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Levels of Polybrominated Diphenyl Ethers in Air-Conditioner Filter Dust Used to Assess Health Risks in Clinic and Electronic Plant Employees

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ABSTRACT

Because consumer products release them, polybrominated diphenyl ethers (PBDEs) are frequently present in a variety of indoor environments including homes and workplaces. Our goal was to investigate PBDEs contamination of workplace air-conditioner filter dust to assess the health effects of contaminated dust on workers with occupational exposure. Nine medical clinics, four dental clinics, eight factory offices, and six factory clean rooms were selected in southern Taiwan between April 2013 and September 2014. Air-conditioner-filter dust was collected by a high-efficiency vacuum cleaner and then PBDEs were analyzed by a high-resolution gas chromatography coupled with high-resolution mass spectrometry. The Σ_{14}PBDEs (the sum of BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, and 209) in independent-air-conditioner filter (IAF) dust was not significantly lower in dental clinics (736 ng g^{-1}) than in medical clinics (1600 ng g^{-1}) and electronic plant offices (2570 ng g^{-1}). PBDEs level was distinctly higher by an order of magnitude in central-air-conditioner system filter (CASF) dust in clean rooms (32,600 ng g^{-1}), than in IAF dust. In clinic and the office workers, PBDEs daily intake via indoor dust ingestion varied from 2.96×10^{-8} to 1.25×10^{-7} mg kg b.w.$^{-1}$ day^{-1} (29.6–125 ng kg b.w.$^{-1}$ day^{-1}), which was obviously below the lowest observed adverse effect of level (LOAEL) of 1 mg kg b.w.$^{-1}$ day^{-1}. Assessment of the risk of non-cancer diseases with neurobehavioral effects and of cancer with neurobehavioral effects in clinic and office workers was notably below threshold values (non-cancer: 1.00 and cancer: 1.00×10^{-6}). In conclusion, clinic and office workers had no harmful effects in the currently existing levels of indoor dust PBDEs in workplaces. IAF or CSAF dust can possibly reflect spatial and temporal dust distribution in the microenvironment. It is also suggested that collection of air-conditioner filter dust may be an alternative method to conventional dust sampling for assessment of indoor contamination by PBDEs.

Keywords: Polybrominated diphenyl ethers; Air-conditioner filter; Indoor dust; Clinics; Offices; Health risk.

INTRODUCTION

Developed in the 20th century, the brominated fire retardants (BFRs) delay ignition and slow the spread of fire. Polybrominated diphenyl ethers (PBDEs) are a class of BFRs and man-made non-naturally occurring chemicals

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utilized extensively as additives in a wide variety of consumer products, such as electronic devices, vehicles, building materials, airplanes, ships, and furniture foam and upholstery. Owing to their environmental persistence and accumulation in adipose tissues, PBDEs are ubiquitous in the atmospheric environment (Chao *et al.*, 2014a; Cincinelli *et al.*, 2014; Zhou *et al.*, 2014; Shy *et al.*, 2015), in soils, sediments, and plants (Wang *et al.*, 2015), water and fishes (Chokwe *et al.*, 2015), and marine and terrestrial mammals (Leonel *et al.*, 2014; Vetter *et al.*, 2015). The animal studies showed that PBDEs disrupted thyroid function and spermatogenesis (Bowers *et al.*, 2015; Huang *et al.*, 2015), impaired mitochondrial function (Huang *et al.*, 2015), induced hepatotoxicity (Ćurčić *et al.*, 2015), caused developmental abnormalities (Berger *et al.*, 2014), affects neurological behaviour (Costa and Giordano, 2011). They tend to accumulate in human bodies and have been detected in human milk and umbilical cord blood (Wang *et al.*, 2008; Lin *et al.*, 2011; Koh *et al.*, 2010). Human with prenatal or postnatal exposure to PBDEs has been demonstrated to be associated with several adverse health outcomes (Chao *et al.*, 2014b; Hsu *et al.*, 2014) including low birth length and weight (Chao *et al.*, 2007), menstrual irregularity (Chao *et al.*, 2010), disruption of thyroid and growth hormones (Shy *et al.*, 2012), and interference with neurological development in infants (Chao *et al.*, 2011). In the recent study in the e-waste dismantling areas, the researchers found that serum levels of PBDEs were related with increasing levels of white blood cells, hemoglobin, and platelets (Xu *et al.*, 2015a).

Although pentaBDEs and octaBDEs formulations have not been globally manufactured since 2004 and commercial decaBDE mixtures have been banned in Europe and voluntarily phased out in the USA since 2012, decaBDE is still produced and widely used in most countries outside of Europe and America. It has been demonstrated that levels of BFRs including PBDEs are much lower in outdoor than indoor air and that the source of BFRs outdoors may be indoor air (Newton *et al.*, 2015). Humans tend to spend more time indoors than outdoors, so direct PBDEs exposure is a public health concern. PentaBDEs and octaBDEs, though phased out of production, continue to be released into the indoor environment or microenvironments from PBDE-containing consumer goods manufactured before the ban and still in service. Exposure to PBDEs occurs in the indoor environment through several routes including ingestion, inhalation, and dermal contact. Much attention has been paid to indoor dust, which is a major sink for PBDEs and an important pathway for human exposure to PBDEs (Chao *et al.*, 2014c; Canbaz *et al.*, 2015; Newton *et al.*, 2015). Furthermore, several reports indicate that most PBDEs exposures are from indoor dust and not food (Lorber, 2008; Johnson-Restrepo and Kannan, 2009; Kang *et al.*, 2011). A recent study failed to find a significant relationship between PBDEs in mattress dust (indoor dust) and development of childhood asthma (Canbaz *et al.*, 2015).

