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Polybrominated diphenyl ethers and novel brominated flame retardants in indoor dust of different microenvironments in Beijing, China

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ABSTRACT

The occurrence levels of eight polybrominated diphenyl ethers (PBDEs) and four novel brominated flame retardants (NBFRs) were determined and compared in indoor dust from different microenvironments (21 homes, 23 offices and 16 day care centers) in Beijing, China. Concentrations of Σ_8PBDEs in dust were 430–17,000 ng/g, 690–8600 ng/g, and 90–2300 ng/g for homes, offices, and day care centers, respectively, and were dominated by BDE-209. Concentrations of Σ_4NBFRs ranged from 310 to 17,000 ng/g, 300 to 4300 ng/g, and not detected to 500 ng/g for homes, offices, and day care centers, respectively, and were dominated by bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (BEH-TEBP) and decabromodiphenylethane (DBDPE) across microenvironments. The results showed an increasing detection and elevated concentration of NBFRs (especially BEH-TEBP), indicating that monitoring of NBFRs in dust samples should be of concern in future studies. A notable finding was that the BFR concentrations in dust samples from day care centers were generally one order of magnitude lower than those from homes and offices in the present study. This implies that previous estimates of toddler exposure via dust ingestion on data from homes may be overestimated. Concentrations of BDE-209 and Σ_8PBDEs were found to be significantly higher in elevated surface dust than floor dust from day care centers. The estimates of daily intakes of BFRs via dust ingestion for Chinese adults and toddlers using Monte Carlo analysis were 2–5 orders of magnitude lower than the corresponding reference daily intakes.

1. Introduction

Brominated flame retardants (BFRs) are chemicals added to a diverse group of products (textiles, carpets, electrical devices, furniture foams, cables, building materials, etc.) to reduce their flammability and meet flame retardant standards (de Wit, 2002). Polybrominated biphenyls ethers (PBDEs), hexabromocyclododecane (HBCDD), and tetrabromobisphenol A (TBBPA) are three of the most extensively used BFRs. Studies have demonstrated the pervasive environmental occurrence, bioaccumulation potential and toxicity of these BFRs (de Wit, 2002; Besis and Samara, 2012; Stieger et al., 2014; Yu et al., 2016). These chemicals can also cause thyroid disorders, diabetes, neurobehavioral and developmental disorders (Kim et al., 2014; Lyche et al., 2015). As a result, there have been extensive bans and phase-out of some PBDEs, as well as restrictions of the use of HBCDD (Besis and Samara, 2012; Al-Omran and Harrad, 2016; Besis et al., 2017), resulting in increased production and marketing of novel BFRs (NBFRs) as substitutes for the phased-out chemicals. Therefore, there have recently been increasing concerns regarding NBFRs as they have been found in various environmental media as well as humans (Newton et al., 2015; Cristale et al., 2016; Tao et al., 2016; Besis et al., 2017; He et al., 2017; Zheng et al., 2017; Sun et al., 2018; Wang et al., 2018).

Studies have shown that the indoor environment is an important source of BFRs (Covaci et al., 2011). Moreover, ingestion of indoor dust is among the most important pathways of human exposure to these indoor released chemicals (Frederiksen et al., 2009; Harrad et al., 2010;

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Besis and Samara, 2012), especially for toddlers and children because of their frequent hand-to-mouth behavior (Jones-Otazo et al., 2005; Johnson-Restrepo and Kannan, 2009). The relatively higher exposure of children than adults to PBDEs has also been revealed in biomonitoring studies (Toms et al., 2009; Lunder et al., 2010). Moreover, toddlers and children are thought to be more vulnerable to these contaminants as they have a decreased metabolic capacity to eliminate xenobiotic contaminants (Landrigan et al., 2004). These facts highlight the significance of including toddlers and children as a particularly relevant group in assessments of human exposure to BFRs in indoor dust.

Numerous studies have already evaluated the occurrence and human exposure to BRFs in indoor dust from homes and offices of different regions and countries (Harrad et al., 2008b; Ali et al., 2011; Ali et al., 2012; Cao et al., 2014; Cequier et al., 2014; Qi et al., 2014; Cao et al., 2015; Hassan and Shoeib, 2015; Li et al., 2015; Al-Omran and Harrad, 2016). These studies are important but could possibly be insufficient to accurately assess the exposure of toddlers and children to BFRs through dust ingestion, because toddlers and children would also spend considerable time in other important microenvironments, such as day care centers (Harrad et al., 2010; Malliari and Kalantzi, 2017; Larsson et al., 2018). However, data regarding BFRs in dust of day care centers are still limited at present (Malliari and Kalantzi, 2017). Furthermore, knowledge regarding differences in the BFR occurrence in dust of day care centers to other microenvironments is needed.

Therefore, in the present study, BFRs were measured in dust samples from different microenvironments in Beijing, China, including homes, offices and day care centers. The specific goals of our study were to (1) compare BFR levels in indoor dust from different microenvironments, and (2) estimate human exposure by differentiating microenvironments relevant to adults and toddlers. Moreover, BFR levels in elevated surface dust (ESD) and floor dust (FD) were compared for selected day care centers. Although some PBDEs are no longer produced, old consumer products that contain these chemicals could still be in use in indoor microenvironments. Hence, both legacy PBDEs and NBFRs were considered in the present study. The NBFRs considered were 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), decabromodiphenylethane (DBDPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), and bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (BEH-TEBP).

