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# Novel brominated flame retardants in house dust from Shanghai, China: levels, temporal variation, and human exposure

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## Abstract

**Background:** Novel brominated flame retardants (NBFRs) have been increasingly used as alternatives to legacy BFRs (e.g., PBDEs and HBCDs) in consumer products, but are liable to emigrate and contaminate indoor dust. In this study, a total of 154 house dust samples including floor dust (FD) and elevated surface dust (ESD) were collected in the biggest metropolitan area (Shanghai) of East China in 2016. Limited information about temporal variation of NBFRs indoors is available, while the period of sampling is influential in human exposure estimates. Levels, temporal variation, and human exposure of seven target NBFRs such as decabromodiphenylethane (DBDPE), 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), 2-ethylhexyl 2,3,4,5-tetrabromobenzoate (EHTBB), and bis(2-ethylhexyl) tetrabromophthalate (BEHTEBP) were investigated in indoor house dust.

**Results:** Concentrations of  $\sum_7$ NBFRs ranged from 19.11 to 3099 ng/g with a geomean of 295.1 ng/g in FD, and from 34.74 to 404.6 ng/g with a geomean of 117.9 ng/g in ESD. The geomeans of DBDPE were 219.6 ng/g in FD and 76.89 ng/g in ESD, accounting for 90.5% and 80.5% of  $\sum_7$ NBFRs. Levels of EHTBB, BTBPE, and DBDPE in FD exceeded significantly those in ESD. The temporal variation in  $\sum_7$ NBFRs in FD was ranked as summer > winter > autumn > spring. The daily exposure doses (DEDs) of  $\sum_7$ NBFRs via dust ingestion decreased as: infants > toddlers > children > teenagers > adults. Infants showed the highest DED in FD, 9.1 ng/kg bw/day.

**Conclusions:** DBDPE clearly dominated the NBFRs in both FD and ESD, but the concentrations of DBDPE in this study were generally moderate compared with the other international studies. Dust ingestion was the major pathway of human exposure to NBFRs indoors. About eightfold difference in exposure estimates between infants and adults showed that infants faced elevated exposure risks in FD. This study highlighted the necessity to estimate human exposure of NBFRs for different age groups using FD and ESD, respectively.

**Keywords:** Novel brominated flame retardants, Floor dust, Elevated surface dust, Temporal variation, Human exposure

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## Introduction

Brominated flame retardants (BFRs) are commonly used to inhibit or suppress combustion for industrial and commercial applications (e.g., building materials, furniture, textiles, and e-products) [1]. Over the past decades, there have been of great concern over the prevalence and environmental fate of some legacy BFRs such as polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs). Due to their persistent, bioaccumulative, toxic, and long-range transportable properties [2–6], Tetra- through Hepta-BDEs, Deca-BDE, and HBCDs had been listed in Annex A of the Stockholm Convention as persistent organic pollutants (POPs) for elimination by 2017 [7]. To meet flammability standards for goods and materials, the usage of alternative or novel brominated flame retardants (NBFRs) has rapidly increased in recent years [8]. Decabromodiphenylethane (DBDPE), as an alternative to the Deca-BDE formulation, has been produced with a capacity of 12,000 tons in China (2006), and its production increases at 80% per year [8]. 1,2-Bis(2,4,6-tribromophenoxy) ethane (BTBPE) is used as a replacement for Octa-BDEs in ABS, thermoplastics, and textile applications [9]. 2-Ethylhexyl 2,3,4,5-tetrabromobenzoate (EHTBB) and bis(2-ethylhexyl) tetrabromophthalate (BEHTEBP) are replacements for penta-BDEs, exist in the commercial mixtures such as Firemaster 550, BZ-54 and DP-45, which is mostly used in polyurethane foam (PUF) applications [10, 11]. Pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), and hexabromobenzene (HBB) are often used with the other flame retardants (FRs) in polyesters [12]. Similar to PBDEs, these NBFRs were reported with semi-volatile and non-reactive properties [13, 14], and were constantly detected in environmental matrices. Moreover, NBFRs like EHTBB, BEHTEBP, BTBPE, and DBDPE display potential adverse environmental behaviors; for example, persistence and bioaccumulation [15, 16]. Animal studies showed that EHTBB, BEHTEBP, BTBPE, and DBDPE posed the potential health risks of endocrine disruption, hepatotoxicity, impaired reproductive physiology, and gene expression [17–24].

Recently, the occurrences of NBFRs have been increasingly investigated in indoor dust from various countries, including USA [25, 26], Canada [27], UK [17, 28, 29], Belgium [30], Norway [31, 32], Spain [33], Sweden [34, 35], Australia [13], New Zealand [1], China [36–38], Pakistan [39], and South Africa [40]. In China, EHTBB, BEHTEBP, BTBPE, and DBDPE were frequently detected in indoor dust samples from e-waste sites in South China [37] and occupational areas in Beijing [38, 41]. However, information about house indoor contamination with NBFRs in Shanghai remains scant. In a previous study, considerable DBDPE concentrations (100–9500 ng/g dw) were

detected only in floor dust (FD) samples from 15 dwellings in East China, while elevated surface dust (ESD) samples were not fully considered. [36]. Oral ingestion and dermal absorption of house dust are considered as the major pathways of BFR exposure, accounting for 56–77% of the total intake [42]. Dust ingestion is a more important exposure pathway for young infants and toddlers due to their frequent hand-to-mouth behaviors [43]. Infants and toddlers may have more contacts with FD, while older age groups (children, teenagers, and adults) may face more NBFR exposure risks from the ESD on furniture and household electric appliances. As a whole, it will be more accurate to individually estimate human exposure risk of NBFRs for five different age groups in FD and ESD.