Information on the use, fate, concentrations, and distribution of workplace PBDEs in Taiwan is limited. Recent studies reported high levels of PBDEs in dust from offices, retail stores, libraries, and other workplaces (Harrad *et al.*, 2008; Batterman *et al.*, 2010; Watkins *et al.*, 2013; Li *et al.*, 2015; Xu *et al.*, 2015b). Not much is known on accumulation of PBDEs in air-conditioner filters even through central air-conditioning systems and independent air conditioners have been widely used in many indoor environments such as offices, schools, laboratories, commercial buildings, and retail stores (Ni *et al.*, 2011; Besis *et al.*, 2014; Xu *et al.*, 2015b; Yu *et al.*, 2013). To my knowledge, few studies have investigated PBDEs accumulation in air-conditioner filters in indoor workplaces (e.g., clinics and electronics factories) and thereby assessed health risks to exposed medical staff and factory employees.

METHODS

Sampling Location

The sampling areas were in southern Taiwan and included Tainan, Kaohsiung, and Pingtung. In the beginning, we contacted with local medical and dental associations in Kaohsiung City and Pingting County to obtain the inventories of the clinics. There are 2221 medical clinics (1581 clinics in Kaohsiung and 640 clinics in Pingtung) and 1006 dental clinics (859 clinics in Kaohsiung and 147 clinics in Pingtung), respectively, in the sampling areas. We selected the candidate lists from the inventories based on our study design. The associations noticed the clinics to let them know our study's aim. Of the 20 candidate medical clinics and 20 candidate dental clinics randomly selected from the local medical and dental association listings, respectively, in Kaohsiung City and Pingtung County, and contacted by research team members, only 9 medical clinics and 4 dental clinics agreed to participate our study. Between April 2013 and September 2013, indoor environments were checked and samples of dust from air filters in independent air-conditioning systems (IAF dust) were collected.

In the begin of 2014, our team members visited at three facilities that manufacture color filters in southern Taiwan (including one in Lujhu [Kaohsiung City] and two in Xinshi [Tainan City]) to discuss the sampling process with managers or engineers according to our study design. Finally, 8 offices and 6 clean rooms were selected based on plant manager recommendations, the indoor environment, our sampling strategy, and our project budget. IAF dust from the offices and central air-conditioner system filter (CASF) dust from the clean rooms were collected from March 2014 to September 2014.

Reagents and Chemicals

Fourteen native PBDE standards, including 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). The internal standards, ^{13}C-labeled PBDEs (BDE-28, -47, -99, -153, -183, -197, -207, and -209), were from Wellington Laboratories (Guelph, Canada). Residue-grade chemicals and HPLC-grade solvents such as toluene, n-hexane, silica gel, potassium oxalate, alumina oxide, and sodium sulfate were obtained from Tedia, Inc. (Fairfield, OH, USA), Sigma-Aldrich (St. Louis, MO, USA), and Merck (Darmstadt, Germany).

Sampling Collection and Analysis

Before sampling, the researchers assessed the indoor environment and conducted a questionnaire survey. The questionnaire elicited the age of building, air conditioner type, the time of the last redecoration, frequency and times of filter cleaning, number of computers and printers, number of product consumers, number of workplace staff or employers, and daily work hours. New workplaces were defined as new buildings or premises redecorated within one year. IAF and CASF dust were sampled using a high efficiency particulate air filter (HEPA filter) vacuum cleaner (Nilfisk Advance Euroclean UZ934 HEPA canister vacuum cleaner) and a strict protocol as described previously (Chao *et al.*, 2014c). Cross contamination of the samples was prevented by washing the vacuum cleaner thoroughly with deionized distilled water and soapy water and wiping the vacuum cleaner with n-hexane-impregnated disposable wipes (Ali *et al.*, 2011). IAF or CASF dust was collected from each site two or three times within a six-month period, depending on dust weight. After collection, each sample was immediately transported to the laboratory at National Pingtung University of Science and Technology, washed through a 50-mesh filter with shaking for 10 minutes, homogenized, stored in a chemically clean glass bottle with a Teflon cap at –20°C until analysis, and sent to Super Micro Research and Technology Center, Mass Cheng Shiu University in Kaohsiung, Taiwan to determine PBDE concentrations.

The analytical PBDE method in the present study was modified from that used in previous studies (Chao *et al.*, 2014c; Wu *et al.*, 2014). Samples (1–3 grams of dust) were spiked with $^{13}C_{12}$-labeled PBDE internal standards prior to extraction by a Soxhlet extractor with 200-ml toluene for at least 16 hours. The extracted solution was concentrated to dryness in a rotary evaporator. For the cleanup procedure, the extract was treated with concentrated sulfuric acid, and passed through a multi-layered silica column and an acid alumina column. The eluate was placed in a vial and evaporated to near dryness using a gentle stream of gaseous nitrogen. The final volume of the eluate prior to injection was 0.2 mL. Fourteen PBDE levels were examined and determined by a high resolution gas chromatography (Hewlett-Packard 6970, H-P Inc., Palo Alto, CA) and using a high resolution mass spectrometry system (Micromass Autospec Ultima, Micromass, Beverly, MA) equipped 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 with constant helium flow of 1 mL min^{-1} at 280°C. The temperature programs of GC oven temperature was held at 100°C for 4 min to increase from 100°C to 200°C at a rate of 40 °C min^{-1}, to stay at 200°C for 3.5 min, to change to 325°C by 10 °C min^{-1}, 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. A field blank was considered during each sampling procedure to evaluate PBDEs contamination in the field. Laboratory blanks were evaluated for each batch of 8–10 samples during each run. The limits of detection (LODs) and quantification (LOQs) were defined as a signal-to-noise (S/N) ratio of higher than 3 and 10, respectively, for each chromatographic peak. For 14 PBDE congeners, the LOD of BDE-209 was 297 pg g^{-1} and LODs for the remaining PBDE congeners were between 0.218 and 42.0 pg g^{-1}. The mean recoveries of PBDE from internal and labelled cleanup standards were within acceptable limits (70–130%). The data in the chemical assay were not blank corrected in this study.