2. Materials and methods

2.1. Sample collection

Indoor dust samples were collected from 21 homes, 23 offices and 16 day care centers located in the urban area of Beijing, China between spring 2012 and summer 2013. Of the 16 day care centers, nine were selected to collect both FD and ESD (e.g., furniture, tables) samples. The other seven were not sampled because no furniture or tables were available for sampling. All dust samples were collected using a vacuum cleaner in conjunction with nylon sampling socks (25 μm, Guangzhou Qixin Filter Ltd., China). The nozzle of the vacuum cleaner was cleaned thoroughly with acetone before and after each sampling event to prevent the possible cross contamination. For homes, composite dust samples were collected from the floors of the living room, bedroom and dining room, while the entire floor was sampled in offices and day care centers. After sampling, each sample was passed through a 500 μm stainless steel sieve and wrapped with solvent rinsed aluminum foil. All dust samples were sealed in plastic zip lock bags and stored at −20 °C until analysis.

2.2. Analytical procedure

2.2.1. Sample extraction and clean-up

The sample pretreatment method was adapted from van den Eede et al. (2012). Briefly, PBDEs and NBFRs were simultaneously extracted from dust samples by ultrasonic extraction. Before extraction, about 50 mg of dust was spiked with 100 μL internal standards (IS) of BDE-77 (50 pg/μL), −128 (50 pg/μL) and ^{13}C-BDE-209 (500 pg/μL) to estimate the recoveries. Extraction was conducted in an ultrasonic bath with 3 mL solvent mixture of *n*-hexane/acetone (3:1, v/v) for 10 min (repeated three times). After each extraction, the samples were centrifuged at 3500 rpm for 2 min and the supernatants were collected and combined for further processing.

The combined extract was subsequently concentrated to approximately 1 mL in a rotary evaporator and applied to a Florisil cartridge (Supelclean ENVI-Florisil, 500 mg/6 cc, Supelco) that had been preconditioned with 6 mL *n*-hexane. The sample was subsequently eluted with 10 mL of solvent mixture of *n*-hexane/dichloromethane (DCM) (2:1, v/v) as the first fraction (F1). The sample was then further eluted with 8 mL ethyl acetate (F2), after which this fraction was used to determine the BEH-TEBP. The F1 volume was reduced to 1 mL and transferred to a 44% acidic silica gel column (600 mg/6 cc) that was subsequently eluted with 10 mL of a solvent mixture of *n*-hexane/DCM (1:1, v/v). Both fractions were concentrated to incipient dryness (approximately 5 μL) under a gentle nitrogen stream and made up in 100 μL of injection standards (50 pg/μL PCB209). Information on chemicals and reagents is given in detail in the Supplementary materials.

2.2.2. Instrumental analysis

The target analysis of the eight PBDE congeners and four NBFRs was conducted using a Shimadzu GC2010 gas chromatograph coupled with a QP2010 mass spectrometer (GC–MS, Kyoto, Japan). The MS was operated in electron capture negative ionization (ECNI) mode. Aliquots of sample extracts (1 μL) were injected into a DB5-MS capillary column (15 m × 0.25 mm × 0.10 μm) with helium as the carrier gas (1.5 mL/min flow rate). The temperature of the ion source and interface was set at 280 °C and 300 °C, respectively. The IS calibration procedure was used for quantification of BFRs. Further details of the GC-ECNI/MS method can be found in the Supplementary materials.

2.3. Quality assurance and quality control

To evaluate our method performance, four solvent washed (*n*-hexane/acetone, 3:1, v/v) dust samples were spiked with known amounts of target BFRs at two concentration levels. As shown in Table S1, the determined concentrations were in accordance with the spiked values at each spiked concentration, indicating that our method performed well for the target BFRs. Procedure blanks were run with each batch of samples to evaluate possible contamination during analysis and the levels of target compounds were below the limit of quantification (LOQ) in most cases. The LOQs of BFRs were estimated as the concentration when a signal to noise ratio was 10:1 (S/N = 10). The LOQs ranged from 0.06 (BDE-28) to 6.5 ng/g (BDE-209) for PBDEs, and from 0.60 (BTBPE) to 16 ng/g (DBDPE) for NBFRs, assuming an final extract volume of 100 μL and dust samples of 50 mg. The LOQs for each individual compound are provided in Table S1. The average recoveries of IS were 116 ± 15%, 72 ± 17%, and 86 ± 26% for BDE-77, BDE-128, and ^{13}C-BDE-209, respectively.

2.4. Estimation of daily intake

The estimated daily intake (EDI, ng/kg bw/day) of BFRs via dust ingestion was calculated according to the following equation (Abdallah and Covaci, 2014; Mizouchi et al., 2015):

$$\mathrm{EDI} = [(C_H F_H) + (C_O F_O) + (C_D F_D)] \times RR \times AF/BW \quad (1)$$

where C_H, C_O and C_D are BFR concentrations (ng/g) in dust samples from homes, offices, and day care centers, respectively; and F_H, F_O and F_D, which are the average percentages of time spent in each microenvironment, are 62.5%, 37.5%, and 0% for adults and 62.5%, 0%, and 37.5% for toddlers (Wu et al., 2016). Additionally, RR is the daily dust

ingestion rate (g/day), which was assumed to be 20 and 50 mg/day for adults and toddlers, respectively (Jones-Otazo et al., 2005); AF is the fraction of BFRs absorbed and assumed to be 100%; and BW is the body weight (kg), which were set at 57.5 and 11.8 kg for average adults and toddlers, respectively (NHFPC, 2002).

Considering that every parameter varies because of interference by external confounding factors without control, the range of EDIs was estimated using Monte Carlo analysis to interpret the possible variations in each parameter used in the calculation. To accomplish this, each parameter was assigned an uncertainty factor and distribution type based on the best judgment to each input parameter. Detailed information regarding parameter values and distribution type can be found in Table S2.