There is insufficient information regarding temporal variation of NBFRs indoors, while the period of sampling is influential in human exposure estimates. In a previous British study, over a 9-month sampling period, higher concentrations of PBEB, EHTBB, BEHTEBP, and BTBPE were observed in indoor dust of warmer seasons, but elevated levels of DBDPE were observed in colder seasons, which might be attributed to the low vapor pressure that facilitated partitioning to indoor dust [28]. Over a 10-month sampling period, Cao et al. [41] reported that NBFR (EHTBB, BEHTEBP, BTBPE, and DBDPE) levels in office dust of China were generally constant among different seasons. To our knowledge, no firm conclusions on temporal variation of NBFRs in indoor dust are available in the literature.

In the present study, the occurrence of seven NBFR compounds was investigated in FD and ESD samples of Shanghai, China. Our goals of this study were to: (1) determine the concentrations of seven target NBFRs in both FD and ESD; (2) compare the difference of NBFRs between FD and ESD; (3) understand the temporal variation of NBFRs in FD; (4) estimate human exposure to NBFRs via dust ingestion and dermal absorption in five age groups.

## Materials and methods

### Chemicals and standards

Individual standards of PBT, PBEB, HBB, EHTBB, BEHTEBP, BTBPE, DBDPE, and PBDE congeners (BDE-77, -128, -138) were purchased from AccuStandard, Inc. (New haven, CT). BDE-77 and BDE-138 were used as surrogate standards in this study. <sup>13</sup>C-decabromodiphenylether (<sup>13</sup>C-BDE-209) was provided by Cambridge Isotope Laboratories, Inc. (Andover, USA) and used as an internal standard for DBDPE. BDE-128 was used as an internal standard for other six NBFR compounds and two surrogate standards (BDE-77, -138). Additional file 1: Table S1 lists physical properties of these chemicals. All

solvents used during the analysis were of pesticide grade. Silica gel (100–200 mesh) was acquired from Merck (Darmstadt, Germany).

### Sample collection

From 22 dwellings in Shanghai, a total of 154 dust samples, including 132 FD samples and 22 ESD samples, were collected in 2016 (Fig. 1). More details about sampling information were presented in Additional file 1. Briefly, FD samples were collected separately from the living room, washroom, kitchen, and bedrooms of each home at the same time. To provide sufficient dust mass for analysis, FD samples from each home every second month were then mixed up to obtain one composite FD sample. In terms of ESD samples, they were collected from the elevated surface of shelves, tables, or other furnishings (typically 0.5–2 m height). All collected dust samples were sieved with pre-cleaned and dried stainless steel sieves with 500  $\mu\text{m}$  diameter of mesh, and thereafter, the sieved dust samples were homogenized thoroughly and stored in aluminum foil at  $-20\text{ }^{\circ}\text{C}$  until extraction.

### Dust extraction and purification

Analysis of target NBFRs in house dust was conducted following the similar extraction and cleanup methods as reported elsewhere with minor modifications [44]. Accurately weighted aliquots of house dust ( $\sim 150\text{ mg}$ ) were spiked with a mixture of surrogate standards (BDE-77 and BDE-138) in *n*-hexane, extracted by ultrasonication with 25 mL acetone/*n*-hexane (1:1, v/v) for three times (20 min each time), and then, the supernatant was evaporated by a rotary vacuum evaporator (R-210/215, BUCHI Labortechnik). Evaporated extracts were reconstituted with 3 mL *n*-hexane and further concentrated to incipient dryness under a gentle nitrogen stream. The purification was carried out using a Pasteur pipette packed with

silica gel (0.1 g neutral bottom layer/0.9 g acidic top layer-containing 0.3 g concentrated sulfuric acid). Columns were conditioned with 3 mL *n*-hexane; the extracts were loaded and then eluted with 10 mL dichloromethane/*n*-hexane (1:1, v/v). The purified extracts were blown up to incipient dryness, before resolubilisation in 200  $\mu\text{L}$  *n*-hexane containing BDE-128 and  $^{13}\text{C}$ -BDE-209 at 200  $\text{pg}/\mu\text{L}$  ready for instrumental analysis.