Health Assessment in the Workplaces

The data from medical staff in the clinics and employees in the electronics plants with occupational exposure to PBDEs via IAF or CASF dust ingestion were obtained to calculate daily intake (DI_{dust}), non-cancer risk (Hazard quotient, HQ), and cancer risk (R). Non-dietary PBDE daily workplace intake was assessed using the following equation from Besis *et al.* (2014):

$$DI_{dust}\,(\text{ng kg b.w.}^{-1}\ \text{day}^{-1}) = (C_{\text{indoor dust}} \times IEF_{\text{indoor exposure fraction}} \times IR_{\text{indoor dust}} \times AB_{\text{absorption rate}})/(BW_{\text{body weight}}) \quad (1)$$

where, $C_{\text{indoor dust}}$ is Σ_{14}PBDEs in IAF or CASF dust from the workplaces. $EF_{\text{indoor exposure fraction}}$ is defined as the percentage of time spent in the workplace each day. $IR_{\text{indoor dust}}$ and $AB_{\text{absorption rate}}$ are the respective average rates of indoor dust ingestion and PBDEs absorption (via indoor dust) in the human intestinal tract. $BW_{\text{body weight}}$ is average body weight. The $IEF_{\text{indoor exposure fraction}}$ for employees in the clinics and electronic plants was defined as 0.3333, which corresponds to an 8-hour work day. Adults ingest 30 mg g^{-1} of indoor dust ($IR_{\text{indoor dust}}$) according to the "Child-specific Exposure Factors Handbook" (US EPA, 2008). The $AB_{\text{absorption rate}}$ was 0.508 for tri- to nona-BDEs and 0.139 for BDE-209 calculated from estimates of accessibility to PBDEs through air-conditioning filters (Yu *et al.*, 2013). $BW_{\text{body weight}}$ was 70 kg for male adults and 56 kg for female adults, respectively, in Taiwan, according to the Nutrition and Health Survey in Taiwan (NAHSIT) conducted from 2005 to 2008 by the Health Promotion Administration of the Ministry of Health and Welfare (MOHW) (National Health Research Institute, 2014).

The risk of non-cancer diseases with neurobehavioral effects and of cancer due to intake via indoor dust were also evaluated in the present study as the follows.

$$\text{Chronic daily intake (CDI)} = (DI_{dust} \times EF \times ED)/(AT \times 365) \quad (2)$$

$$HQ = CDI/RfD;$$
$$R = CDI \times SF_{\text{slope factor.}}$$

Human risk assessment was calculated using a modification of the equation from Lim *et al.* (2014) and Li *et al.* (2015a), where EF is exposure frequency per year (days year^{-1}), which is defined as 220 days year^{-1} in this study, ED is exposure duration (years), which is assumed to be 40 years, and AT is average lifespan in Taiwan (76.43 and 82.82 years for men and women, respectively estimated from [NAHSIT] data) (National Health Research

Institute, 2014). Oral reference doses for chronic exposure (RfDs) to BDE-47, -99, -153, and -209 (0.0001, 0.0001, 0.0002, and 0.007 mg kg $^{-1}$ day^{-1}, respectively) were provided by the US EPA Integrated Risk Information System (IRIS) for non-cancer diseases with neurobehavioral effects (US EPA, 2008). The cancer slope factor (SF $_{\text{slope factor}}$) for oral BDE-209 is 0.0007 per mg kg $^{-1}$ day^{-1} (US EPA, 2008).

Statistical Analysis

Measurements of PBDEs below LODs were set at zero. A Monte Carlo simulation was used to assess human health risks in the workplace. Nonparametric statistical analyses including Spearman's rho correlation coefficient, Mann-Whitney *U*, and Kruskal Wallis *H* tests were performed because indoor dust levels of PBDEs in various indoor environments are not normally distributed. Statistical analysis was carried out using the Statistical Product and Service Solutions (SPSS) software (version 12.0).

RESULTS AND DISCUSSION

Indoor Dust PBDE Concentrations in Clinics and Electronic Plants

Table 1 shows concentrations of PBDEs in IAF or CASF dust from medical and dental clinics and from offices and clean rooms in electronic color filter manufacturing facilities. Mean levels of Σ_{14}PBDEs in IAF or CASF dust were 1600 (median [Md]: 508, standard deviation [SD]: 2230), 736 (Md: 826, SD: 360), 2590 (Md: 2710, SD: 1530), and 33,600 (Md: 25,900, SD: 36,500) ng g^{-1} in the medical (n = 13) clinics, dental (n = 4) clinics, factory offices (n = 8), and factory clean rooms (n = 6), respectively. IAF dust levels of PBDEs in the clinics and offices were lower than floor dust and settled dust levels (i.e., electronic dust) in Taiwanese houses (Chao *et al.*, 2014c). The environments listed (in order from low to higher average level of Σ_{14}PBDEs in air-conditioner filter dust) were dental clinics < medical clinics < offices < clean rooms. The extremely high levels of PBDEs in CASF dust from clean rooms were surprising, though comparable to levels in indoor dust from e-waste recycling areas (38,685 ng g^{-1}) in China (Jiang *et al.*, 2014).