2.5. Statistical analysis

Descriptive analysis was performed using Microsoft Excel 2016 and SPSS (version 19.0). Concentrations of BFRs below the LOQs were left at zero. The Mann-Whitney *U* test was executed to test the differences between concentrations of BFRs in dust samples from the three microenvironments. The Wilcoxon signed rank test was utilized to investigate the difference in BFR concentrations between floor and elevated surface dust samples collected from the same day care center. A p value ≤ 0.05 was considered statistically significant. Monte Carlo analysis (10,000 simulations) was employed to calculate the best estimates of means, confidence intervals and medians of EDIs for adults and toddlers, and this was accomplished using the Crystal Ball (Version 11.2) add-in in Microsoft Excel 2016.

3. Results

3.1. BFRs in different microenvironments

3.1.1. PBDEs

BDE-209, -47 and -99 were found with detection frequencies of 100% in all dust samples from homes and offices, followed by BDE-183, -28, -100 and -153 with detection frequencies varying from 86% to 96%. However, in dust samples from day care centers, only BDE-209 was found at 100% of sampling sites. Other congeners, including BDE-47, -100 and -99, had a relatively lower detection frequency (69–81%). Note that BDE-153 was not found in any of the day care center samples (Table S3).

The concentrations of Σ_8PBDEs ranged from 430 to 17,000 ng/g, 690 to 8600 ng/g, and 90 to 2300 ng/g in dust samples from homes, offices, and day care centers, respectively. Significant differences were observed for Σ_8PBDEs levels among the three microenvironments ($p < 0.05$), and the median Σ_8PBDEs concentrations followed the order of: offices > homes > day care centers (Fig. 1). The differences were also significant for individual PBDE congener across the three microenvironments and followed the same order as Σ_8PBDEs concentration levels. BDE-209 was the predominant congener in all dust samples of different microenvironments (Fig. S1), consisting 65–99% of the total PBDEs with an average value of 95% for all dust samples ($n = 60$). As for tri- to hepta-BDEs (sum of BDE-28, -47, -100, -99, -154, -153, -183), median concentrations of $\Sigma_{tri\text{-}hepta}$PBDEs followed the same trend as that for Σ_8PBDEs among the three microenvironments (Fig. 1). The congener profiles were similar for samples from homes and offices, where BDE-47, -99, -183 and -153 predominated the constitution of tri- to hepta-BDEs, but for day care centers, BDE-100, -47, and -99 were the predominant congeners (Fig. S2).

3.1.2. NBFRs

DBDPE and BEH-TEBP were detected in almost all of the dust samples (88–100%). A lower detection frequency (50%) for BTBPE in the dust samples from day care centers was observed than in homes (100%) and offices (96%). The detection frequency of EH-TBB in samples from offices and day care centers was 48% and 25%, respectively, which was lower than that in homes (86%) (Table S3).

The median concentration of Σ_4NBFRs in dust samples from homes was highest, followed by that of offices, whereas the median value was one order of magnitude lower for that of day care centers (Fig. 1). The higher concentration of Σ_4NBFRs in homes than in day care centers and offices occurred because the BEH-TEBP was detected at high concentrations in dust samples from homes. Statistical analysis revealed concentrations of DBDPE in offices significantly exceeded those in homes, while no significant differences between offices and homes were found for EH-TBB ($p = 0.363$) or BTBPE ($p = 0.120$). Concentrations of each individual NBFR in day care centers were significantly lower than in the other two microenvironments (Fig. 1). BEH-TEBP was most abundant in almost all of the samples from homes, whereas the composition profiles in dust samples from offices and day care centers were slightly different. Two obvious patterns were found; specifically, DBDPE was the predominant compound for some sampling locations, while DBDPE and BEH-TEBP made similar contributions to the total NBFRs for other locations (Fig. S3).

3.2. BFR occurrence in ESD versus FD in day care centers

Statistical descriptors of BFR concentrations were calculated for both ESD and FD samples from nine day care centers. The results shown in Table 1 indicate that the median ESD to FD ratios were 1.5–2.0 for BDE-209, Σ_8PBDEs, BTBPE, DBDPE and Σ_4NBFRs, suggesting higher chemical concentrations in ESD than FD. Similar concentrations in ESD to FD were observed for BDE-47, -99 and $\Sigma_{tri\text{-}hepta}$PBDEs as the median ESD to FD ratios were around 1.0. However, the median concentrations of BDE-100, EH-TBB and BEH-TEBP were lower in the ESD than the FD. To further investigate the differences between ESD and FD, a Wilcoxon signed rank test was run and the results showed that the differences between ESD and FD were only statistically significant for BDE-209 ($Z = -2.380$, $p = 0.017$) and Σ_8PBDEs ($Z = -2.192$, $p = 0.028$).

3.3. EDIs for adults and toddlers

The median values of EDIs for our investigated BFRs ranged from 1.3×10^{-3} (BDE-154) to 3.1 (BEH-TEBP) ng/kg bw/day for toddlers, and from 2.8×10^{-4} (BDE-28) to 0.60 (BDE-209) ng/kg bw/day for adults (Figs. 2 and 3, Table S4). The median EDI values for Σ_8PBDEs and Σ_4NBFRs were estimated to be 0.65 and 0.47 ng/kg bw/day for adults, respectively, whereas they were approximately 4 and 10 times higher for toddlers. The upper bounds of EDIs at the 90% confidence level (the worst-case scenario) were 4.4×10^{-3}–15 and 1.2×10^{-3}–2.2 ng/kg bw/day for toddlers and adults, respectively, which is still 2–5 orders of magnitude lower than the corresponding reference daily intakes (Hardy et al., 2008; USEPA, 2008) (Figs. 2 and 3). These findings indicate the low health risk for toddlers and adults exposed to BFRs via dust ingestion in our investigated sites. However, potential health risks should not neglected given the long-term and multi pathway exposure to BFRs.

