighting of the glass fiber filters.

Extraction, Cleanup, and Analysis

The extraction, cleanup procedures, and analysis of airborne PBDEs in the present study were followed by our previous publication with minor modification (Chao *et al.*, 2014; Shy *et al.*, 2015). Briefly, pre-labeled isotopes and identifiable surrogate standards were spiked in the filters to evaluate the PBDEs loss during the sampling process prior to aerosol sampling indoors and outdoors. Owing to monitoring of the extraction and cleanup processes, the internal standards were spiked into the samples and well mixed with toluene before aerosol samples were extracted with toluene for 24 h in a Soxhlet extractor. After extraction, the extracts were then concentrated, treated with concentrated sulfuric acid, and passed through a multicolumn system installed with acid silica, alumina, and activated carbon columns. The eluate solution was gathered, evaporated, and concentrated to near dryness by using a gentle stream of gaseous nitrogen prior to transference into 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 elute of PBDEs in a vial was re-dissolved by toluene. The final extract was reduced in volume to 0.2 mL under a stream of nitrogen.

PBDEs in the final extract were determined by high-resolution gas chromatography with high-resolution mass spectrometry (HRGC/HRMS) (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. A DB-5HT column (L = 15 m, i.d. = 0.25 mm, film thickness = 0.1 μm) (J&W Scientific, Folsom, CA) was installed on HRGC in splitless mode at 280°C with constant helium flow of 1 mL min^{-1}. The temperature program of HRGC oven stably maintained at 100°C in the first 4 min, steadily increase to 200°C at a rate of 40 °C min^{-1} from 100 to 200°C, hold at 200°C for 3.5 min, rapidly increase to 325°C at a rate of 10 °C min^{-1}, and continued at 325°C for the last 2.5 min. The electron energy and source temperature were specified at 35 eV and 250°C in HRMS, 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^{-1} and the LOD of BDE-209 was 314 pg g^{-1}. 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.

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

The indoor and outdoor air quality in the present study was shown in Table 1. Median levels of PM_{10}, $PM_{2.5}$, and CO were 54.8 μg m^{-3}, 24.4 μg m^{-3}, and 0.600 ppm in the indoor air and 35.8 μg m^{-3}, 18.7 μg m^{-3}, and 0.330 ppm in the outdoor air, respectively. In the indoor air quality (IAQs) from various microenvironments including hospitals, rail stations, department stores, offices, and supermarkets, median concentrations of CO_2, O_3, HCHO, and TVOCs were investigated as 706, 0.0278, 0.0421, and 0.0254 ppm, respectively. Measurements of most regulated IAQ pollutants were beneath Taiwanese IAQ standards except for TVOCs in a department store and HCHO in a supermarket. For bioaerosol in the indoor environment, TBC (median: 730 CFU m^{-3}) and TFC (439 CFU m^{-3}) concentrations met the Taiwanese IAQs standards. Our IAQ values in the present study were comparable to our previous report (Hsu *et al.*, 2015). In comparison with outdoor PM_{10}, indoor PM_{10} levels are higher. The same phenomena is also observed in $PM_{2.5}$ levels. Several studies revealed that indoor PM including coarse, fine, and submicron particulate possibly originated from outdoor sources (Chen *et al.*, 2016; Raysoni *et al.*, 2016; Wang *et al.*, 2016). The presence of $PM_{2.5}$ and PM_{10} in the indoor environment might be mainly attributed to the indoor equipment, occupant activities, and outdoor air including building structure, ventilation system, air exchange rate, and ambient conditions (Tippayawong *et al.*, 2009; Madureira *et al.*, 2012; Hassanvand *et al.*, 2014; Chen *et al.*, 2016; Raysoni *et al.*, 2016; Wang *et al.*, 2016). A close indoor and outdoor relations for $PM_{2.5}$ was found during unoccupancy and occupancy while the only weak correlation appeared to be that of PM_{10} during occupancy in primary schools (Chen *et al.*, 2016). Othman *et al.* (2016) reported a lower indoor PM_{10} level due to low occupant activities during office hours. Aside from occupancy, the presence of many consumer products like printers in the indoor environment also affects PM_{10} levels as reported by several studies (Lee *et al.*, 2001; He *et al.*, 2007; Kagi *et al.*, 2007).

Table 1. Air quality in indoor and outdoor air.

