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RESEARCH ARTICLE

Occurrence and partitioning of brominated flame retardants (BFRs) in indoor air and dust: a 15-month case study in a test home

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Abstract

Ten polybrominated diphenyl ethers (PBDEs) and 16 novel brominated flame retardants (NBFRs) were measured in air and dust samples collected in a test home in Harbin, China, from January 2017 to June 2018. The PBDE and NBFR concentrations in indoor air were in the ranges of 0.598–14.5 pg m^{-3} and 9.28–686 pg m^{-3}, respectively. The ranges of the PBDE and NBFR concentrations in indoor dust were 221–1060 ng g^{-1} and 71.9–1160 ng g^{-1}, respectively. Brominated flame retardant (BFR) concentrations in indoor air were affected by the temperature, relative humidity (RH), and ventilation. The BFR concentrations in indoor dust did not show temperature dependence. All dust samples were sieved into 6 size fractions (F1–F6: 1000–2000 μm, 500–1000 μm, 250–500 μm, 125–250 μm, 63–125 μm, and < 63 μm). The mass percentage of BFRs in F6 was the highest. The BFR concentrations did not increase constantly with a particle size decrease, and the concentrations in F2 were higher than those in F3. The partitioning behavior of BFRs illustrates that the dust-air partitioning coefficient approximately approached equilibrium within F5, F6, and the total dust fraction (FA) in the test home when logK_{OA} was between 9.1 and 11.32. Air-dust fugacity fractions were calculated, and the results suggested that most of the BFRs were mainly transferred from air to dust in the indoor environment for F1–F6.

Keywords PBDEs · NBFRs · Influence factors · Source analysis · Particle size · Dust-air exchange

Introduction

Polybrominated diphenyl ethers (PBDEs) and novel brominated flame retardants (NBFRs) are brominated flame retardants (BFRs) commonly added to commercial materials and consumer goods to reduce fire possibility. With the ban or restriction of commercial penta-, octa-, and deca-BDE, the market demand for alternative flame retardant is increasing to comply with flammability standards for consumer products (Batterman et al. 2009). For example, decabromodiphenyl ethane (DBDPE) is used as an alternative flame retardant for deca-BDE, with production volumes in China and other countries reaching 25,000 t a^{-1} and 30,000 t a^{-1}, respectively (Covaci et al. 2011; Zhang and Gu 2013).

Studies have concluded that BFRs have negative effects on human and animal health (Johnson et al. 2013; Lema et al. 2007; Yu et al. 2019). Due to the persistent presence of PBDEs and the increasing use of NBFRs in indoor environments, they have been detected in indoor air and dust in many countries (He et al. 2018a; Sakhi et al. 2019; Wemken et al.

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2019; Xiong et al. 2019). Air inhalation, dust ingestion, and dermal contact are major pathways of human exposure to BFRs (Allen et al. 2007; Besis and Samara 2012; Ni et al. 2013). Therefore, the pollution characteristics and fate in indoor environments are important for studying the potential harm of BFRs to human health.

In recent years, few studies have focused on the variations in BFR concentrations in the same indoor environment (Bennett et al. 2015; Niu et al. 2018; Vorkamp et al. 2011). In these studies, concentrations were related to the emission source, ventilation, temperature, and relative humidity (RH) (Muenhor and Harrad 2012; Niu et al. 2019; Stapleton et al. 2008; Yu et al. 2012). Studies have reported that ventilation affects emission and sorption rates by impacting the mass transfer coefficient (Liagkouridis et al. 2014; Melymuk et al. 2011). Emission test chambers have shown that increased temperature leads to increased emission rates for plasticizer phthalate and PBDEs (Kajiwara and Takigami 2013; Kemmlein et al. 2003; Sun et al. 2018). Research has indicated that moisture prevents the adsorption of BFRs in dust (Kuang et al. 2016). It is worth noting that most sampling rooms were inhabited, and corresponding influential factors were more complex than those in chamber studies. Therefore, a study on the influential factors in the test home is crucial to evaluate the variability of the BFR concentrations.

Particle size is an important factor that influences the distribution of pollutants in dust (Xu et al. 2016). Compounds tend to be enriched in finer particles when surface adsorption plays a dominant role in compound accumulation (Al-Omran and Harrad 2016; Wei et al. 2009). However, BFR concentrations did not increase with the decreasing particle size in indoor environments, such as home, office, and hotel (Cao et al. 2014a; Cao et al. 2013; Chao et al. 2014; He et al. 2018b). The distribution of BFR concentrations under different particle sizes varies in different microenvironments.

In addition, the partitioning and exchange of BFRs between indoor air and dust are important factors that affect the pollution characteristics of BFRs in indoor environments. The dust-air partition coefficient (K_D) and fugacity fraction (*ff*) are commonly applied to characterize the pattern of dust-air partitioning and exchange of BFRs, respectively (Harner et al. 2001; Weschler and Nazaroff 2010). For BFRs, the relationship between $\log K_D$ and the octanol-air partition coefficient ($\log K_{OA}$) has been investigated in homes in the USA, Norway, and Australia, and the results suggested that these factors are significantly correlated (Cequier et al. 2014; Dodson et al. 2015; He et al. 2018a; Wemken et al. 2019; Weschler and Nazaroff 2010). Few studies have investigated the exchange of BFRs between indoor dust and air.