We also calculated the relative contribution of human exposure to BFRs by ingestion of home dust to that via all types of indoor dust, which was expressed as EDI_{home}/EDI_{total} (Figs. 2 and 3, right axis). Interestingly, home dust contributed over 70% to toddlers' exposure to BFRs via dust for most investigated chemicals. However, for adults, < 30% of the PBDEs were from ingestion of home dust, while home dust contributed 50–80% of NBFRs encountered by adults via dust.

4. Discussion

4.1. Comparisons of BFR levels

The BFR concentrations in dust samples available in the literature were compared to those observed in the present study to put our results

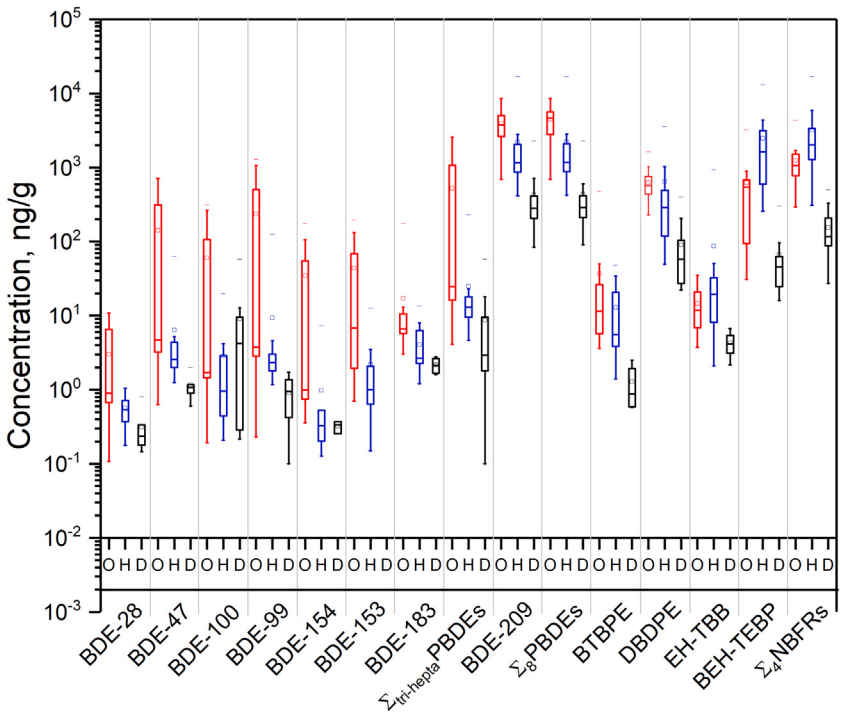

Fig. 1. Box plot showing the concentrations of BFRs in dust samples from the three microenvironments (O, H and D represents offices, homes and day care centers, respectively). The lower, middle, and upper lines in the box represent the 25th, 50th, and 75th percentile values, whereas the rectangle symbol and lower and upper bars represent the mean, minimum and maximum values, respectively.

into perspective. It should be noted that an accurate comparison of total PBDE concentrations across studies would be difficult as PBDE congeners investigated in dust differed from each other among existing studies. Therefore, the concentrations of BDE-209 were used for comparison between studies because BDE-209 was generally the predominant congener of indoor dust samples across almost all studied sites and could be used to reflect the total PBDE levels (Yu et al., 2016; Malliari and Kalantzi, 2017).

4.1.1. Home dust

The median concentration of BDE-209 in our dust samples from homes was moderate compared to those of 50 to 55,100 ng/g that were previously reported in home dust in China (Huang et al., 2010; Chen et al., 2011; Kang et al., 2011; Zheng et al., 2011; Yu et al., 2012; Zhu et al., 2013; Chen et al., 2014; Jiang et al., 2014; Qi et al., 2014; Wang et al., 2014; Wang et al., 2015; Zheng et al., 2015; Zhu et al., 2015; Meng et al., 2016; Sun et al., 2016; He et al., 2017; Peng et al., 2017; Zheng et al., 2017; Sun et al., 2018; Wang et al., 2018) (Table S5). When compared to the global levels summarized by Malliari and Kalantzi (2017), our median level of BDE-209 was comparable to those in Australia and New Zealand, higher than those in Africa, Asian countries (except Japan and South Korea) and Europe (except the United Kingdom), but lower than those reported in North America and the United Kingdom.

The median level of BEH-TEBP in home dust was significantly higher than that previously reported in China (Qi et al., 2014; Zheng

Table 1
Comparison of concentrations of BFRs (ng/g) in elevated surface dust (ESD) and floor dust (FD) from selected day care centers.

Compound	Elevated surface dust (n = 9)				Floor dust (n = 9)				Comparison of ESD and FD	
	DF (%)	Min.	Median	Max.	DF (%)	Min.	Median	Max.	Ratio (median)	p[a]
BDE-28	44	ND	0.20	0.36	56	ND	0.23	0.34	–	–
BDE-47	100	0.50	0.92	2.4	100	0.60	1.1	1.8	1.1	0.779
BDE-100	67	ND	0.37	0.51	67	ND	4.6	13	0.04	0.753
BDE-99	78	ND	0.81	1.9	89	ND	0.95	1.6	1.0	0.735
BDE-154	11	ND	0.57	0.57	0	–	–	–	–	–
BDE-153	44	ND	0.52	2.9	0	–	–	–	–	–
BDE-183	56	ND	2.7	17	44	ND	2.1	2.8	–	0.285
$\Sigma_{tri\text{-}hepta}$PBDEs[b]	–	0.57	4.0	26	–	0.89	2.8	18	1.2	0.859
BDE-209	100	90	800	19,000	100	200	360	2300	1.8	0.017*
Σ_8PBDEs[c]	–	91	810	19,000	–	210	360	2300	1.8	0.028*
BTBPE	56	ND	1.7	42	67	ND	1.3	2.5	1.5	1.000
DBDPE	78	ND	200	430	100	23	58	400	1.7	0.310
EH-TBB	33	ND	3.5	3.8	44	ND	4.1	6.6	0.5	0.180
BEH-TEBP	78	ND	30	110	89	ND	58	300	0.5	0.116
Σ_4NBFRs[d]	–	ND	270	550	–	27	127	500	1.6	0.398

Abbreviations: DF, detection frequency; ND, not detected.
[a] Wilcoxon sign rank test.
[b] Sum of BDE-28, -47, -100, -99, -154, -153, and -183.
[c] Sum of BDE-28, -47, -100, -99, -154, -153, -183, and -209.
[d] Sum of BTBPE, DBDPE, EH-TBB, and BEH-TEBP.
* $p \leq 0.05$.