	Indoor air[1] Range (median)	Outdoor[2] Range (median)
Particulate matter		
PM_{10} (μg m^{-3})	26.3–458 (54.8)	24.1–73.0 (35.8)
$PM_{2.5}$ (μg m^{-3})	14.4–28.0 (24.4)	14.4–42.3 (18.7)
Indoor quality		
CO (ppm)	0.375–0.840 (0.600)	0.134–0.356 (0.330)
CO_2 (ppm)	606–931 (706)	-
O_3 (ppm)	0.0163–0.0458 (0.0278)	-
HCHO (ppm)	0.0118–0.101 (0.0421)	-
TVOC (ppm)	0.0109–0.816 (0.0254)	-
Bacteria (CFU m^{-3})	297–974 (730)	-
Fungi (CFU m^{-3})	205–901 (439)	-

[1] Monitoring of Indoor air quality in the present study is regulated by Taiwanese EPA.
[2] Monitoring of PM_{10}, $PM_{2.5}$, and CO in the present study was obtained from the ambient monitoring sites between May 6 and May 8 or between October 2 and October 5 in 2013.

Table 2 shows the $PM_{2.5}$-bound Σ_{14}PBDEs mean concentrations in different outdoor locations in central and southern Taiwan which were grouped under metropolis areas, industrial areas, and rural areas. The mean concentrations of $PM_{2.5}$-bound Σ_{14}PBDEs in the metropolis areas which includes Xitun, Douliu, and Tainan are 74.3, 76.3, and 53.0 pg m^{-3}, respectively. For industrial areas like Mailiao and Taixi, the observed mean concentrations are 54.5 and 169 pg m^{-3}. Lastly, the rural areas which include Hengchun and Meinong, has mean concentrations of 74.3 and 51.0 pg m^{-3}, respectively. However, in the present study the mean levels in rural areas are relatively similar to that of the metropolis areas (urban) and the highest mean concentrations of Σ_{14}PBDEs in $PM_{2.5}$ was observed to be in industrial areas particularly in Taixi. Only few studies investigated $PM_{2.5}$-bound PBDEs in outdoor air (Table 3). Compared with $PM_{2.5}$-bound ΣPBDEs in the previous studies (Dong *et al.*, 2015; Liu *et al.*, 2016), our values in Taiwanese outdoor air were comparable to outdoor air levels in the metropolis areas in China, but still 60 to 100-fold higher than the background levels of atmospheric $PM_{2.5}$-bound PBDEs in East China Sea (Li *et al.*, 2015). Although outdoor $PM_{2.5}$-bound Σ_{12}PBDEs in the ambient air of Valencia, Spain were presented in extremely low magnitudes, the high molecular weights of PBDEs like octa-BDEs, nona-BDEs, and deca-BDEs were not analyzed in Beser's report (Beser *et al.*, 2014). PBDEs particle distribution in the atmosphere was found to be associated with fine particles of diameters smaller than 0.49 μm as reported by Besis *et al.* (2015) in traffic and urban background sampling sites located in Thessaloniki, northern Greece. In a ten-year-ago study (Deng *et al.*, 2007), outdoor mean Σ_{22}PBDEs in $PM_{2.5}$ concentrations from e-waste recycling sites (industrial) in Guiyu, Southeast China to that of the urban sites in Guangzhou and Hong Kong, South China were analysed wherein it showed that Guiyu has the highest atmospheric mean $PM_{2.5}$-bound PBDEs levels (16.6 ng m^{-3}) compared to the latter two (33.8–372 pg m^{-3}) and other urban sites from all over the world with an estimated exposure through inhalation of 133 and 332 ng day^{-1} for both adult and children. Although the surveillance of ambient air PBDEs (n = 180) were done in metropolis (Hong Kong and Guangzhou) and heavily contaminated industrial areas (Guiyu) (Deng *et al.*, 2007), we have concerns regarding with Deng's report (Deng *et al.*, 2007) due to two reasons as follows: (1) the HRGC/LRMS with EI mode was used to detect PBDE congeners. The resolution and sensitivity is not good enough to distinguish the neighbour peaks, and (2) high molecular weights of PBDEs from 196 to 209 were not measured in their study. Most PBDE congeners from octa-BDEs to deca-BDE are possible dominating congeners among total PBDEs in the outdoor air.