To identify the distribution of BFR concentrations and the dust-air partitioning pattern for different particle sizes in the same indoor environment over a period of time, this study measured 10 PBDEs and 16 NBFRs in air and dust in a test home in Harbin, China, from January 2017 to June 2018. The aims of this study are (1) to determine the occurrence of BFRs in indoor air and dust, (2) to investigate the distribution of BFRs among the particle size fractions of indoor dust, and (3) to examine the partitioning and exchange of BFRs between indoor air and dust.

Materials and methods

Chemicals

10 PBDEs, including BDE-28, 47, 66, 100, 99, 85, 153, 183, 138, and 209, and 16 NBFRs, including 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EHTBB), bis(2-ethylhexyl)tetrabromophthalate (BEHTBP), 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), ally-2,4,6-tribromophenyl ether (ATE), α/β-tetrabromoethylcyclohexane (α/β-TBECH), α/β-1,2,5,6-tetrabromocyclooctane (α/β-TBCO), 2,3,5,6-tetrabromo-p-xylene (p-TBX), 2-bromoallyl-2,4,6-tribromophenyl ether (BATE), pentabromotoluene (PBT), 1,2,3,4,5-pentabromobenzene (PBBZ), pentabromoethylbenzene (PBEB), hexabromobenzene (HBBZ), 2,3-dibromopropyl-2,4,6-tribromophenyl ether (DPTE), pentabromobenzyl acrylate (PBBA), tetrabromo-o-chlorotoluene (TBCT), and DBDPE were analyzed in this study (Tables S1 and S2).

Sample collection

An ordinary residence located on the 5th floor in Harbin, China, was selected as the test home, and the external environment and plane structure of the test home are shown in Fig. S1. Details on the electrical appliances, furniture, and interior decoration of the test home are given in Table S3.

Indoor air samples were collected once every 2–3 months via passive air samplers from March 2017 to June 2018. The living room (LR), bedroom 1 (BR1), bedroom 2 (BR2), south balcony (LS), and north balcony (LN) were chosen as indoor air sampling rooms. A total of 9 batches of samples were collected (A1–A9, Table S4). The polyurethane foam (PUF) disks were placed in treated aluminum boxes and stored at − 20 °C until extraction. Sampling information, such as the sampling time, temperature, and RH, was recorded in detail (Table S4).

Indoor dust samples were collected from January 2017 to June 2018 once every 2–3 months with a pretreated vacuum cleaner from the floor surface of various rooms: LR, BR1, and BR2. A total of 6 batches of samples were collected (D1–D6, Table S4). The dust samples were wrapped in aluminum foil and stored at − 20 °C until extraction. Since this home had no human interference except for taking samples, the low mass of the dust collected from three rooms was integrated into one indoor dust sample for analysis. All dust samples were sieved into 6 size fractions through different meshes made of stainless steel (F1–F6: 1000–2000 μm, 500–1000 μm, 250–500 μm, 125–250 μm, 63–125 μm, and < 63 μm).

Sample extraction

The PUF was spiked with CB-155 and $^{13}C_{10}$-BDE-209 and then Soxhlet-extracted with a mixed solution of acetone:hexane (1:1, v:v, 350 mL) for 24 h. After the extraction, 3 mL of isooctane was added and concentrated under a vacuum to evaporate to 2–3 mL. The extract was concentrated and solvent-exchanged to isooctane with a gentle stream of nitrogen, stored at − 20 °C until GC-MS analysis.

The dust sample was placed in a glass centrifuge tube and mixed with a mixture of acetone:hexane (1:4, v:v, 5 mL) and surrogate standards (CB-155 and $^{13}C_{10}$-BDE-209). Samples were ultrasonically extracted for 20 min before shaking extraction for 30 min and centrifuged at 4000 rpm for 5 min. The extraction was performed three times, and the three extracts were combined. The extract was concentrated and solvent-exchanged to isooctane with a gentle stream of nitrogen, stored at − 20 °C until GC-MS analysis.

QA/QC and data analysis

In the process of sample treatment, a method blank and a spiked blank were run with each batch of 10 samples. The average recoveries of target BFRs ranged from 72.3 to 114% in spiked samples, and the recoveries of the surrogate standards ($^{13}C_{12}$-BDE-209, CB-155) in samples ranged from 68.4 to 123%. The detection limit of BFRs in indoor air samples was between 0.017 and 3.61 pg m^{-3}, and the detection limit of BFRs in indoor dust samples was between 0.004 and 21.0 ng g^{-1} (Table S5). Statistical analysis was performed using SPSS 20.0. Statistical significance was measured if $p < 0.05$. The data analysis and related equations are presented in the SI.

Results and discussion

BFR occurrence in air

Concentrations and profiles of BFRs in air

The concentrations of individual BFRs in the air samples collected in five rooms are listed in Table 1. BDE-85, 153, 138, 183, and 209, β-TBCO, TBCT, PBT, EHTBB, BTBPE, and DBDPE were not detected in the air samples.