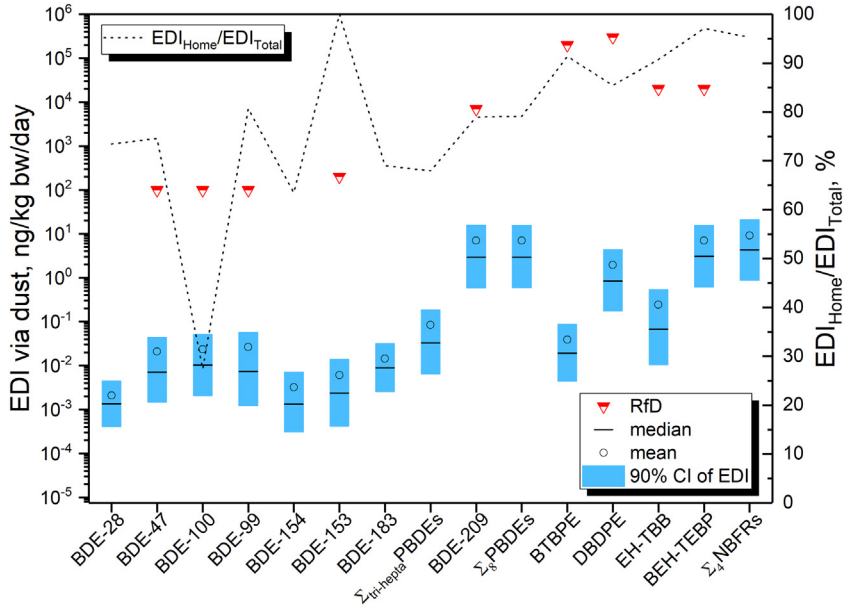

Fig. 2. Estimated daily intake (EDI) of BFRs via indoor dust ingestion for toddlers compared to the reference daily intakes (RfD) (left axis), and the ratio of BFR intake through home dust (EDI_{Home}) to that via all types of indoor dust (EDI_{Total}) (right axis). RfD values were from Hardy et al. (2008) and the USEPA (2008).

et al., 2015; Peng et al., 2017; Sun et al., 2018), and at the high end among studies worldwide (Malliari and Kalantzi, 2017). The median level of EH-TBB was comparable to those reported in previous studies in China (Qi et al., 2014; Zheng et al., 2015; Peng et al., 2017). According to Malliari and Kalantzi (2017), the highest concentration of EH-TBB (> 1000 ng/g) was found in North America, whereas the EH-TBB levels reported in Europe and other Asian countries were similar to those observed in the current study.

The levels of DBDPE and BTBPE observed in home dust samples were comparable or slightly lower than those reported in most studies conducted in China (Zheng et al., 2011; Chen et al., 2014; Qi et al., 2014; Zheng et al., 2015; He et al., 2017; Peng et al., 2017; Zheng et al., 2017; Sun et al., 2018; Wang et al., 2018) and worldwide (Malliari and Kalantzi, 2017).

4.1.2. Office dust

Table S6 shows a global comparison of BFR concentrations in office dust. The median concentration of BDE-209 observed in the present study was comparable to that in South-Central China (Huang et al., 2010), slightly higher than that in Shanghai (Li et al., 2015) and Hong Kong (Kang et al., 2011), but significantly higher than that in Guangzhou (Zheng et al., 2017), Hangzhou (Sun et al., 2016) and another study in Beijing (Wang et al., 2018). When compared to studies conducted worldwide, our results were similar to those reported for office dust in Brazil (Cristale et al., 2018), the United States (Watkins et al., 2011) and the United Kingdom (Tao et al., 2016), but higher than those in Greece (Besis et al., 2014), Turkey (Kurt-Karakus et al., 2017), Belgium (D'Hollander et al., 2010), Germany (Brommer et al., 2012), Sweden (Newton et al., 2015), Australia (He et al., 2018; McGrath et al., 2018), and Nigeria (Olukunle et al., 2015). Overall, the offices