### Instrumental analysis

Analysis of target NBFRs was performed using Agilent 7890A gas chromatography coupled to a 5975C mass spectrometry (GC/MS). The GC was equipped with a programmable temperature vaporizer (PTV) injector and DB-5MS capillary column (15 m  $\times$  0.25 mm i.d.  $\times$  0.10  $\mu\text{m}$  film thickness; Agilent J&W). 1  $\mu\text{L}$  purified extracts were automatically injected in splitless mode at an injector temperature of 280  $^{\circ}\text{C}$ . Negative chemical ionization (NCI) with helium as carrier gas (1.5 mL/min) was used, and the MS was operated in the selected ion-monitoring (SIM) mode. The ion source, quadrupole, and transfer line temperatures were set at 250, 150, and 290  $^{\circ}\text{C}$ , respectively. The oven temperature was initially held at 110  $^{\circ}\text{C}$  for 1.5 min, ramped 20  $^{\circ}\text{C}/\text{min}$  to 200  $^{\circ}\text{C}$  held for 1 min, and then ramped 10  $^{\circ}\text{C}/\text{min}$  to 310  $^{\circ}\text{C}$ , and held for 5 min. The identification of target compounds was based on their retention times and the relative intensity of the fragment ions. The selected fragment ions ( $m/z$ ) were as follows: 495/497 for  $^{13}\text{C}$ -BDE-209, 384/462 for BEHTEBP, 357/359 for EHTBB, and 79/81 for other target compounds.

### Quality assurance and quality control

All glasswares used in the laboratory were washed thoroughly with detergent, rinsed with distilled water, and dried and baked at 400  $^{\circ}\text{C}$  for 5 h. Silica gel was activated at 300  $^{\circ}\text{C}$  for 12 h prior to use. To prevent photolytic degradation of the target compounds, ultraviolet-light protection was mounted on windows, and the glassware was covered with aluminum foil whenever possible. To assess any possible contamination during the process of sample treatment and analysis, one laboratory blank was processed in parallel with every set of eight dust samples. Blank correction was performed by subtracting the mean value detected in the laboratory blanks from the same batch. Spike/recovery experiments ( $n = 3$ ) involving fortification of seven target compounds produced satisfactory recovery rates (Table 1), demonstrating good method accuracy. Seven-point calibration plots were created for quantification and calibration curves with reliable regression coefficients ( $> 0.997$ ) were obtained. Determination of the limit of detection (LOD) and limit of quantification (LOQ) was performed according to Newton et al.



**Fig. 1** The sampling sites of indoor dust in Shanghai

**Table 1** Quality assurance and quality control results

	PBT	PBEB	HBB	EHTBB	BTBPE	BEHTEBP	DBDPE	References
Limit of detection (LOD)	0.02	0.03	0.12	0.09	0.21	0.06	0.69	This study
Limit of quantification (LOQ)	0.06	0.08	0.43	0.31	0.74	0.21	2.3	This study
Average in blanks	na	na	0.32	0.23	0.51	0.12	2.1	This study
Blank detection (%)	na	na	67	70	90	72	96	This study
Recovery in spiked blanks (%)	92 (5)	93 (4)	95 (4)	93 (7)	97 (5)	90 (3)	101 (5)	This study
Mean (SD) concentration in SRM 2585	na	1.5 (2.2)	na	35 (4.3)	43 (10)	312 (121)	<5	This study
	na	na	na	<30	<0.8	145 (17)	<10	[11]
	na	na	na	40	32	652	<20	[45]
	na	na	na	26 (2)	39 (14)	574 (49)	<7.1	[46]
	na	na	na	36 (2.4)	39 (4.9)	1300	<10	[47]
	na	na	na	35 (6)	76 (4)	857 (73)	na	[48]
	na	8.1 (2.0)	na	34 (5.2)	53 (9.2)	869 (110)	<6	[29]

na not detected or available

[35]. For the analytes not present in blanks, LOD was estimated based on a signal-to-noise ratio (S/N) 3:1 and LOQ was set to S/N=10. For compounds detected in blanks, the LOD and LOQ were defined as three times the mean values plus 3 and 5 standard deviations, respectively (Table 1). As a further quality control procedure, an indoor dust Standard Reference Material (SRM 2585, National Institute of Standards and Technology, Gaithersburg, MD;  $n=3$ ) was processed for target NBFRs, concentrations of NBFRs in SRM 2585 were in agreement with those reported in the previous studies (Table 1). The recoveries of surrogate standards were  $82 \pm 12\%$  for BDE-77,  $89 \pm 10\%$  for BDE-138. However, the reported concentrations in this study were not corrected with surrogate recovery.

#### Data analysis

The statistical analyses of the data were performed using Microsoft Excel 2011 and GraphPad Prism 6 (GraphPad Software, La Jolla, USA). Any differences in NBFR concentration between two dust categories and between seasons (or months) were examined using paired-samples *T* test and one-way analysis of variance (ANOVA), respectively. Shapiro–Wilk normality test was conducted to confirm our data with skewed distribution, and thereafter, data were log-transformed prior to *T* test or ANOVA. The two-tailed Spearman's correlation coefficient was calculated to analyze the correlations between the variables. A *P* value of less than 0.05 was defined as statistical significance.