PBDEs BDE-28, 47, 66, 99, and 100, as the main components of penta-BDE, were detected in air with a detection frequency (DF) higher than 60%. The concentrations of $\sum_5$PBDEs ranged from 0.598 to 14.5 pg m^{-3}, with a median of 3.12 pg m^{-3} and an average of 4.19 pg m^{-3}. The PBDE congener profile (average concentration percentage ± std. dev.) was dominated by BDE-28 (53% ± 14%) > BDE-100 (27% ± 21%) > BDE-47 (20% ± 9%) > BDE-99 (11% ± 10%) > BDE-66 (7% ± 3%). In this study, the concentrations of $\sum_5$PBDEs were comparable to those in the Czech Republic (Venier et al. 2016) and Sweden (Newton et al. 2015) and lower than those in the USA, Canada (Venier et al. 2016), and Ireland (Wemken et al. 2019) (Table S6).

NBFRs The concentrations of $\sum_{11}$NBFRs varied from 9.28 to 686 pg m^{-3}, with a median of 37.5 pg m^{-3} and an average of 157 pg m^{-3}. The DF of NBFRs was high, with > 60% detection, except for p-TBX, DPTE, and PBBA, which had 31%, 47%, and 49% DFs, respectively. This result suggested that most BFRs commonly persist in indoor air (Niu et al. 2019; Wemken et al. 2019). HBBZ was the predominant contaminant, with an average concentration of 125 pg m^{-3}, followed by BATE (16.2 pg m^{-3}), β-TBECH (6.09 pg m^{-3}), α-TBECH (5.27 pg m^{-3}), BEHTBP (3.61 pg m^{-3}), PBBZ (3.01 pg m^{-3}), DPTE (1.45 pg m^{-3}), ATE (1.41 pg m^{-3}), PBEB (0.434 pg m^{-3}), p-TBX (0.422 pg m^{-3}), and PBBA (0.222 pg m^{-3}). The HBBZ concentrations in this study were comparable to those in Besiktas, Turkey (Kurt-Karakus et al. 2017), and much higher than those in Norway, Sweden, the UK, the USA, Canada, and the Czech Republic (Cequier et al. 2014; Newton et al. 2015; Tao et al. 2016; Venier et al. 2016) (Table S7).

The average concentration and composition of PBDEs and BFRs in indoor air from five different rooms during the monitoring period are illustrated in Fig. S2. The values imply that the levels of PBDEs and NBFRs both decreased in the following order: LR > LS > LN > BR2 > BR1; moreover, BDE-28 and HBBZ were the main pollutants in each room. The highest average concentrations of PBDEs and NBFRs observed in LR with more household products than the other areas were 1 to 9 times higher than those in other rooms, suggesting spatial variability within the test home. The differences in the application of emission sources affected BFR concentrations (Allen et al. 2007; Hazrati and Harrad 2006). The correlations among the BFR concentrations in air in different rooms are discussed in the SI.

Influential factors

To further evaluate the variability of the BFR concentrations without external interference and determine the main influential factors, the relationships between BFR concentrations and

Table 1 Concentrations of BFRs in indoor air and dust

	Indoor air (pg m^{-3})						Indoor dust (ng g^{-1})				
	DF (%)	Average	Std. dev.	Median	Range		DF (%)	Average	Std. dev.	Median	Range
BDE-28	100	2.14	1.79	1.66	0.332–9.56	BDE-28	100	6.61	16.9	0.668	0.294–57.2
BDE-47	96	0.971	0.92	0.503	nd–3.51	BDE-47	82	2.27	2.92	1.03	nd–9.46
BDE-66	60	0.357	0.26	0.305	nd–0.942	BDE-66	91	8.36	15.2	0.140	nd–45.1
BDE-100	69	0.971	1.41	0.541	nd–7.24	BDE-100	82	13.3	14.2	5.33	nd–41.4
BDE-99	60	0.463	0.47	0.296	nd–2.19	BDE-99	82	2.40	5.60	0.450	nd–17.2
BDE-85	-	-		-	-	BDE-85	73	3.82	5.03	1.47	nd–12.6
BDE-153	-	-		-	-	BDE-153	100	2.90	7.38	0.494	0.164–25.0
BDE-183	-	-		-	-	BDE-183	100	3.51	4.88	1.20	0.390–16.2
BDE-138	-	-		-	-	BDE-138	45	0.0288	0.0160	0.0296	nd–0.0488
BDE-209	-	-		-	-	BDE-209	100	409	230	235	195–904
$\sum_5$PBDEs[a]		4.19	3.17	3.12	0.598–14.5	$\sum_{10}$PBDEs[c]		447	334	259	221–1060
α-TBECH	93	5.27	2.31	4.78	nd–10.7	α-TBECH	-	-		-	-
β-TBECH	96	6.09	4.77	4.59	nd–26.9	β-TBECH	100	8.81	14.3	4.92	0.801–51.5
β-TBCO	-	-		-	-	β-TBCO	91	4.13	3.34	2.78	nd–10.4
ATE	96	1.41	1.38	1.14	nd–8.56	ATE	-	-		-	-
p-TBX	31	0.422	0.340	0.271	nd–1.20	p-TBX	-	-		-	-
TBCT	-	-		-	-	TBCT	91	4.10	4.30	2.36	nd–13.8
PBBZ	100	3.01	3.03	1.81	0.0246–13.8	PBBZ	100	3.04	6.03	0.816	0.247–21.0
HBBZ	100	125	202	5.78	0.210–665	HBBZ	100	36.2	80.9	7.07	0.279–276
PBT	-	-		-	-	PBT	100	11.2	10.6	10.5	1.20–37.9
BATE	82	16.2	14.5	9.93	nd–63.5	BATE	-	-		-	-
PBEB	100	0.434	0.300	0.368	0.117–1.36	PBEB	64	0.190	0.145	0.233	nd–0.413
DPTE	47	1.45	1.66	1.02	nd–6.21	DPTE	73	10.4	12.7	7.03	nd–38.5
PBBA	49	0.222	0.120	0.183	nd–0.516	PBBA	27	1.76	2.45	0.493	nd–4.58
EHTBB	-	-		-	-	EHTBB	100	106	244	10.7	0.308–823
BTBPE	-	-		-	-	BTBPE	100	5.23	7.94	2.17	0.469–27.9
BEHTBP	67	3.61	13.2	0.387	nd–71.9	BEHTBP	100	92.5	96.0	43.6	6.81–244
DBDPE	-	-		-	-	DBDPE	55	368	297.5	325	nd–882
$\sum_{11}$NBFRs[b]		157	213	37.5	9.28–686	$\sum_{13}$NBFRs[d]		479	374.2	353	71.9–1160