BDE-209 was the predominant congener in IAF or CASF dust collected from these four kinds of workplaces. BDE-209 in IAF or CASF dust was 53.4%, 61.3%, 64.1% and 29.6% of the indoor Σ_{14}PBDEs in dust from medical clinics, dental clinics, offices, and clean rooms, respectively. PBDE patterns in IAF dust from the clinics and offices were similar (Figs. 1(A)–1(C) and Figs. 2(A) and 2(B)). Conversely, nona-BDEs (the sum of BDE-206, -207, and -208) and octa-BDEs (BDE-196, 197, and 203) accounted for only 33.0% and 31.4% of the total in CASF dust collected from clean rooms (Fig. 1(D) and Fig. 2(B)). Levels of tri- to nona-BDEs and Σ_{14}PBDEs in IAF dust were lower in the new medical clinics than in the dental and old medical clinics (Fig. 2(A)). Before IAF dust sampling began, we anticipated that PBDEs contamination in IAF dust from the new medical clinics would exceed that from old medical clinics, but the results showed the opposite. The unrelatedness of heavier PBDEs contamination of IAF dust

Table 1. PBDE concentrations in air-conditioner filter dust from medical clinics, dental clinics, factory offices, and factory clean rooms (ng g^{-1}).

PBDEs	Medical clinics[a]	Dental clinics[a]	Electronic color filter manufacturing facilities: Offices[a]	Electronic color filter manufacturing facilities: Clean rooms[b]
	Mean ± SD (range)	Mean ± SD (range)	Mean ± SD (range)	Mean ± SD (range)
BDE–28	0.660 ± 1.01 (0.10–3.29)	0.500 ± 0.300 (0.090–0.800)	1.32 ± 2.45 (0.146–7.33)	1.05 ± 1.10 (0.18–3.01)
BDE–47	20.0 ± 40.3 (2.55–127)	9.44 ± 9.87 (1.92–23.4)	40.4 ± 81.7 (2.25–242)	7.32 ± 6.54 (1.73–19.8)
BDE–99	45.1 ± 113 (3.45–347)	11.9 ± 11.1 (3.33–27.1)	40.3 ± 61.3 (2.93–179)	15.4 ± 15.4 (2.48–18.0)
BDE–100	12.6 ± 31.4 (1.11–96.1)	2.56 ± 2.06 (0.610–5.10)	4.31 ± 6.79 (0.660–20.9)	2.56 ± 3.24 (0.24–8.89)
BDE–153	46.1 ± 113 (2.31–345)	7.31 ± 3.42 (2.59–10.8)	18.1 ± 11.2 (5.14–32.9)	92.2 ± 138 (4.27–362)
BDE–154	10.4 ± 25.1 (0.900–77.2)	2.26 ± 1.08 (1.00–3.35)	7.85 ± 5.30 (2.69–16.7)	37.8 ± 66.5 (2.34–172)
BDE–183	170 ± 359 (5.76–1070)	20.1 ± 17.0 (3.87–40.4)	33.6 ± 17.8 (21.4–64.6)	1840 ± 2840 (23.6–6570)
BDE–196	61.8 ± 133 (3.24–409)	25.0 ± 36.1 (1.15–78.7)	52.6 ± 35.4 (22.6–136)	4240 ± 7610 (44.7–19500)
BDE–197	104 ± 238 (2.21–724)	11.9 ± 10.5 (0.980–23.9)	18.3 ± 10.3 (6.60–39.5)	1910 ± 3440 (23.4–8750)
BDE–203	35.9 ± 70.0 (1.93–216)	25.6 ± 44.0 (1.72–91.6)	51.8 ± 28.4 (25.7–98.8)	4400 ± 7950 (29.5–20300)
BDE–206	91.4 ± 103 (21.0–324)	82.3 ± 59.4 (13.6–144)	368 ± 388 (70.5–1300)	4870 ± 5320 (232–13600)
BDE–207	119 ± 203 (16.2–635)	52.1 ± 30.3 (9.40–76.1)	204 ± 167 (50.3–600)	4530 ± 5750 (152–14800)
BDE–208	29.5 ± 24.5 (9.27–84.3)	34.1 ± 25.4 (6.39–65.6)	84.2 ± 79.0 (16.2–272)	1690 ± 2170 (45.9–5440)
BDE–209	855 ± 971 (143–2320)	451 ± 192 (176–600)	1662 ± 937 (252–3030)	9930 ± 7390 (954–201000)
Σ_{14}PBDE	1600± 2230 (268–6620)	736 ± 360 (224–1070)	2570 ± 1530 (486–5590)	32600 ± 36200 (1760–101000)

[a] Independent air-conditioning unit filter dust (IAF dust); [b] Central air-conditioning system filter dust (CASF dust).