## Results and discussion

### Concentrations of NBFRs in FD and ESD

The concentrations of seven NBFRs (PBT, PBEB, HBB, EHTBB, BEHTEBP, BTBPE, and DBDPE) were

determined in FD and ESD. Table 2 lists detection frequency, minimum, maximum, median, mean, and geomean concentrations detected with respect to target NBFRs in ng/g of dw house dust. Our results indicated that BTBPE and DBDPE were detected in all dust samples, while detection frequencies of PBT, PBEB, HBB, EHTBB, and BEHTEBP were between 55.2 and 96.8%. The concentrations of  $\sum_7$ NBFRs in FD ranged from 19.11 to 3099 ng/g with a geomean of 295.1 ng/g, while the levels of  $\sum_7$ NBFRs in ESD ranged from 34.74 to 404.6 ng/g with a geomean of 117.9 ng/g. The geomean levels of DBDPE in two dust categories were generally one or two orders of magnitude higher than the second dominant compound BTBPE. The concentrations of NBFR compounds in FD were ranked as DBDPE  $\gg$  BTBPE > HBB > EHTBB > BEHTEBP > PBT > PBEB, while the similar trend in ESD was found as DBDPE  $\gg$  BTBPE > EHTBB > HBB > BEHTEBP > PBT > PBEB. Seven target NBFR compounds were discussed individually in the following parts.

### DBDPE

The concentrations of DBDPE ranged from 14.70 to 2931 ng/g with a geomean of 219.6 ng/g in FD and ranged from 16.90 to 349.4 ng/g with a geomean of 76.89 ng/g in ESD, respectively. DBDPE was the predominant compound of target NBFRs in house dust samples of this study, contributing to 90.5% and 80.5% of total NBFRs in FD and ESD. The domination of DBDPE in indoor dust was in line with the other reports in China [36, 49, 50]. Compared with PBDEs in our previous study [51], DBDPE concentrations rivaled those of Deca-BDE, revealing that DBDPE has been widely used in China following the phase-out of PBDEs. Peng et al. [36] reported higher concentration range of DBDPE in indoor dust from Shanghai (100–9500 ng/g), but the concentration

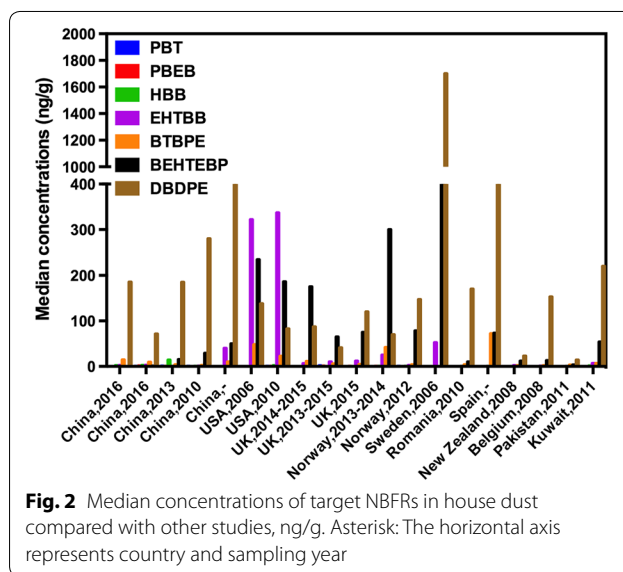
**Table 2** Statistical data of NBFR concentrations (ng/g) in two dust categories

Dust category/compound	PBT	PBEB	HBB	EHTBB	BTBPE	BEHTEBP	DBDPE	Σ <sub>7</sub> NBFRs
FD sample (n = 132)								
Detection (%)	93.5	88.9	96.8	63.6	100	55.2	100	–
Minimum	<LOD	<LOD	<LOD	<LOD	0.58	<LOD	14.70	19.11
5th Percentile	0.06	0.04	0.34	0.17	2.97	0.11	34.68	43.33
Median	0.32	0.20	2.13	2.26	14.49	0.13	185.4	290.8
95th Percentile	2.81	2.35	17.14	59.37	184.5	53.75	1571	1630
Maximum	25.77	7.00	27.32	143.7	526.1	224.0	2931	3099
Mean	0.96	0.45	3.65	12.64	42.68	12.1	419.4	491.8
Std. deviation	2.83	0.90	4.78	23.76	84.03	28.50	575.4	579.7
Geomean	0.38	0.21	2.11	1.85	17.26	0.84	219.6	295.1
ESD sample (n = 22)								
Detection (%)	77.3	81.8	100	90.9	100	63.6	100	–
Minimum	<LOD	<LOD	0.61	<LOD	0.37	<LOD	16.90	37.74
5th Percentile	0.03	0.04	0.61	0.17	0.37	0.11	16.9	37.74
Median	0.59	1.20	2.41	2.80	9.30	0.98	71.43	104.8
95th Percentile	7.87	7.87	16.9	26	37.88	45.43	349.4	404.6
Maximum	7.87	7.87	16.9	26.10	37.88	45.43	349.4	404.6
Mean	1.31	1.81	4.00	6.58	14.02	8.09	102.5	136.9
Std. deviation	2.03	2.15	4.34	7.04	11.48	12.36	88.05	88.28
Geomean	0.45	0.73	2.60	3.24	9.59	1.17	76.89	117.9

range was still in the same order of magnitude with this study. Qi et al. [50] also reported the comparable median level of DBDPE in indoor dust across China (280 ng/g). Compared with the international studies, the median level of DBDPE in this study (185.4 ng/g) was an order of magnitude lower than that in Swedish indoor dust (1700 ng/g) [47]. However, the median concentration of DBDPE in USA [25], UK [29] and Norway [31] was 2.2, 2.1 and 2.6 times lower than that in this study. The levels of DBDPE in house dust detected from different countries varied in two orders of magnitude; DBDPE concentrations in this study were generally moderate, while DBDPE in house dust from Sweden [47] and Spain [33] exhibited elevated concentrations (Fig. 2; Additional file 1: Table S2), probably due to different fire safety regulations or living habits (e.g., ventilation time, the amount of e-products).