"-" and nd means not detected

[a] $\sum_5$PBDEs is the sum of the BDE-28, 47, 66, 100, and 99

[b] $\sum_{11}$NBFRs is the sum of the α-TBECH, β-TBECH, ATE, p-TBX, PBBZ, HBBZ, BATE, PBEB, DPTE, PBBA, and BEHTBP

[c] $\sum_{10}$PBDEs is the sum of the BDE-28, 47, 66, 100, 99, 85, 153, 138, 183, and 209

[d] $\sum_{13}$NBFRs is the sum of the β-TBECH, β-TBCO, TBCT, PBBZ, HBBZ, PBT, PBEB, DPTE, PBBA, EHTBB, BTBPE, BEHTBP, and DBDPE

indoor environmental variables, such as temperature, RH, and ventilation, were analyzed in this study.

Temperature and RH The temporal variations in the PBDE and NBFR concentrations in BR1, BR2, and LR and the corresponding temperature and RH (average) are depicted in Figs. S3 and S4, respectively. The temporal variation of PBDE concentrations was similar to that of temperature and RH, especially for the temperature (Fig. S3).

The NBFR concentrations did not show obvious temperature dependence (Fig. S4). To determine the differences, the concentration and composition of NBFRs for A5–A8 were analyzed, and the results indicated that HBBZ and BATE led to the differences. Figure S5 shows that for $\sum_9$NBFRs, except for HBBZ and BATE, the temporal variations in the concentration coincided with those in the temperature. Most BFR concentrations were mainly affected by the temperature and RH. The Pearson correlation showed that the concentrations of $\sum_5$PBDEs and $\sum_9$NBFRs were significantly correlated with the temperature and RH ($p < 0.05$), respectively (Table S8). The correlation between concentration and temperature was attributed to the volatility of BFRs. The correlation between concentration and RH may be due to moisture inhibiting the sorption of BFRs in dust (Kuang et al. 2016).

The temporal variations in the BFR concentrations in this study were consistent with those in a previous study; notably, the PBDE concentrations in warmer months were generally higher than those in colder months during the monitoring period in indoor environments (Hazrati and Harrad 2006). In addition, the temporal variations in the penta- and octa-BDE levels in outdoor air were similar to those in ambient temperature (Ding et al. 2016).

Ventilation and other factors The concentrations of HBBZ and BATE increased significantly in A5–A8 (autumn and winter), possibly due to indoor ventilation in autumn and an increase in air temperature near heating pipes during the heating season. Studies have reported that ventilation increases the emission rate and sorption rate of pollutants by impacting the mass transfer coefficient and increases the passive sampling rate by reducing the thickness of the air-side boundary layer (Bartkow et al. 2005; Liagkouridis et al. 2014; Melymuk et al. 2011). The increased temperature leads to an increase in the emission rate of pollutants (Kajiwara and Takigami 2013; Kemmlein et al. 2003; Sun et al. 2018; Zhang et al. 2009). The results indicated that indoor ventilation and heating have an effect on the BFR concentrations in indoor air.