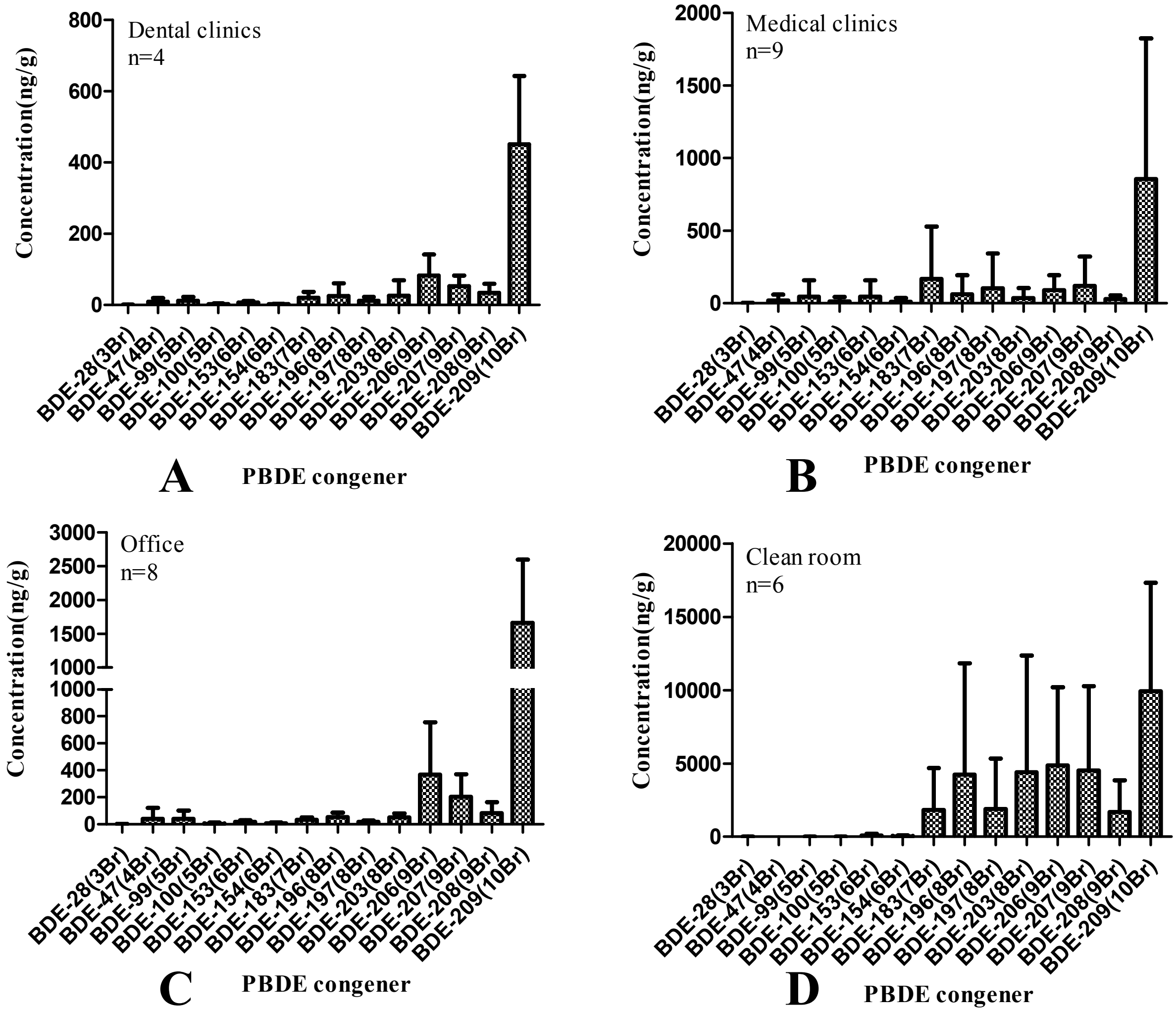

Fig. 1. Concentrations and patterns of PBDEs in the air-conditioner filter dust from workplaces including (A) Dental clinics, (B) Medical clinics, and (C) Offices, and (D) Clean rooms in electronic color filter manufacturing plants.

to new indoor environments led us to conclude that the bulk of PBDEs are probably not released from consumer products and construction materials used for redecoration or building. Ni *et al.* (2011) also found that levels of indoor dust PBDEs were lower in newer buildings. In the present study, despite the involuntary phase-out of commercial pentaBDEs and octaBDEs since 2004 in Taiwan, low levels of brominated PBDEs from tetra to hepta were still present in indoor workplaces. It is believed that pentaBDEs and octaBDEs are continually present on the surface of furniture, consumer electronics, carpets, textiles, and building materials in indoor workplaces in Taiwan. In an American study (Batterman *et al.*, 2010), BDE-47, -71, and -99 were detected in most indoor dust, suggesting that currently on the market consumer products or household appliances were still significant sources of PBDEs despite the 2004 phase-out of pentaBDEs and octaBDEs in commercial products.

Indoor PBDE concentrations may be influenced by several environmental factors including ventilation, room temperature, building materials, and numbers and types of consumer electronics (de Wit *et al.*, 2012). In the present study, PBDE levels in IAF and CASF dust were not correlated with numbers of computers or other electronic devices in the workplace (data not shown). Conversely, a recent South African study revealed an association of house dust BDE-47 with household mattresses and foam-containing furniture and an association of indoor dust BDE-209 with electronic appliances in both homes and offices (Abafe and Martincigh, 2014). Although not correlated with the number of electrical appliances or computers in office microenvironments, PBDE levels in office dust were significantly positively correlated with the rate of office power usage (Li *et al.*, 2015). In addition, indoor dust PBDE concentrations may be associated with human activity and socioeconomic development. Zhu *et al.* (2015) showed evidence for the possible relationship for indoor dust PBDEs to higher population density in urban areas or metropolitan cities.