**BTBPE**

BTBPE was the second dominant compound of target NBFRs, ranging from 0.58 to 526.1 ng/g with a geomean of 17.26 ng/g in FD and ranging from 0.37 to 37.88 ng/g with a geomean of 9.59 ng/g in ESD, respectively. In this study, the full detection frequency of BTBPE indicates the extensive use of this emerging compound in indoor environment. The median level of BTBPE (14.49 ng/g) was very comparable to that in USA [25], UK [29], and Pakistan [39], but was three-to-five times lower than reported



**Fig. 2** Median concentrations of target NBFRs in house dust compared with other studies, ng/g. Asterisk: The horizontal axis represents country and sampling year

levels in Norway [31] and Spain [33]. As a whole, the concentrations of BTBPE presented in this study were of middle level compared with the international studies (Fig. 2; Additional file 1: Table S2). In the previous studies of China, BTBPE in indoor dust showed very comparable levels to this study, about 10–20 ng/g, and concentrations of BTBPE were also one order of magnitude lower than DBDPE in these studies [36, 50].



### **EHTBB and BEHTEBP**

EHTBB and BEHTEBP were detected in more than half of the house dust samples. EHTBB was, in general, more frequently detected than BEHTEBP in both FD and ESD. ESD samples showed higher concentrations of EHTBB and BEHTEBP, with a geomean of 3.24 ng/g and 1.17 ng/g, respectively. Geomean concentrations of EHTBB were 2.2 and 2.8 times higher than BEHTEBP in FD and ESD. Compared with the other regions, levels of EHTBB and BEHTEBP in this study were about one or two orders of magnitude lower than those in USA [11, 25] and some European countries such as UK [17], Sweden [47], and Norway [31], whereas they were very close to those in New Zealand [1] and Pakistan [39], indicating that Europe and USA have apparently a higher consumption of EHTBB and BEHTEBP, to comply with different fire safety standards [12, 52, 53].

Of particular interest to note was the ratio of EHTBB/BEHTEBP. Three commercial mixtures Firemaster 550, Firemaster BZ-54, and DP-45 contain about 35% (15%), 70% (30%), and 0% (100%) of EHTBB and BEHTEBP [54]. Another study reported that the ratio of EHTBB/BEHTEBP in Firemaster 550 is approximately 4:1 by mass [11]. Firemaster 550 might have ratios of EHTBB/BEHTEBP between 2.33 (35%:15%) and 4. However, among the dust samples analyzed the ratio ranged from 0.01 to 80.3 with a geomean of 2.20 in FD, while the ratio range was from 0.08 to 122 with a geomean of 2.77 in ESD. The wide ratio range in house dust may reflect various sources of these emerging compounds in indoor environment; for example, the household electric products and building materials which contain different commercial mixtures of EHTBB and BEHTEBP. Besides, EHTBB and BEHTEBP have both been reported to undergo sequential reductive debromination [10]. The photodegradation of BEHTEBP may result in the formation of tetrabrominated analogs, which have similar properties as di(2-ethylhexyl) phthalate (DEHP), and then may influence the ratios of EHTBB/BEHTEBP in collected dust samples. Moreover, there was possible breakdown of BEHTEBP during cleanup due to the breaking of two ester bonds, which affected the fate of BEHTEBP. Thus, the degradation of these emerging compounds may be another influencing factor to elucidate the wide ratio range of EHTBB/BEHTEBP in this study. Further investigations need doing to draw firm conclusions.

### **HBB**

In this study, HBB was detected in almost all the dust samples (>96.8%). The levels of HBB were comparative to EHTBB, ranging from <LOD to 27.32 ng/g with a geomean of 2.11 ng/g in FD and from 0.61 to 16.9 ng/g with a geomean of 2.60 ng/g in ESD, respectively. Under

the similar conditions (e.g., sampling month), the median concentrations of HBB were approximately fourfold higher than those in 23 provinces across China [50], there might be the possibility of HBB being increasingly used as an alternative BFR in indoor environment over the past few years. From another perspective, HBB could be considered as decomposition product derived from the pyrolysis of polymeric BFRs, as reported by Gouteux et al. [55], which could be another possible source of HBB indoors. Further work is needed to assess the release of HBB from polymeric BFRs.

### **PBEB**

Levels of PBEB in this study ranged from <LOD to a maximum of 7.00 ng/g in FD and from <LOD to a maximum of 7.87 ng/g in ESD. The geomean concentration of PBEB in FD was one order of magnitude lower than HBB and EHTBB, but it was very similar to PBT. In ESD, the geomean value of PBEB was approximately 3.6 and 4.4 times lower than HBB and EHTBB, while it was slightly higher than PBT. Compared with the previous studies in USA [25], UK [17, 56], and Norway [31], the levels of PBEB in China were very close to them, mostly below 1 ng/g (Fig. 2; Additional file 1: Table S2).