Source analysis

Principal component analysis (PCA) was used to investigate the possible sources of $\sum_5$PBDEs and 8 NBFR compounds (DF > 60%) in air. Figure S6 shows that 79.3% of the total variance was explained by 2 principal components, and the variances of the two factors are 56.3% and 23.0%. The correlation and rotated component matrix of the BFRs in air are presented in Tables S9 and S10, respectively. PC1 was dominated by α-TBECH, β-TBECH, and $\sum_5$PBDEs, and there was an extremely significant correlation among these factors ($p < 0.01$). TBECH and $\sum_5$PBDEs can both be used in textiles (Andersson et al. 2006). One sample collected from a futon contained penta-BDE (0.5% by weight) (Stapleton et al. 2009). PC2 showed high loading values in PBEB, BEHTBP, and ATE. Pearson correlation showed that BEHTBP and ATE were extremely significantly correlated ($p < 0.01$). The BEHTBP concentrations found in polyurethane foam ranged from 1.1 to 1.5% by weight (Stubbings et al. 2018). ATE can also be used in polyurethane foam (Covaci et al. 2011). In summary, the main indoor sources of BFRs in indoor air are textiles and polyurethane foam.

BFR occurrence in dust

Concentrations and profiles of BFRs in dust

The concentrations of BFRs in the total dust fraction calculated using Eq. (S1) are listed in Table 1. BATE, ATE, p-TBX, and α-TBECH were not detected in dust samples.

PBDEs $\sum_{10}$PBDEs ranged from 221 to 1060 ng g^{-1}, with a median of 447 ng g^{-1} and an average of 259 ng g^{-1}. The DF of PBDE congeners was higher than 70%, except for BDE-138 (45%). BDE-209 was the predominant congener in indoor dust, with a median of 235 ng g^{-1} and an average of 409 ng g^{-1}, constituting 79.9 to 99.3% of $\sum_{10}$PBDEs. BDE-28, 47, 66, 100, 99, 85, 153, and 138 were included in the calculation of $\sum_8$PBDE as penta-BDE, with a median of 17.3 ng g^{-1} and an average of 35.4 ng g^{-1}, respectively, constituting 0.6–19.2% of $\sum_{10}$PBDEs. BDE-183 (defined as octa-BDE) constituted 0.1–1.5% of $\sum_{10}$PBDEs. The concentrations of $\sum_{10}$PBDEs in this study were lower than those in Canada, the USA (Venier et al. 2016), Sweden (Newton et al. 2015), Australia (He et al. 2018a; McGrath et al. 2018), the UK (Tao et al. 2016), Turkey (Kurt-Karakus et al. 2017), Norway (Cequier et al. 2014), and other cities in China (Peng et al. 2017; Sun et al. 2016; Zheng et al. 2011) (Table S11).

NBFRs The concentrations of $\sum_{13}$NBFRs in indoor dust varied from 71.9 to 1160 ng g^{-1}, with a median of 353 ng g^{-1} and an average of 479 ng g^{-1}, respectively. The DF of individual NBFRs was higher than 60%, except for DBDPE (55%) and PBBA (27%). DBDPE, BEHTBP, and EHTBB (average concentration percentage ± std. dev.) were the predominant contaminants in indoor dust, accounting for 73% ± 19%, 23% ± 23%, and 14% ± 24% of $\sum_{13}$NBFRs, respectively. In addition, the value of EHTBB/BEHTBP among the dust samplers ranged from 0.01 to 6.1, with median and average values of 0.34 and 0.93, respectively. Compared with the NBFR concentrations measured in previous studies, the EHTBB and HBBZ concentrations in this study were higher than those in the Czech Republic, Sweden, and the UK, and the BEHTBP levels were lower than those in these countries (Newton et al. 2015; Tao et al. 2016; Venier et al. 2016) (Table S12).

Influential factors

Emission source and ventilation Studies have demonstrated that there are three main factors (the emission source, ventilation, and temperature) that influence the BFR concentrations in dust. Previous research observed that changes in BFR concentrations are related to variations in emission sources (Allen et al. 2008; Brommer et al. 2012; Liagkouridis et al. 2014; Peng et al. 2017; Takigami et al. 2008). In addition, studies have also found that indoor ventilation and temperature affect the levels of BFRs in indoor dust (Muenhor and Harrad 2012; Yu et al. 2012). In this study, no change existed in indoor emission sources, except for the replacement of one computer monitor during the monitoring period, and the dust sample in autumn was too small to be collected for analysis. Therefore, changes in emission sources and ventilation could be excluded.

Temperature The total concentrations of PBDEs and NBFRs measured in the test home floor dust during the monitoring period are shown in Fig. S7, and the highest concentrations of BFRs appeared from January to June 2018. There was a difference in BFR concentrations but no obvious temporal or temperature dependence. This result was similar to the results observed for PBDEs in other indoor dust samples (Brommer et al. 2012; Cao et al. 2014b; Muenhor and Harrad 2012). Pearson correlation showed that there was no significant correlation ($p > 0.05$) among the BFR concentrations, temperature, and RH in the test home (Table S13). BFRs migrate to indoor dust through the volatilize-adsorption and abrasion of indoor materials (Cao et al. 2015). Therefore, the sources of BFRs in dust are more complicated than those in air, resulting in a change in the BFR concentrations in dust due to the effects of the temperature and RH being less than those of other influential factors.