Many atmospheric studies investigated PBDEs in outdoor air. Harrad and Hunter (2006) found that PBDEs favour higher ratios in air as compared in the soil (47:99 ratio) and that its main source is the indoor environment. Rural sites in Ontario, Canada have been reported to have a mean Σ_{21}PBDEs in total air concentrations of range between 6 and 85 pg m^{-3} which are still lower as compared in urban sites (Gouin *et al.*, 2005). In addition, a study conducted in Izmir, Turkey on PBDE concentrations of outdoor and indoor organic films on window glasses showed that industrial sites have the highest PBDE concentrations compared to that of the offices, laboratories, and urban, suburban, and rural homes (Cetin and Odabasi, 2011). Syed *et al.* (2013) suggested that industrial activities played a key role in the PBDE distribution in areas located near industrial sites. Combustion process in Taiwanese industrial locations (Wang *et al.*, 2011) as well as dismantling and dumping of e-wastes (Deng *et al.*, 2007) are listed as reasons for PBDEs emission in industrial sites (O'Driscoll *et al.*, 2016).

As shown in Table 4, the indoor $PM_{2.5}$-bound Σ_{14}PBDEs mean concentrations for different indoor locations which include libraries, rail stations, department stores, offices, hospitals, and supermarkets are 357, 35.3, 50.2, 73.2, 59.2, and 124 pg m^{-3}, respectively. The highest Σ_{14}PBDE level in $PM_{2.5}$ was found in the indoor air of library. Many libraries have already been modernized and incorporated with printers and computers and many other electronic systems which may have contributed more to the indoor PBDEs level. A positive correlation ($p < 0.001$) was found between the number of electronic appliances and polyurethane foam

Table 2. PBDE concentrations on $PM_{2.5}$ particulate in outdoor aerosol from central and southern Taiwan (pg m^{-3}).

	Metropolis areas			Industrial areas		Rural areas	
	Xitun[1,3] (n = 1)	Douliu[1,3] (n = 1)	Tainan[1,3] (n = 1)	Mailiao[1,3] (n = 1)	Taixi[1,3] (n = 1)	Hengchun[2,3] (n = 1)	Meinong[2,3] (n = 1)
BDE-28	3.73	2.47	2.95	17.4	11.0	3.73	10.3
BDE-47	32.0	16.9	16.0	8.35	7.15	32.0	9.40
BDE-100	2.85	1.54	1.57	0.308	0.428	2.85	0.493
BDE-99	10.9	5.83	6.78	1.40	2.26	10.9	1.95
BDE-154	1.06	0.755	0.503	0.238	0.403	1.06	0.345
BDE-153	0.843	1.01	0.850	0.130	0.470	0.843	0.453
BDE-183	1.03	2.35	0.653	0.678	0.785	1.03	0.683
BDE-197	0.368	0.863	0.145	0.330	0.295	0.368	0.145
BDE-203	0.518	0.632	0.275	0.110	0.110	0.518	0.505
BDE-196	0.375	0.580	0.310	0.303	0.380	0.375	0.360
BDE-208	0.883	1.45	0.815	0.610	2.49	0.883	0.808
BDE-207	1.47	2.60	1.49	1.20	3.98	1.47	1.53
BDE-206	1.24	2.58	1.49	1.10	3.88	1.24	1.48
BDE-209	16.9	36.8	19.1	22.4	135	16.9	22.7
Σ_{14}PBDEs	74.3	76.3	53.0	54.5	169	74.3	51.0

[1] The sampling time was from May 5 to May 8 in 2013.
[2] The sampling time was from October 2 to October 5 in 2013.
[3] Four $PM_{2.5}$ samples from the continuously four-day sampling in the same TEPA air monitoring site were pooled together.

Table 3. Summary of the current data on $PM_{2.5}$-bound PBDE levels in outdoor air.