Current Data on PBDEs in IAF or CASF Dust Worldwide

The current global data on PBDEs in IAF or CASF dust are presented in Table 2. Levels of BDE-209 and ΣPBDEs in IAF dust from Taiwanese clinics had a similar order of

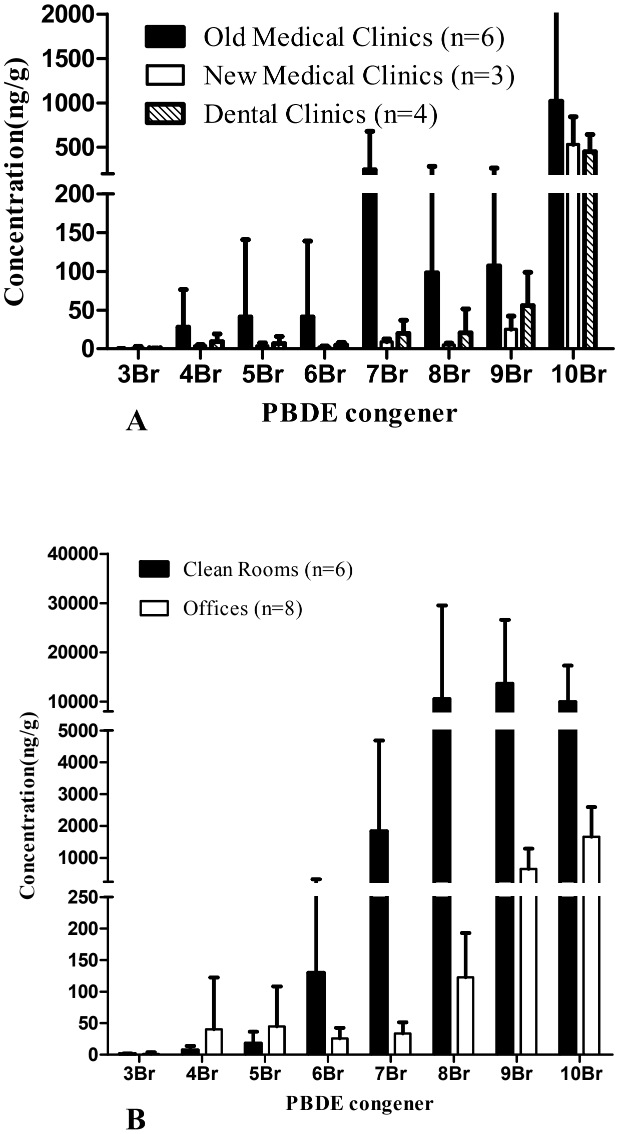

Fig. 2. PBDE homologue concentrations in the air-conditioner filter dust from: (A) Old and new medical clinics and dental clinics and (B) Offices and clean rooms in electronic color filter manufacturing facilities.

magnitude as levels in CASF dust from Chinese offices (Ni *et al.*, 2011) and Greek workplaces (Besis *et al.*, 2014). Mean levels of BDE-209 were notably lower in IAF dust from clinics and offices in southern Taiwan than that from a university in the Philippines (Fulong and Espino, 2013). Levels of BDE-209 and ΣPBDEs in CASF dust were extremely high in retail stores located in the United States (Xu *et al.*, 2015b) and in clean rooms in Taiwan (the present study). The most abundant BDE congener was BDE-209 in our study as well as other studies listed in Table 2 (Ni *et al.*, 2011; Besis *et al.*, 2014; Xu *et al.*, 2015b). In Table 2, higher brominated PBDEs, such as nona-BDEs and deca-BDE, accounted for 64.4–89.4% of ΣPBDEs in the present study (medical clinics: 68.4%, dental clinics: 84.2%, offices: 89.4%, and clean rooms: 64.4%), 93.9% of ΣPBDEs in the Greek study (Besis *et al.*, 2014), and 81.1% of ΣPBDEs in the Chinese study (Ni *et al.*, 2011). The possible reasons for the high percentage of higher brominated PBDEs in IAF or CASF dust are the presence of commercial decaBDE mixtures in consumer products currently used in indoor workplaces and the probable debromination of BDE-209 to form tetra- to nona-PBDEs.