Source analysis

BFRs (DF > 60%) in indoor dust were used to determine the possible sources. It is worth noting that BDE-28, 47, 66, 100, 99, 85, and 153 were the major components of penta-BDE and they were extremely significantly correlated ($p < 0.01$). Figure S8 shows that 94.2% of the total variance was explained by two principal components, and the variances of the two factors were 77.4% and 16.8%, respectively. The correlations and rotated component matrix of BFRs in dust are presented in Tables S14 and S15, respectively. PC1 was dominated by HBBZ, EHTBB, BDE-183, BEHTBP, PBBZ, BTBPE, penta-BDE, BDE-209, PBT, and β-TBCO, and there were obviously significant correlations ($p < 0.01$) among them, except for BDE-209, PBT, and β-TBCO. EHTBB, BEHTBP, and penta-BDE are used in textiles and polyurethane foam. The DF of penta-BDE congeners exceeded 80% in textiles (Vojta et al. 2017). In 156 sofas, penta-BDE was detected at a frequency of 48% in all samples (Cooper et al. 2016). The use of EHTBB and BEHTBP is similar to penta-BDE, since penta-BDE has been replaced by FM-550 containing EHTBB and BEHTBP (Stapleton et al. 2008). According to previous studies, PBT and β-TBCO can be applied to polyurethane foam and HBBZ can be used in textiles (Covaci et al. 2011; Kurt-Karakus et al. 2017; Wang et al. 2018). BDE-183, BTBPE, and BDE-209 were found in electrical and electronic equipment (Vojta et al. 2017). PC2 showed high loading in TBCT and β-TBECH, indicating that they are from common sources. In summary, the main sources of BFRs in indoor dust are textiles, polyurethane foam, electrical equipment, and electronic equipment.

Distributions of BFRs among different size fractions

Dust mass percentage

Figure S9 shows that the average dust mass percentage of F6 was the highest, accounting for 34% of the total dust; the lowest dust mass percentage corresponded to F1, accounting for 8%. Additionally, the dust mass percentages of F2, F3, F4, and F5 were comparable. According to the relevant literature, considering the lack of human activities in the test home, the main sources of F5 and F6 were infiltration, deposition, and the loss of abrasion-resistant materials (Cao et al. 2014a; Cousins et al. 1999; Weschler et al. 2008). F1, F2, and F3 were mainly caused by the abrasion of soft materials, wind, and outdoor dust carried into the room by people.

Concentration percentage

The distributions of the PBDE and NBFR concentration percentage in each particle size are shown in Fig. 1, and those of the BFR concentration percentage are shown in Fig. S10. The average concentration percentages of PBDEs and NBFRs in F6 were the highest, accounting for 28% and 33% of the total dust, respectively, followed by those in F2, F5, F4, F3, and F1. The PBDE and NBFR concentrations in F2 were higher than those in other particle sizes, except for F6, indicating that the concentrations of BFRs had no inverse relation to the particle size. The migration of pollutants to indoor dust caused by the abrasion of soft materials may be the main reason for the increase in the BFR concentrations in F2. The reasons why the concentrations of BFRs in F3–F6 increased with decreasing particle size are mainly as follows. First, BFRs in the gas phase tend to accumulate in fine particles with a large specific surface area (Lewis et al. 1999). Second, this behavior may be caused by the loss of abrasion-resistant materials (Cao et al. 2014a). In a previous study, the PBDE concentrations in cars appeared to increase with decreasing dust particle size, but the concentrations in homes did not (Wei et al. 2009).

Detailed information on the BFR concentrations for each particle size is presented in Table S16. For each particle size, BDE-209, DBDPE, and BEHTBP were the main pollutants. The concentration changes of each pollutant within F1–F6 suggested that the trends of most BFR individual concentrations within different particle sizes were the same as those of the PBDEs and NBFRs, except for β-TBCO, PBT, BTBPE, and BDE-183 (Fig. S11). The concentrations in F5 were the highest, suggesting that surface adsorption might not be the

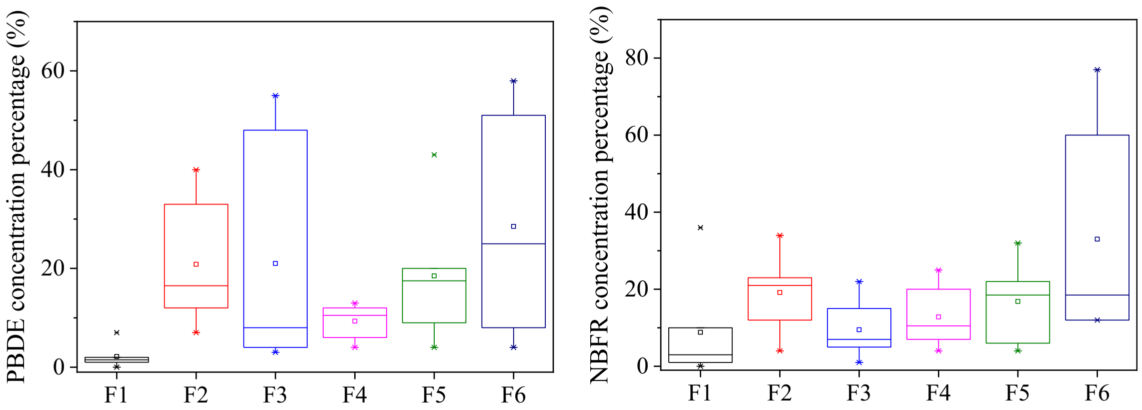

Fig. 1 The concentration percentages of PBDEs and NBFRs in indoor different particle sizes

driving mechanism of these pollutants. Through Pearson correlation analysis, BTBPE and BDE-183 in F5 were extremely significantly correlated ($p < 0.01$), but β-TBCO and PBT were not correlated ($p > 0.05$). This implied that BTBPE and BDE-183 were from the same pollution source, and β-TBCO and PBT had similar release patterns. In a previous study, for $\sum_7$Tri-hepta-BDEs and BEHTBP, concentrations were higher in finer dust particles, but not for BDE-209 and DBDPE (Al-Omran and Harrad 2016).