### **PBT**

PBT was detected in over 77.3% of all the dust samples. One of FD samples showed the highest concentration of PBT at 25.77 ng/g, and the maximum of PBT in ESD was 7.87 ng/g. The geomean levels of PBT in FD and ESD were both below 0.5 ng/g, 0.38 ng/g, and 0.45 ng/g, respectively. Like PBEB, levels of PBT in house dust of the other regions were generally at low levels [17, 25, 32, 50], and house dust of UK showed the marginally higher level of PBT, about 1.8 ng/g (Additional file 1: Table S2).

### **Comparison and correlation of NBFRs between FD and ESD**

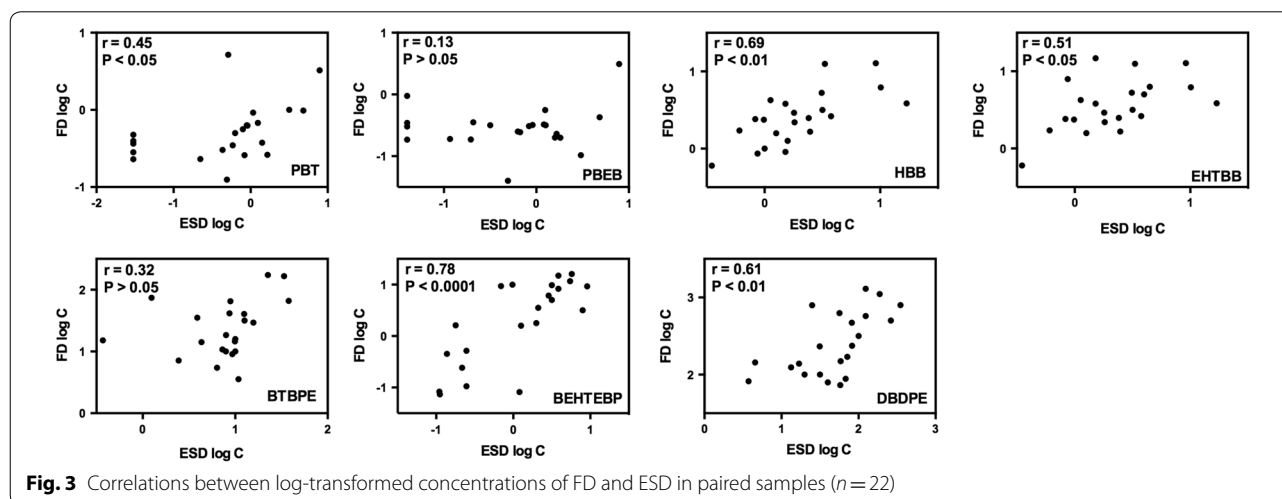
In this study, the difference of NBFRs between FD and ESD was compared testing whether it was necessary to separately collect two dust categories for analyzing NBFRs, and then for further estimating human exposure risks in different age groups. Following log transformation of concentrations expressed on a dry dust weight basis, paired-samples *T* tests were applied to test whether concentrations of target NBFRs in FD would exceed those in ESD ( $n=22$ ). Concentrations of EHTBB, BTBPE, and DBDPE in FD exceeded significantly those in ESD, with ( $P<0.05$ ) 0.0169, 0.008, and <0.0001 respectively. Al-Omran and Harrad [28] reported that spatial variations in BFR concentrations were probably driven by various potential emissions or abrasion sources from household products. Owing to possible abrasion of household products containing NBFRs that causes

precipitation to the floor by gravity, and the low vapor pressure that may facilitate partitioning to FD, EHTBB, BTBPE, and DBDPE were possibly more accumulated on the floor than on elevated surfaces of household products in indoor environment, but it was not wholly true. Multiple mechanisms might influence the fate of each NBFR compound. Moreover, Harrad et al. [57] found that concentrations of PBDEs were inversely correlated with surface loadings (ng dust per m<sup>2</sup> of surface area) in indoor environment. The negative correlation between levels of NBFRs and surface loadings may exist under similar circumstances. It was plausible that, relative higher dust loadings of EHTBB, BTBPE, and DBDPE in ESD resulted in lower concentrations. In terms of relative abundance, there was not statistically difference at a significance level of 5% for target NBFR compounds in two dust categories, except for DBDPE and BEHTEBP. DBDPE accounted for about 90.5% and 80.5% in FD and ESD, respectively, while larger percentage of BEHTEBP was observed in ESD, with a value of 11.1% (Additional file 1: Fig. S1). In summary, paired-samples *T* tests showed the difference of NBFR compounds between FD and ESD, possibly indicating different distributions of NBFRs in two dust categories. As young infants and toddlers contact more with FD, it will be more valid to estimate human exposure to NBFRs for them using FD. For older age groups (e.g., adults), ESD may be preferable for human exposure estimates.