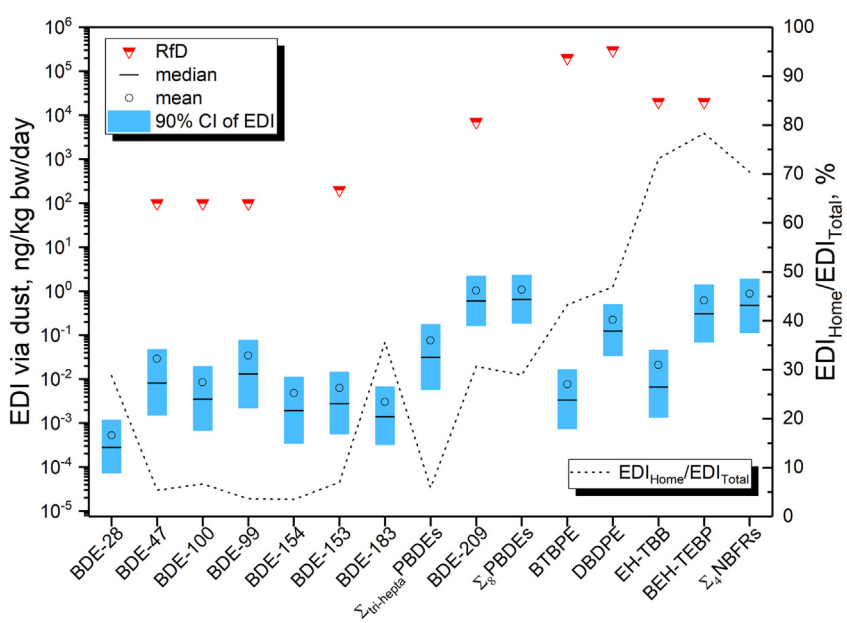

Fig. 3. Estimated daily intake (EDI) of BFRs via indoor dust ingestion for adults compared to the reference daily intakes (RfD) (left axis), and the ratio of BFR intake through home dust (EDI_{Home}) to that via all types of indoor dust (EDI_{Total}) (right axis). RfD values were from Hardy et al. (2008) and the USEPA (2008).

investigated in this study had a relatively higher concentration of BDE-209, whereas the DBDPE levels were at the low end among concentrations reported to date.

Another predominant NBFR observed in office dust samples in the present study was BEH-TEBP. Our median concentration of BEH-TEBP (546 ng/g, Table S3) was lower than that reported by Cristale et al. (2018) for Brazil office dust, but higher than those reported in other studies (Ali et al., 2011; Newton et al., 2015; Tao et al., 2016; Kurt-Karakus et al., 2017; Sun et al., 2018). Much higher concentrations for EH-TBB were found in office dust from Turkey and Brazil (Kurt-Karakus et al., 2017; Cristale et al., 2018), while similar concentrations were reported by others (Ali et al., 2011; Newton et al., 2015; Tao et al., 2016; McGrath et al., 2018). The BTBPE levels observed in the present study were similar to those reported in China (Sun et al., 2018; Wang et al., 2018), Belgium (Ali et al., 2011), Turkey (Kurt-Karakus et al., 2017) and Sweden (Newton et al., 2015), but slightly lower than those reported in the United Kingdom (Tao et al., 2016).

4.1.3. Day care center dust

Very few studies reported concentrations of BFRs in dust samples from day care centers in China. We previously reported the occurrence of BFRs in indoor dust of four kindergartens in Beijing (Cao et al., 2014). Another study has reported the occurrence of PBDEs in indoor dust of day care centers in China to date. The occurrence of BDE-47, -99, and -100 in indoor dust of kindergartens in Hongkong ranged from 0.08 to 1.1 ng/g, 0.75 to 7.6 ng/g, and 0.22 to 90 ng/g, respectively (Deng et al., 2016), which is similar to the results of the present study. Moreover, limited studies of BFRs in day care centers have been conducted worldwide, even though it is thought to be an important microenvironment for assessment of toddler exposure to BFRs via dust ingestion. The existing relative studies are compiled in Table S7. The BFR concentrations (except BEH-TEBP) observed in the present study were consistently lower than those reported in Brazil (Cristale et al., 2018), Spain (Cristale et al., 2016), Sweden (Larsson et al., 2018), the United Kingdom (Harrad et al., 2010; Ali et al., 2011) and the United States (Stubbings et al., 2018). The BEH-TEBP levels observed in the present study were higher than those reported in Brazil (Cristale et al., 2018) and the United States (Stubbings et al., 2018), similar to those reported in Sweden (Larsson et al., 2018), but lower than those reported in the United Kingdom (Ali et al., 2011) and Spain (Cristale et al., 2016).

4.2. Comparisons between microenvironments

Our results demonstrated that the predominant PBDE congener in dust samples of the three investigated microenvironments was BDE-209, which is in accordance with the findings in most previous studies in China and worldwide (Yu et al., 2016; Malliari and Kalantzi, 2017). The contribution of BDE-209 to total PBDEs in dust samples was similar to that of commercial deca-BDE (Schecter et al., 2005), indicating the extensive use of commercial deca-BDE in our investigated sites and China (Yu et al., 2016). Deca-BDE was included as a persistent organic pollutant by the Stockholm Convention in 2017, but was produced in China in large quantity before then (Ji et al., 2017) and has been applied in older consumer products. Therefore, the monitoring of deca-BDE in the environment in China is still needed in future studies. Conversely, the small proportion of lower brominated PBDE congeners indicates that phase-out of commercial penta- and octa-BDE products in 2009 has had a significant impact on the contamination profile of BFRs in indoor environments.

Another general trend in the present study was that some alternative BFRs, e.g., DBDPE and BEH-TEBP, were detected at considerable frequencies and concentrations in all types of dust (Table S3). The increasing detection and elevated concentration of these NBFRs indicate that monitoring of NBFRs in dust samples of different microenvironments should be of concern in future studies, as the shift of consumption pattern from traditional to emerging BFRs has been emphasized (Wang et al., 2018). DBDPE is used as a replacement for deca-BDE, and China is one of the major producers of DBDPE worldwide (Zheng et al., 2011). As consumer products containing deca-BDE are phased out, more attention should be paid to the possible prevalence of DBDPE in Chinese environments. Another notable finding was that the BEH-TEBP levels in our investigated microenvironments were at the high end among studies in China and worldwide. BEH-TEBP and EH-TBB are applied as commercial mixtures (Firemaster 550 and Firemaster BZ-54) for replacement of penta-BDE, and BEH-TEBP is also used in the commercial mixture DP-45 (Stapleton et al., 2008; Venier et al., 2016). According to Ma et al. (2012), the ratio of EH-TBB to BEH-TEBP (f_{EtoB}) is around 0.7–0.8 in the commercial products Firemaster 550 and Firemaster BZ-54, but the f_{EtoB} ratios in our dust samples from the three microenvironments were all significantly lower than 0.7. Similarly, lower f_{EtoB} ratios were reported in indoor dust of previous studies (Stapleton et al., 2008; Ali et al., 2011; Qi et al., 2014). Therefore, BEH-TEBP detected in our investigated sites could be attributed to the use of commercial DP-45 in China. Another possible explanation could be that BEH-TEBP is more recalcitrant to degradation than EH-TBB (Davis and Stapleton, 2009).