Dust-air partitioning

BDE-28, BDE-47, BDE-66, BDE-99, BDE-100, β-TBECH, PBBZ, HBBZ, PBEB, and BEHTBP were detected in dust and air. Therefore, in this study, the concentrations of these compounds collected in BR1, BR2, and LR were used in Eq. (S2–S8) (Harner et al. 2001; Weschler and Nazaroff 2010; Yadav et al. 2018) to describe the dust-air partitioning and exchange of BFRs within F1–F6 and FA in the test home. The median values of $\log K_D$ and *ff* are provided in Table S17.

BFR distribution between indoor dust and air

The measured median $\log K_D$ values of PBDEs and NBFRs within different particle sizes ranged from −0.606 to 5.63 and from −1.97 to 6.33, respectively (Table S17). For HBBZ, BDE-47, BDE-66, BDE-99, BDE-100, and BEHTBP, the $\log K_D$ values increased with decreasing particle size (Fig. S12). For β-TBECH, PBBZ, BDE-28, and PBEB, the $\log K_D$ values of F2 were higher than those of F3 (Fig. S12). For each particle size, the $\log K_D$ of BFRs increased with increasing $\log K_{OA}$ (Fig. S13), suggesting that BFRs with high $\log K_{OA}$ values tended to accumulate in indoor dust, which may be the main storage source for long-term migration.

The relationships between the $\log K_D$ values of BFRs for different particle sizes and the $\log K_{OA}$ value for each compound were estimated, as shown in Fig. 2. The median values of $\log K_D$ within F1–F6 and FA were linearly associated with $\log K_{OA}$, and they were correlated (Table S18). Similar relationships for the semi-volatile organic compounds (SVOCs) were detected in indoor environments in the USA, Norway, Australia, and China (Cequier et al. 2014; Dodson et al. 2015; He et al. 2018a; Li et al. 2019). Compared with the slope of 1 ($\log K_D = \log K_{OA} - 7$) predicted from equilibrium partitioning (Weschler and Nazaroff 2010), the slopes of the linear fit function based on the actual concentrations were 0.2 and 0.3 in this study. These results indicate that for BTBPE, with a high $\log K_{OA}$ (16.9) value, the measured concentrations within F1–F6 and FA were all significantly lower than those predicted at equilibrium. This result might be because the dust-air partitioning of BFRs with high $\log K_{OA}$ values within F1–F6 and FA was at steady-state and could not achieve equilibrium in the indoor environment. The non-equilibrium state was similar to the gas-particle partitioning steady-state model of PBDEs observed in global air (Li et al. 2015). Furthermore, previous studies have demonstrated that the steady-state model is also applicable to the dust-air partitioning of SVOCs in different microenvironments (Li et al. 2019; Venier et al. 2016; Vykoukalova et al. 2017; Zhang et al. 2011).

For β-TBECH, which had the low $\log K_{OA}$ (8.01) value, the measured concentrations in F1–F6 and FA were higher than those predicted at equilibrium, which might be attributed to two factors. First, the β-TBECH concentrations in dust were overestimated due to the abrasion of materials containing β-TBECH. Second, the β-TBECH concentrations in air were underestimated because the PUF-air partitioning of β-TBECH did not achieve equilibrium (Li et al. 2019).

The $\log K_D$ values of BFRs ($\log K_{OA} = 9.1$–11.32) were similar to the $\log K_D$ at equilibrium, especially within F5, F6, and FA, suggesting that the distribution between indoor air and dust tended to achieve equilibrium for these particle sizes. Dust-air partitioning was more likely to be the main source of BFRs in F5 and F6, since BFRs in the gas phase tend to accumulate in fine particles. In addition, the BFR concentrations in F5 and F6 were higher, implying that they dominate the BFR concentrations in FA. Therefore, the distribution of