Scatterplots and Spearman correlation coefficients examining relationships of seven target NBFRs between two dust categories were selected in this study. As shown in Fig. 3, log-transformed concentrations of some target compounds in FD, including PBT, HBB, EHTBB, BEHTEBP, and DBDPE, were significantly correlated with those in ESD ( $P < 0.05$ ). Particularly, strongly

significant correlations ( $P < 0.01$ ) of HBB, BEHTEBP, and DBDPE were found between FD and ESD, with respective correlation coefficient values of 0.69, 0.78, and 0.61. The results indicated that these compounds might be from similar emission sources such as polymeric materials, which was consistent with the report by Covaci et al. [8]. Similar results were also reported in house dust from Iraq [58]. However, no significant correlations of PBEB or BTBPE were obtained between FD and ESD, though PBEB and BTBPE were both used in thermoset resins [8]. The number and type of household products containing PBEB or BTBPE at different sampling heights may be potential factors that influence the environmental fate of PBEB and BTBPE.

The correlations among NBFRs in FD or ESD were also investigated. In terms of FD, the results in Table 3 pointed to a positive statistically significant correlation between concentrations of PBT, PBEB, and HBB, which may give grounds for assuming similar sources of their emission and transport mechanism to house dust. Interestingly, a weak positive statistically significant correlation was also found between the levels of HBB and DBDPE in FD, which may be associated with the photodegradation of DBDPE partially into HBB [55]. No significant correlation was examined between EHTBB and BEHTEBP, possibly indicating various emission sources of EHTBB and BEHTEBP from commercial mixtures (e.g., firemaster 550, BZ-54, DP-45) or different behavior of the two. Table 4 shows the tests of relationship among target NBFRs in ESD. Similar to FD, a stronger positive statistically significant association was found between PBT and PBEB in ESD ( $r = 0.879, P < 0.01$ ). Furthermore, a significant correlation ( $P < 0.05$ ) was also found between EHTBB and other four compounds PBT, PBEB, HBB, and DBDPE with respective correlation coefficient values



of 0.635, 0.573, 0.676, and 0.469, possibly indicating the similar source in indoor environment.

**Temporal variations of NBFrs in FD**

The geomean concentrations of  $\sum_7$ NBFrs were ranked as August (327.0, ng/g) > December (320.7) > October (258.1) > February (251.5) > June (216.9) > April (162.3) (Additional file 1: Fig. S2). The concentrations of

$\sum_7$ NBFrs in August were higher than those in the other months by testing with one-way ANOVA ( $P < 0.05$ ). April, (June, August), and October (December, February) represent the seasons of spring, summer, autumn, and winter, respectively. Geomean concentrations of  $\sum_7$ NBFrs in FD decreased in the order: summer > winter > autumn > spring (Fig. 4). Higher room temperature, which led to more volatile emissions of NBFrs

**Table 3 Spearman’s rank correlation coefficient (r) between concentrations of NBFrs in FD**

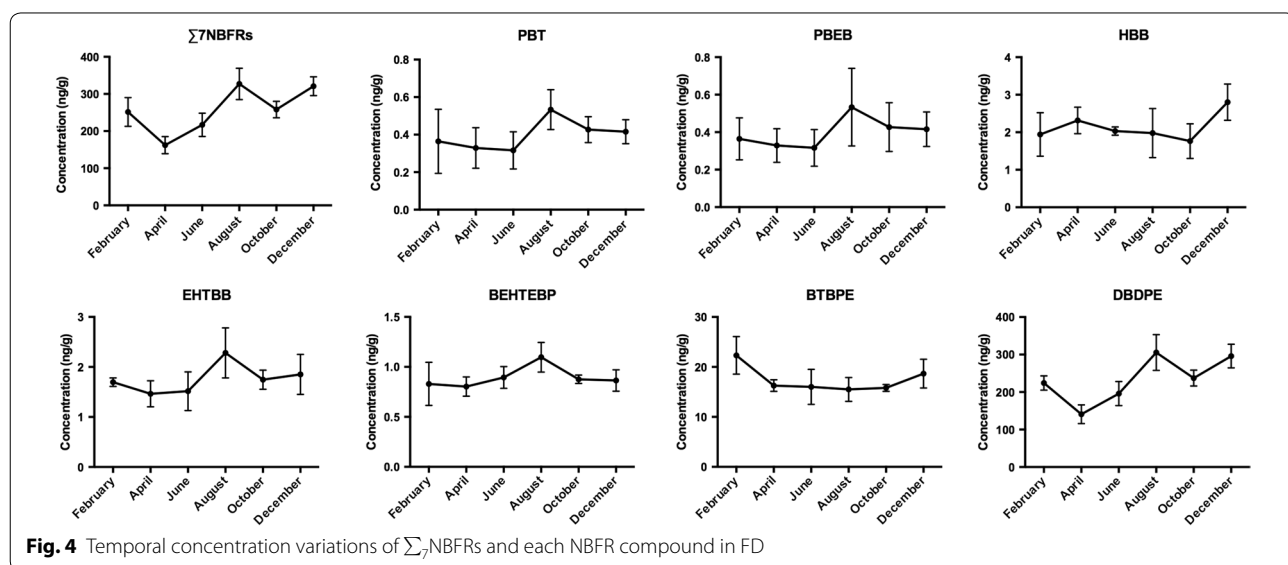
	PBT	PBEB	HBB	EHTBB	BTBPE	BEHTEBP	DBDPE
PBT		0.571**	0.031	-0.055	0.095	0.012	0.031
PBEB	0.571**		0.203*	-0.061	0.085	-0.022	0.109
HBB	0.031	0.203*		-0.100	0.089	-0.020	0.186*
EHTBB	-0.055	-0.061	-0.100		-0.066	0.023	-0.114
BTBPE	0.095	0.085	0.089	-0.066		-0.008	0.005
BEHTEBP	0.012	-0.022	-0.020	0.023	-0.008		-0.016
DBDPE	0.031	0.109	0.186*	-0.114	0.005	-0.016	