Location	Style	Mean or mean ± SD	PBDE congeners	Reference
Metropolis areas (China)[1]	Urban	35 ± 150 pg m^{-3}	Σ_9PBDEs: BDE-28, 35, 47, 99, 100, 153, 154, 183, and 209	Liu *et al.*, 2016
Beijing (China)	Industrial	146 pg m^{-3}	Σ_{42}PBDEs: BDE- 1, 2, 3, 7, 8, 10, 11, 12, 13, 15, 17, 25, 28, 30, 32, 33, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 116, 118, 119, 126, 138, 153, 154, 155, 166, 181, 183, 190, 205, 206, and 209	Dong *et al.*, 2015
Hong Kong (China)	Urban	88.3 ± 54.3 pg m^{-3}	Σ_{22} PBDEs: 3, 7, 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 236, 153, 154, 183, 184, and 191	Deng *et al.*, 2007
Guangzhou (China)	Urban	159 ± 81.0 pg m^{-3}		
Guiyu (China)	Industrial	16600 ± 13300 pg m^{-3}		
East China Sea (China)	Sea	0.97 ± 0.52 pg m^{-3}	Σ_{11}PBDEs: BDE-17, 28, 47, 66, 71, 85, 99, 100, 153, 154, 183 and 209	Li *et al.*, 2015b
Region of Valencia Government (Spain)	Residential	0.213 pg m^{-3}	Σ_{12}PBDEs: 28, 47, 49, 66, 99, 100,119, 139, 153, 154, 155, and183	Beser *et al.*, 2014
Central and southern Taiwan (Taiwan)	Urban	67.9 ± 12.9 pg m^{-3}	Σ_{14}PBDEs: 28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, and 209	The present study
	Industrial	112 pg m^{-3}		
	Rural	62.7 pg m^{-3}		

[1] The metropolis areas in China including Beijing, Shanghai, Guangzhou, Nanjing, Wuhan, Taiyuan, Chengdu, Lanzhou, Guiyang, and Xinxiang.

in chairs against PBDE levels in the indoor environment (Harrad *et al.*, 2004). It has been understood that PBDEs can migrate from the disposed plastic parts of electronic equipment to indoor dust and that the printed wiring board (PWB) which is found in most of electronic appliances emit additional PBDEs when heated (Guo *et al.*, 2015b, c; Anh *et al.*, 2016). The second highest $PM_{2.5}$-bound Σ_{14}PBDEs mean concentration is found in supermarkets. Supermarkets highly merchandise electronic devices and appliances coated with BFRs which may have contributed to its relatively high $PM_{2.5}$-bound Σ_{14}PBDEs mean concentrations. Due to the high air exchange rate and efficacy of ventilation, the rail station result appeared to be the lowest level of $PM_{2.5}$-bound Σ_{14}PBDEs among the public environments. Different workplaces have different levels and composition concentrations of PBDEs in the present study. The Greek study revealed that mean levels of PBDEs in offices, internet cafes/computer rooms, and computers/electronic shops were significantly higher compared with those in furniture stores, homes, and outdoor air (Mandalakis *et al.*, 2008). In Table 5, levels of $PM_{2.5}$-bound ΣPBDEs were shown in kindergartens, household, workplaces, and recycle plant. Our Σ_{14}PBDE levels in the various indoor environments had the similar magnitudes compared with those in household

Table 4. Concentrations of PBDEs on $PM_{2.5}$ fine particulate in the indoor aerosol from various environments (pg m^{-3}).

	Library[1] (n = 1)	Rail Station[1] (n = 1)	Department Store[1] (n = 1)	Office[1] (n = 1)	Hospital[1] (n = 2)	Supermarket[1] (n = 2)
BDE-28	0.0795	0.0795	0.0797	0.0797	0.211	0.340
BDE-47	< LOD	< LOD	1.71	3.52	1.71	2.86
BDE-100	0.0945	0.0945	0.0943	3.60	0.245	0.525
BDE-99	0.768	0.768	0.768	14.9	0.768	0.768
BDE-154	0.0886	0.0886	0.472	3.68	0.204	0.644
BDE-153	0.260	0.260	1.75	3.35	0.260	1.70
BDE-183	0.523	0.523	4.42	3.05	1.14	7.63
BDE-197	0.292	0.292	1.65	0.292	0.600	4.80
BDE-203	1.20	0.888	1.73	0.756	1.37	7.02
BDE-196	1.04	0.807	1.84	0.663	1.08	5.26
BDE-208	9.52	0.581	0.581	0.581	1.21	4.84
BDE-207	14.1	0.957	3.00	3.01	1.73	8.89
BDE-206	15.6	0.651	2.75	2.56	2.34	8.49
BDE-209	314	29.7	29.3	33.2	48.3	69.8
Σ_{14}PBDEs	357	35.3	50.2	73.2	59.2	124

[1] Three $PM_{2.5}$ samples from the same style of the indoor environment were pooled together.