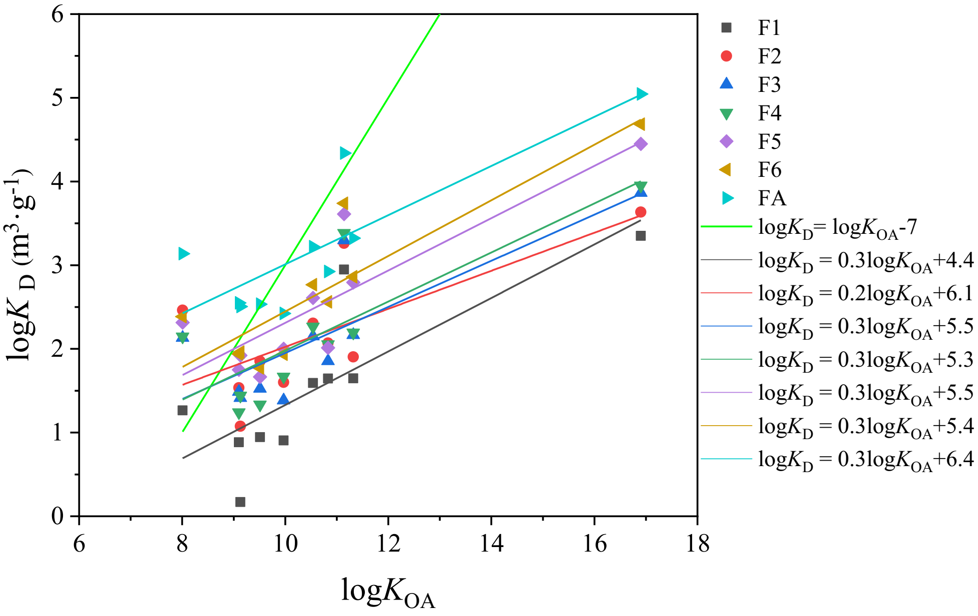

Fig. 2 Scatterplot of the median logK_D of BFRs in different particle sizes against the corresponding logK_{OA}

BFRs (logK_{OA} = 9.1–11.32) between indoor air and dust approximately reached equilibrium in FA, and similar behavior was observed for F5 and F6.

BFR exchange between indoor dust and air

The dust-air exchange of BFRs is an important process that affects the dust-air partitioning in indoor environments. The fugacity fraction (*ff*) is applied to characterize the pattern of dust-air exchange. For different particle sizes, the range of *ff* values of β-TBECH (logK_{OA} = 8.01) was 0.484–0.983; for BFRs (logK_{OA} = 9.1–16.9), the median values of *ff* ranged from 0.005 to 0.872, and the *ff* values of BEHTBP (logK_{OA} = 16.9) were all far from 0.5 (Table S17).

When *ff* = 0.5, the BFR dust-air exchange reaches equilibrium; when *ff* > 0.5, the BFRs mainly enter the air from dust; under other conditions, the BFRs mainly enter the dust from air. The values of *ff* in the range of 0.3–0.7 reflect an equilibrium state due to the approximations used in the calculations (Harner et al. 2001).

The *ff* values of each compound within F1–F6 are shown in Fig. S14. Only β-TBECH exhibited *ff* values much higher than 0.5 in F2–F6, indicating that indoor dust was "saturated" relative to air. Hence, β-TBECH moved from indoor dust to air, where the indoor dust was temporarily stored, causing secondary pollution in the indoor environment. The values of *ff* for most BFRs within each particle size were less than 0.5, indicating that most of these chemicals mainly moved from indoor air to dust. Therefore, they tended to accumulate in indoor dust or achieve dust-air equilibrium. Once deposited, they can cause environmental hazards for a long time. The dust-air exchange status in this study was similar to the soil-air exchange status of BFRs in Tianjin, China (Hong et al. 2018). For each particle size, the dust-air exchange status of the BFRs was similar, implying that particle size has no significant effect on dust-air exchange.

Through Pearson correlation analysis, the *ff* values for different particle sizes had no correlation ($p > 0.05$) with the corresponding temperature. However, temperature had an influence on the dust-air exchange of SVOCs (Wei et al. 2019). In a previous report, the median *ff* values of 2,4,4′-trichlorobiphenyl (PCB-28) and R-HCH identified them as more volatile compounds that decreased during the cold season (Ruzickova et al. 2008). In this study, there was no correlation between *ff* values and temperature, which may be due to the small magnitude of indoor temperature changes or insufficient data. Therefore, the relationship between these factors still needs to be further studied.

Conclusion

BDE-28 and HBBZ were the main pollutants in indoor air, and BDE-209, BEHTBP, and EHTBB were the main pollutants in indoor dust. The concentrations of $\sum_5$PBDEs and $\sum_9$NBFRs in indoor air showed significant temperature dependence. The BFR concentrations in air were affected by the temperature, RH, and ventilation. However, temperature dependence was not observed for BFR concentrations in indoor dust. Textiles, polyurethane foam, electrical equipment, and electronic equipment were the main sources of BFRs in the test home. The mass percentage of BFRs in F6 was the highest, suggesting that indoor dust was mainly composed of F6. Moreover, the BFR concentrations did not increase constantly with the decreasing particle size, and the values in F2 were higher than those in F3. The partitioning of BFRs

($\log K_{\mathrm{OA}} = 9.1$–$11.32$) between indoor air and dust approximately approached equilibrium within F5, F6, and FA. In addition, for BFRs ($\log K_{\mathrm{OA}} = 9.1$–$16.9$), the direction of the net flux was mainly from indoor air to dust for F1–F6. This test home case study indicated that factors such as environment-specific characteristics, particle size, and dust-air partitioning are important for understanding the pollution characteristics of BFRs in indoor environments. The results might not be applicable to all indoor microenvironments, but they are consistent with those of previous studies.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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