There were differences between the three investigated microenvironments. The relative abundances of NBFRs to PBDEs differed across our investigated microenvironments. The median concentration of Σ_4NBFRs was higher than that of Σ_8PBDEs for home dust, while the opposite was observed for dust from day care centers and offices (Fig. 1 and Table S3). Furthermore, as mentioned above, BEH-TEBP was the most abundant NBFR in home dust, while both DBDPE and BEH-TEBP were predominant among NBFRs in offices and day care centers. Such differences could be attributed to the different types and abundances of BFR sources in the three microenvironments (Tao et al., 2016).

As for BFR concentrations, statistical analysis revealed that the concentrations of PBDE congeners and DBDPE in offices significantly exceeded those in homes, which is consistent with the results of a previous study (Tao et al., 2016). No significant differences between offices and homes were found for EH-TBB and BTBPE. For BEH-TEBP, a significantly higher concentration was observed in homes than offices in the present study, which differs from the results reported by Tao et al. (2016). Notably, significantly lower BFR concentrations were observed in dust samples from day care centers than those from homes and offices in the present study. This is in line with our previous observation for flame retardants in dust of different microenvironments in Beijing (Cao et al., 2014; Wu et al., 2016), and could reflect a lack of BFR sources and high cleaning frequency in day care centers (Brommer et al., 2012; Cao et al., 2014).

Few studies conducted to date have been intentionally designed to compare the BFR levels in day care centers to those in other microenvironments. The results of Cristale et al. (2018) showed that concentrations of NBFRs in primary schools were lower than in houses, offices and cars in Brazil, which is consistent with the results of the present study, although concentrations of BDE-209 in primary schools were similar to those in houses, but lower than those in offices in their case study. Another case study by these authors indicated that there were lower levels of BFRs in primary schools than in theaters and homes in Spain (Cristale et al., 2016). Ali et al. (2011) found that median concentrations of BEH-TEBP, BTBPE, DBDPE and EH-TBB in 36 dust samples from day care centers and primary schools in the United Kingdom were generally lower or comparable than those from homes in England, while BDE-209 was present at higher concentrations in day care centers and primary schools than in homes and offices in their study. However, Harrad et al. (2010) reported lower concentrations of PBDEs in primary schools in the United Kingdom, compared to homes and offices. Because few studies have explored the differences in BFR concentrations between day care centers and other microenvironments, we also conducted an inter-study comparison using the compiled supplementary data. The study by Stubbings et al. (2018) showed that the

median concentrations of BDE-209, BEH-TEBP, and DBDPE in their investigated child care centers in California were 160 ng/g, 4.2 ng/g, and 59 ng/g, respectively, which are lower than previously reported values for homes (Malliari and Kalantzi, 2017) and offices (Table S6) in the United States, whereas the median level of EH-TBB (2400 ng/g) of Stubbings et al. (2018) was higher than US homes and offices of other studies. Similar or comparable BFR concentrations were observed in dust samples when we compared the median values for preschools reported by Larsson et al. (2018) to those for general Swedish offices (Table S6) and homes (Malliari and Kalantzi, 2017).

Exposure of toddlers to BFRs via dust ingestion has attracted a great deal of attention because of their behavior (e.g. frequent hand-to-mouth) (Jones-Otazo et al., 2005; Johnson-Restrepo and Kannan, 2009) and low contaminant metabolism (Landrigan et al., 2004). Our results indicated that low BFR concentrations were observed in dust samples from day care centers, which has important implications for estimated exposure to toddlers. Toddlers' exposure to BFRs via dust ingestion is probably overestimated when only relying on the dataset of BFR concentrations in indoor dust samples from homes (activity pattern = 100% home), as they spend considerable time in day care centers, where they can benefit from less BFR contamination. However, it should be noted that the samples analyzed in the present study were limited in the number and spatial coverage of sampling sites. Much more work is needed to confirm this as these data are important to accurate assessment of toddlers' exposure to BFRs via dust ingestion.

4.3. Comparison of BFR concentrations in FD versus ESD

Our findings suggest that the concentrations of most BFRs were higher or similar in ESD than FD in day care centers. However, our results should be interpreted cautiously. Only nine sampling sites (small sample size) were selected to investigate the relationship of BFR concentrations between ESD and FD. Conversely, only BDE-47 and -209 were detected in all paired samples, although comparisons were conducted for all PBDE congeners and NBFRs. Nevertheless, our results demonstrated that the BDE-209 in ESD was significantly higher than that in FD. These findings are consistent with those of previous investigations of other microenvironments. For example, Al-Omran and Harrad (2016) reported median levels of PBDEs were higher in ESD than FD in Iraqi homes. Moreover, Allgood et al. (2017) found that median ESD levels of PBDEs and NBFRs were at least two times higher than those in paired FD samples from an academic environment. Similar findings were also reported by other researchers for dust in Swedish homes (Björklund et al., 2012) and in airplanes (Allen et al., 2013). The present study revealed differences in the concentrations of BFRs in FD to ESD in day care centers, another important indoor microenvironment, which further confirmed the need for future studies to estimate human exposure to BFRs based on a detailed understanding of where and how much time individuals spent in different indoor microenvironments (Allgood et al., 2017).