Table 5. Summary of the current data on $PM_{2.5}$-bound PBDE levels in indoor air.

Location	Style	Mean ± SD or Range	PBDE congeners	Reference
Hong Kong (China)	Kindergartens	0.453 ± 0.345 ng m^{-3}	Σ_{13}PBDE: BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, and 190	Deng *et al.*, 2016
Shanghai (China)[1]	Households Workplaces[2]	0.0485 ± 0.0172 ng m^{-3} 0.105 ± 0.0455 ng m^{-3}	Σ_{18}PBDEs: BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 203, 206, 207, 208, and 209	Xu *et al.*, 2016
Guangzhou (China)	Households	0.239 ng m^{-3}	Σ_{26}PBDEs: BDE-3, 7, 15, 17, 28, 47, 49, 66, 71, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207, and 209	Wang *et al.*, 2014
Hong Kong (China)	Households	0.0438 ng m^{-3}		
Shanghai (China)[3]	Waste TV Recycling plant	9.35–6670 μg g^{-1}	Σ_{12}PBDEs: BDE-28, 47, 99, 100, 153, 154, 183, 203, 206, 207, 208, and 209	Guo *et al.*, 2015c
Central and southern Taiwan (Taiwan)	Workplaces[4]	0.116 ± 0.112 ng m^{-3}	Σ_{14}PBDEs: 28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, and 20	The present study

[1] The composition concentrations of $PM_{2.5}$-bound PBDEs were 2519 ± 790 ng g^{-1} (mean ± SD) and 2345 ± 665 ng g^{-1} in household and workplaces, respectively.
[2] Xu *et al.* (2016) did not shewed the types of workplaces in Shanghai City.
[3] The composition concentrations $PM_{2.5}$ in different sampling sites in the waste TV recycle plant were 9.35 μg g^{-1} in warehouse, 476 μg g^{-1} in TV dismantling, 3740 μg g^{-1} in printer wiring board (PWB) heating, 32.6 μg g^{-1} in PWB recycling, and 6670 μg g^{-1} in plastic crushing.
[4] The workplaces in the present study were library, rail station, department store, office, hospital, and supermarket.

and workplace from Xu's and Wang's reports (Wang *et al.*, 2014; Xu *et al.*, 2016) ($p > 0.05$). The extremely high levels of $PM_{2.5}$-bound ΣPBDEs were found in a waste TV recycle plant particularly for the highly PBDE-contaminated treatment process like heating PWB and plastic crushing (Guo *et al.*, 2015b). Mean level range of indoor $PM_{2.5}$-bound PBDEs in five Hong Kong kindergartens (0.10–0.64 ng m^{-3} or 100–640 pg m^{-3}) (Deng *et al.*, 2016) were still relatively higher than that of the indoor locations except for library (357 pg m^{-3}) and supermarket (124 pg m^{-3}) in the present study. Based on the raw data of PBDEs from five kindergartens in Deng's study (Deng *et al.*, 2016), their data might be not accepted due to most values of hexa-BDEs and hepta-BDEs below LODs.

The distributions of outdoor and indoor PBDE homologues (di-BDE, tri-BDE, tetra-BDE, pentaBDE, hexa-BDE, hepta-BDE, octa-BDE, nona-BDE, and deca-BDE) are shown in Fig. 1. Deca-BDE has the highest distribution levels in both the indoor and outdoor locations at 74.7% and 48.03%. Nona-BDE (BDE-208, -207, and -206) levels (11.6%) is significantly high in the indoor locations while tri- and tert-BDE (11.3% and 16%) in the outdoor locations. Many studies have already reported that deca-BDE (BDE-209) is predominant in both the indoor and outdoor environments (Stapleton *et al.*, 2005; Gevao *et al.*, 2006; Chen *et al.*, 2008; Gou *et al.*, 2016a). But still, BDE-209 is higher in