Risk Assessment of PBDEs through Indoor Dust in Workplaces

The workers with occupational exposure to PBDEs via indoor dust were assessed (Table 3) to determine non-dietary daily intake, non-cancer risk, and cancer risk in clinics and offices. The workers in the selected electronic color filter manufacturing facilities are required to wear masks and protective clothing before entrance of clean rooms. Health risk to workers from ingestion of indoor dust in clean rooms was not examined in the present study mainly due to uncertainties in the effect on exposure of protective masks, protective clothing, and route of exposure. Evaluation of health assessment via indoor particulates for the workers in the clean rooms faced the lot of difficulties and uncertainties in the present study. Firstly, the penetration rate of PBDEs on the indoor particulates from the masks or protective clothing is unknown. Secondly, the exposure route through indoor particulates in the positive environment can not be reviewed based on the current literatures. The magnitude of non-dietary ingestion of Σ_{14}PBDEs DI_{dust} in the medical and dental clinics was 7.12×10^{-8} and 2.96×10^{-8} mg kg b.w.$^{-1}$ day^{-1} for males and 8.89×10^{-8} and 3.71×10^{-8} mg kg b.w.$^{-1}$ day^{-1}, respectively, for females and lower in offices for males than females (1.00×10^{-7} mg kg b.w.$^{-1}$ day^{-1} vs. 1.25×10^{-7} mg kg b.w.$^{-1}$ day^{-1}). DI_{dust} values in the present study were lower than 1 mg kg b.w.$^{-1}$ day^{-1}, which is the lowest observed adverse effect level (LOAEL) recommended by Darnerud *et al.* (2001). DI_{dust} values in the present study were conservatively estimated because PBDEs bioaccessibility was used as the measure of PBDEs absorption rate in the intestinal tract. In contrast to values of indoor PBDE DI_{dust} for adults in most previous studies (131 pg kg b.w.$^{-1}$ day^{-1} in Ni *et al.* (2011); 12 ng day^{-1} in Besis *et al.* (2014); 530 pg kg b.w.$^{-1}$ day^{-1} in Zhu *et al.* (2015); 426 pg kg b.w.$^{-1}$ day^{-1} in Li *et al.* (2015)), values of PBDE DI_{dust} (29.6–125 pg kg b.w.$^{-1}$ day^{-1}) in our study were notably lower because the absorption rate was assumed to be PBDEs bioaccessibility rather than 1.0. If we assumed the absorption rate was 1.00, the DI_{dust} values in the present study were 105–463 pg kg b.w.$^{-1}$ day^{-1} or 7.36–25.9 ng day^{-1}, which are comparable to those in the previously mentioned studies. The values of PBDEs DI_{dust} for medical staff in clinics and employees in electronics plants from the present study were still below global values in occupational microenvironments (i.e., offices) except for e-waste recycling sites.

For the non-cancer risks of workplace PBDEs exposure via indoor dust, none of the HQ values for BDE-47, -99, -153, and -209 (which ranged from 4.04×10^{-7} to 1.19×10^{-5} for medical staff and employees in electronics plants) exceeded the critical value of 1.00, indicating that ingestion of indoor dust with PBDEs was not a lifetime cause of non-cancer neurobehavioral diseases for medical staff and

Table 2. Published PBDE levels accumulated on air-conditioner filters (ng g^{-1}).

Country	Dust type (number)	Sampling year	BDE–209 Mean (median)	ΣPBDEs Mean (median)	PBDE congeners detected	Reference
China	CASF (n = 56) in offices [a]	2008–2009	618 (258)	920 (477)	15 PBDEs: BDE-28, 47, 49, 85, 99, 100, 138, 153, 154, 183, 196, 206, 207, 208, 209	Ni *et al.* (2011)
Philippines	IAF (n = 8) in a university [b]	2010	2172 (NS)	–	1 PBDE: BDE–209	Fulong and Espino (2013)
Greece	CASF (n = 20) in workplaces [c]	2012	1106 (969)	1217 (1092)	21 PBDEs: BDE-15, 17, 28, 47, 66, 71, 77, 85, 99, 100, 119, 138, 153, 154, 166, 181, 183, 203, 206, 207, 209	Besis *et al.* (2014)
USA	CASF (n = 14) in retail stores	–	4960 (1030)	17,900 (6020)	14 PBDEs: BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 209	Xu *et al.* (2015)
Taiwan	IAF (n = 13) in clinics[d]	2013	731 (463)	1330 (606)	14 PBDEs: BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, 209	The present study
Taiwan	IAF (n = 8) in offices	2014	1660 (1840)	2590 (2710)	14 PBDEs: BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, 209	The present study
Taiwan	CASF (n = 6) in clean rooms	2014	9930 (10,800)	33,600 (25,900)	14 PBDEs: BDE-28, 47, 99, 100, 153, 154, 183, 196, 197, 203, 206, 207, 208, 209	The present study

[a] All offices were in the same new office building.
[b] Eight central air-conditioner filter (CASF) dust samples were from 4 offices, 2 libraries, a classroom, and a computing facility.
[c] Twenty CASF dust samples were collected from 2 electronics equipment stores, 5 coffee shops, 3 restaurants, 4 internet cafes, a public library, 3 offices, a chemical laboratory in a university, and a building of a local newspaper.
[d] Thirteen air-conditioner filter (IAF) dust samples were from 9 medical clinics and 4 dental clinics.

Table 3. Monte Carlo simulation assessment of the health risk of PBDEs to employees via ingestion of indoor dust in medical clinics, dental clinics, and offices in electronics plants.