4.4. EDIs

4.4.1. Comparisons between studies

Different models for estimating the EDIs of BFRs from dust with different parameters have been used (Malliari and Kalantzi, 2017). To facilitate comparison among studies, literature reported median EDIs under the mean exposures scenario were compiled. Most of the compiled studies used mean estimates of 20 and 50 mg/day of dust intake for adults and toddlers, with a few exceptions (see Table S8). The available data describing human intake of BDE-209, BEH-TEBP, BTBPE, DBDPE and EH-TBB worldwide are provided in Table S8 and compared with our results in Fig. S4. Human exposure to BFRs via dust intake was found to be highly variable between different parts of the world and even within countries (Table S8). Our median EDIs for BDE-209 were at the moderate or the high end of previously reported EDIs worldwide (Harrad et al., 2008a, 2008b; Sjödin et al., 2008; Roosens et al., 2009; Takigami et al., 2009; D'Hollander et al., 2010; Harrad et al., 2010; Chen et al., 2011; Brommer et al., 2012; Kalachova et al., 2012; Lee et al., 2013; Bradman et al., 2014; Chao et al., 2014; Fromme et al., 2014; Jiang et al., 2014; Olukunle et al., 2015; Sahlström et al., 2015; Ali et al., 2016; Kim et al., 2016; Tao et al., 2016; Tay et al., 2017; Zheng et al., 2017; Cristale et al., 2018; He et al., 2018; Larsson et al., 2018; McGrath et al., 2018; Stubbings et al., 2018; Wang et al., 2018), with the lowest and highest EDIs being reported in Australia (Sjödin et al., 2008) and the United Kingdom (Harrad et al., 2008a), respectively. The median EDIs of DBDPE in the present study were at moderate levels around the globe (Harrad et al., 2008a; Ali et al., 2011; Ali et al., 2012; Brommer et al., 2012; Kalachova et al., 2012; Chen et al., 2014; Fromme et al., 2014; Qi et al., 2014; Sahlström et al., 2015; Ali et al., 2016; Coelho et al., 2016; Tao et al., 2016; He et al., 2017; Tay et al., 2017; Zheng et al., 2017; Cristale et al., 2018; Larsson et al., 2018; McGrath et al., 2018; Stubbings et al., 2018; Wang et al., 2018), and the highest EDIs of DBDPE has been reported in an e-waste recycling area in China (He et al., 2017). Interestingly, the estimated intakes of BEH-TEBP in previous studies were all lower compared to the current study (Ali et al., 2011; Ali et al., 2012; Fromme et al., 2014; Qi et al., 2014; Sahlström et al., 2015; Ali et al., 2016; Tao et al., 2016; Tay et al., 2017; Cristale et al., 2018; Larsson et al., 2018; Stubbings et al., 2018). As for EH-TBB and BTBPE, our estimate was in the middle of previously reported values (Harrad et al., 2008a; Ali et al., 2011; Ali et al., 2012; Chen et al., 2014; Fromme et al., 2014; Qi et al., 2014; Sahlström et al., 2015; Ali et al., 2016; Coelho et al., 2016; Tao et al., 2016; Kurt-Karakus et al., 2017; Tay et al., 2017; Cristale et al., 2018; McGrath et al., 2018; Stubbings et al., 2018; Wang et al., 2018).

4.4.2. Sensitivity analysis

Sensitivity analysis of human exposure models for adults and toddlers was assessed by conducting rank correlation calculations between input and output during Monte Carlo simulations for BDE-209, Σ_8PBDEs, four individual NBFRs, and Σ_4NBFRs. The results are shown in Fig. S5. The most influential parameter for estimating toddlers' EDIs was BFR concentration in home dust, with r values ranging from 0.65 to 0.82, followed by dust ingestion rate (0.53 to 0.70) and then body weight (−0.14 to −0.11). It is worth noting that the BFR concentration in dust of day care centers was not an influential factor for most BFRs. This is because significantly lower BFR concentrations were observed in dust samples from day care centers than homes, which made this parameter less influential for estimation of the daily intake of toddlers in the present study. However, as discussed above, more work is needed to add data regarding BFR levels in dust of day care centers, which is also emphasized in a recent review (Malliari and Kalantzi, 2017). This would facilitate determination of the influence of this parameter on total exposure of toddlers. For estimation of intakes of adults, the order of the most influential parameters was dust ingestion rate (0.59 to 0.83) > concentration in home dust (0.35 to 0.74) > concentration in office dust (0.10 to 0.54). Therefore, research on BFR concentrations in different microenvironments and dust ingestion behavior is of great significance to accurately quantify the human exposure to BFRs via dust.

5. Conclusions

The present study evaluated a suite of BFRs (including legacy PBDEs and NBFRs) in dust samples from homes, offices and day care centers in Beijing, China. BDE-209 was the predominant compound across all microenvironments, followed by DBDPE and BEH-TEBP. NBFRs were found with the rising detection and elevated concentration in our investigated microenvironments and should be of concern in future studies. Our results showed that the median concentrations of most BFRs in the dust of three investigated microenvironments followed the order of offices > homes > day care centers. In particular, the BFR

concentrations in day care centers were found to be one order of magnitude lower than those in homes and offices in the present study. This implies that previous estimates of toddlers' exposure to BFRs via dust ingestion may be overestimated based on data from homes only, as toddlers spend a considerable amount of time in day care centers, where the BFR concentrations were relatively low compared with homes. Moreover, the results suggest that concentrations of Σ_8PBDEs were higher in ESD than those in FD in day care centers. Finally, our estimates of exposure to BFRs via dust ingestion for the general population in Beijing, China were comparable to those previously reported values except BEH-TEBP, for which our estimates were relatively higher than those in previous studies. However, our EDIs fell well below the relevant health-based limit values, even in the worst-case scenario.

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Appendix A. Supplementary data

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