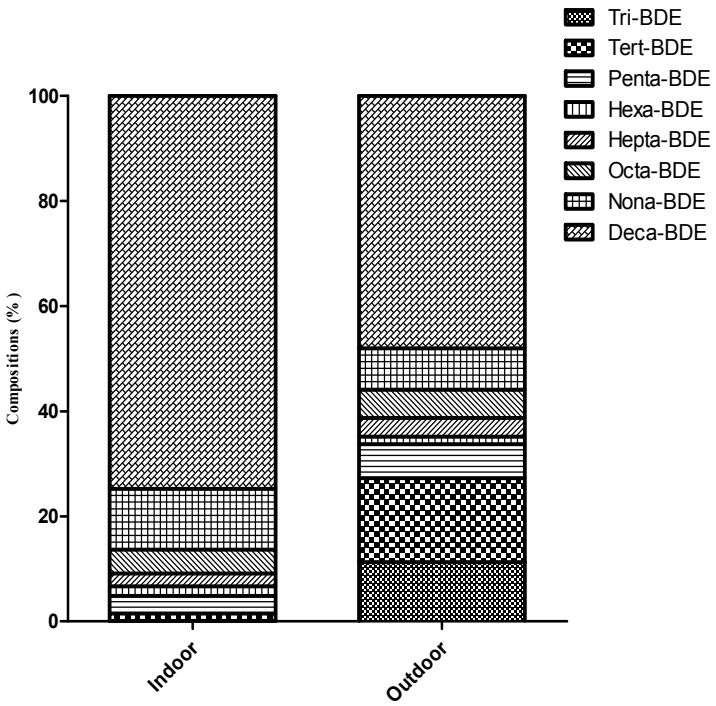

Fig. 1. The composition of PBDEs on the fine particulate ($PM_{2.5}$) in the indoor and outdoor aerosol.

the indoor environment as compared to that in the outdoor environment. Similarly for indoor environments, BDE-209 was found to be predominant in air in Taiwanese residential homes among all 14 PBDE congeners as reported by Shy *et al.* (2015). The opposite is true in Korean elementary schools where the predominating congener was BDE-47 (Lim *et al.*, 2014). BDE-209 in the outdoor is mainly attributed to indoor BDE-209 (Gou *et al.*, 2016a). A high percentage of nona-BDE was also observed in a study conducted on e-waste workshops in China in which debromination of deca-BDE to nona-BDEs through exposure to natural sunlight was inferred to be the possible cause (Xu *et al.*, 2015). However, with the lower vapor pressures of nona- and deca-BDEs, they are mostly particle-bound which makes them harder to degrade photochemically (Raff and Hites, 2007). Various equipment containing PBDEs found in the indoor environment are main sources of PBDE emission into the air via volatilization (Tung *et al.*, 2014). The existence of these higher brominated PBDEs in the indoor air is probably attributed to their migration behaviours from outdoors to indoors. Although the indoor air is a significant source of PBDEs emission into the outdoor environment, the individual sources for each indoor and outdoor PBDEs might be different. A high percentage of tri- and tetra-BDEs are accounted in the outdoor locations. An opposite trend was observed in the study conducted by Gou *et al.* (2016a) wherein the tri-BDE is higher in the indoor than in the outdoor. Ding *et al.* (2016) reported higher BDE-28 levels in the indoor air than BDE-47 while the outdoor air showed higher BDE-47 levels compared to BDE-28. Possibly, the presence of BDE-28 and BDE-47 in the indoor and outdoor air is attributed to photochemical degradation by natural sunlight and migration behaviours from indoors to outdoors of the congeners brought about by their low vapor pressure characteristics which makes them easily evaporate from products (Ding *et al.*, 2016).

CONCLUSIONS

The industrial locations were found to have the highest outdoor $PM_{2.5}$-bound PBDEs levels particularly in Taixi located at the industrial area. This is affected mainly by industrial activities such as combustion or petrochemical production around the area and also the migration of indoor $PM_{2.5}$-bound PBDEs into the outdoor environment. Our study found the highest indoor $PM_{2.5}$-bound PBDEs in the libraries, mainly, due to low air exchange rate, low efficiency of ventilation, the volatilization of PBDEs from the surface of electronic equipment (e.g., computer and printers) which are already common in the present day libraries. Deca-BDE and nona-BDEs predominated in the indoor locations. This is primarily attributed to their dispersion from the indoor equipment and migration behaviors from the outdoor environment. Tri-, tetra-, and deca-BDE levels were high in the outdoor areas. Photochemical degradation or debromination is also a possible source of lower brominated PBDEs in both the indoor and outdoor environments. Migration behaviors from the indoor to the outdoor air and vice versa of the $PM_{2.5}$-bound PBDEs might affect the indoor and outdoor $PM_{2.5}$-bound PBDEs level**s.**

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DISCLAIMER

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

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