	Medical clinics	Dental clinics	Offices
Daily intake (DI $_{dust}$)			
Males (mg kg b.w.$^{-1}$ day^{-1})			
BDE–209	1.70×10^{-8} (8.52×10^{-9})[a]	8.96×10^{-9} (1.02×10^{-8})	3.30×10^{-8} (3.65×10^{-8})
Σ_{14}PBDEs	7.12×10^{-8} (1.18×10^{-7})	2.96×10^{-8} (2.84×10^{-8})	1.00×10^{-7} (8.90×10^{-8})
Females (mg kg b.w.$^{-1}$ day^{-1})			
BDE–209	2.12×10^{-8} (1.07×10^{-8})	1.12×10^{-8} (1.28×10^{-8})	4.12×10^{-8} (4.57×10^{-8})
Σ_{14}PBDEs	8.89×10^{-8} (2.44×10^{-8})	3.71×10^{-8} (3.55×10^{-8})	1.25×10^{-7} (1.11×10^{-7})
Non-cancer risk (HQs)			
Males (dimensionless)			
BDE-47	4.58×10^{-6} (1.62×10^{-5})	2.26×10^{-6} (1.43×10^{-6})	9.25×10^{-6} (3.30×10^{-6})
BDE-99	1.03×10^{-5} (2.02×10^{-5})	2.72×10^{-6} (1.95×10^{-6})	9.23×10^{-6} (9.18×10^{-6})
BDE-153	5.28×10^{-6} (5.55×10^{-6})	8.37×10^{-7} (9.09×10^{-7})	2.07×10^{-6} (1.83×10^{-6})
BDE-209	7.65×10^{-7} (3.84×10^{-7})	4.04×10^{-7} (4.60×10^{-7})	1.49×10^{-6} (1.65×10^{-6})
Females (dimensionless)			
BDE-47	5.28×10^{-6} (1.37×10^{-6})	2.49×10^{-6} (1.65×10^{-6})	1.07×10^{-5} (2.16×10^{-5})
BDE-99	1.19×10^{-5} (2.33×10^{-6})	3.14×10^{-6} (2.25×10^{-6})	1.06×10^{-5} (1.62×10^{-5})
BDE-153	6.09×10^{-6} (6.40×10^{-7})	9.65×10^{-7} (1.65×10^{-6})	2.39×10^{-6} (1.48×10^{-6})
BDE-209	8.83×10^{-7} (4.23×10^{-7})	4.66×10^{-7} (5.31×10^{-6})	1.71×10^{-6} (9.67×10^{-7})
Cancer risk (Rs)			
Males (dimensionless)			
BDE-209	3.75×10^{-12} (1.88×10^{-12})	1.98×10^{-12} (2.25×10^{-12})	7.28×10^{-12} (8.07×10^{-12})
Females (dimensionless)			
BDE-209	4.32×10^{-12} (2.17×10^{-12})	2.28×10^{-12} (2.60×10^{-12})	8.40×10^{-12} (4.74×10^{-12})

[a] Mean (median).

office employees. Like the present study, previous studies reported HQ values that were much below the threshold value of 1.00 (Li *et al.*, 2015; Zhu *et al.*, 2015). Zhu *et al.* (2015) suggested that these low HQ values (at least two orders of magnitude below the critical value of 1.00) are an indication of safe and acceptable exposure to indoor dust PBDEs through ingestion.

Based on the information from the US EPA IRIS, BDE-209 is the only BDE congener associated with human cancer risk with neurobehavioral effects. In our study, the Rs for occupational exposure to BDE-209 via ingestion of indoor dust were < 1.00×10^{-6} in medical staff and factory employees. Therefore their exposure was considered to be at levels incapable of causing cancer with neurobehavioral effects during their lifetime. Our R values for indoor dust ingestion in an office environment were consistent with those reported by Li *et al.* (2015) and approximately several orders of magnitude below the threshold level of 1.00×10^{-6} (Li *et al.*, 2015).

Watkins *et al.* (2013) suggested the use of surface swipes to assess exposure in indoor environments because PBDE levels in indoor air is highly associated with dust levels of surface wipe and floor and a convenient way to collect indoor dust. Distribution of dust PBDEs in the indoor environment is affected by ventilation, air exchange, the use of electronic devices, human activity, and building materials. Spatio-temporal variability of indoor dust deposition can affect the amount of PBDEs contaminating indoor dust (Batterman *et al.*, 2010; Li *et al.*, 2015). Collection of floor dust and indoor air is reflective of short-term PBDEs contamination in the indoor environment. Xu *et al.* (2015b) proposed collecting dust PBDEs from air filters in building heating, ventilation, and air-conditioning (HVAC) systems as an alternative to conventional dust sampling. Independent air conditioners are popular and widely used in indoor environments including homes and workplaces in Taiwan, particularly southern Taiwan. Southern Taiwan is located in a tropical zone and air conditioners are continuously used for 7–8 months. Collection of IAF dust to detect PBDEs may be a convenient and effective way to monitor PBDEs exposure in indoor environments.

Advantages and Limitation

There are several advantages for the use of the air-conditioner filter dust as indoor dust to represent PBDEs contamination in the indoor environment. Firstly, air-conditioner filer dust is the indoor environmental sample with consideration of spatial and temporal distribution. Secondly, most PBDEs contaminated on the dust compared with other environmental matrices. Thirdly, air-conditioner filter dust is more convenient to collect compared with the floor dust. There is also some limitation in the present study. Degradation and debromination of PBDEs on the air-conditioner filter dust did not considered in the present study. Health assessment of the clean-room workers was excluded probably due to lack of risk parameters when the workers used the masks and protective clothing. Workers in this study are referred to the employees in the medical and dental clinics and the offices of the color filter manufacture facilities.

CONCLUSIONS

In this study, we investigated PBDE levels in air-conditioner filter dust from medical clinics, dental clinics, and factory offices and factory clean rooms in electronic color filter manufacturing plants. The levels of PBDEs in dust were lower from dental clinics than from clinics and electronics plants. Extremely high levels and notably different patterns of PBDEs were found in CASF dust collected from clean rooms in electronic color filter manufacturing plants compared to those in IAF dust collected from the clinics and offices. Workers in the clinics and offices with occupational exposure to PBDEs through indoor dust ingestion were below threshold values and therefore unlikely to cause adverse health effects. In contrast, compared with the general population exposure to house dust, the health risks of workers with occupational exposure to PBDEs in the clinic or office dust might be lower.

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DISCLAIMER

The authors declare no conflicts of interest.

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