\*  $P < 0.05$ ; \*\*  $P < 0.01$

**Table 4 Spearman’s rank correlation coefficient (r) between concentrations of NBFrs in ESD**

	PBT	PBEB	HBB	EHTBB	BTBPE	BEHTEBP	DBDPE
PBT		0.879**	0.445	0.635**	0.086	-0.083	0.556*
PBEB	0.879**		0.249	0.573*	0.205	-0.090	0.564*
HBB	0.445	0.249		0.676**	0.345	-0.116	0.161
EHTBB	0.635**	0.573*	0.676**		0.154	0.014	0.469*
BTBPE	0.086	0.205	0.345	0.154		-0.091	0.014
BEHTEBP	-0.083	-0.090	-0.116	0.014	-0.091		-0.270
DBDPE	0.556*	0.564*	0.161	0.469*	0.014	-0.270	

\*  $P < 0.05$ ; \*\*  $P < 0.01$



**Fig. 4** Temporal concentration variations of  $\sum_7$ NBFrs and each NBFr compound in FD



from sources (e.g., electrical appliances and furnishings), may be one possible explanation for higher concentrations of NBFRs in summer, but this is not wholly true. Interestingly, FD in winter showed the second highest concentration of  $\sum_7$ NBFRs. Household heating system indoors may counterbalance the effect of lower temperature in winter. Moreover, ventilation in winter is poorer, which may contribute to the residence of NBFRs indoors. Similar conclusions were reported in former studies [41, 59].

It was not surprising that the temporal trend of DBDPE was very analogous to that of  $\sum_7$ NBFRs (Fig. 4). The temporal variation of DBDPE might determine the temporal trend of  $\sum_7$ NBFRs, due to the domination of DBDPE in indoor dust. FD in summer showed the highest concentration of DBDPE (305.4 ng/g), while FD in spring the lowest (140.9 ng/g) (Additional file 1: Fig. S2). Over twofold difference of DBDPE between the maximum (winter) and minimum (spring) indicated that the sampling period is a vital influencing factor for further investigations of human exposure risk assessment. FD in summer also showed the highest levels of PBT, PBEB, EHTBB, and BEHTEBP except for HBB and BTBPE (Fig. 4). Higher levels of BTBPE appeared in colder months (February and December), indicating different fate of BTBPE in indoor environment over the monitoring period. Other influencing factors like the introduction or remove of FR-containing goods, floor materials, and humidity of rooms might also affect the levels of NBFRs indoors. To sum up, PBT, PBEB, EHTBB, BEHTEBP, and DBDPE in FD exhibited higher concentrations in warm summer. HBB and BTBPE, which were probably more favored at lower temperature, showed elevated concentrations in colder seasons.

### Estimation of human exposure to NBFRs

Human exposure to NBFRs via oral ingestion and dermal contact of house dust was estimated through application of the exposure factors recommended by the U.S. Environmental Protection Agency (EPA) and Ministry of Ecology and Environment of the People’s Republic of China (MEEPRC) [60, 61]. 5th percentile, mean/geomean, and 95th percentile concentrations were used to calculate various plausible low-end, ‘typical’, and high-end exposure scenarios. Five age groups were taken into consideration: infants (<1 year), toddlers (1–5 years), children (6–11 years), teenagers (12–19 years), and adults ( $\geq 20$  years). The recommended daily exposure values are listed in Table 5.

Calculated daily exposure doses (DEDs) of NBFRs with different exposure scenarios are presented in Table 6. The contribution of dust ingestion to NBFR exposure is nearly one order of magnitude higher than the exposure from dermal absorption in FD and ESD. Dust ingestion was the more important pathway of human exposure to NBFRs. On the basis of mean concentration, the DED of NBFRs via dust ingestion (DED<sub>i</sub>) in both FD and ESD was decreased in the order: infants > toddlers > children > teenagers > adults. The highest DED was in the group of infants via dust ingestion at 95th percentile in FD, 9.10 ng/kg bw/day, which was about 1, 2, 4, and 8 times higher than that for toddlers, children, teenagers, and adults, respectively. The DED via dermal absorption (DED<sub>da</sub>) of NBFRs present in house dust was estimated based on the assumption that only 3% of the dose is absorbed through the skin [42, 64, 65]. All five age groups had lower DED<sub>da</sub> than DED<sub>i</sub>, and the highest DED<sub>da</sub> was 0.49 ng/kg bw/day for infants (FD). Total DEDs of NBFRs for five age groups were also calculated through summation of the DED via these two pathways (Additional

**Table 5 The recommended daily exposure values of five age groups for estimating daily exposure dose via dust ingestion and dermal absorption**

Dust exposure pathway	Factors	Daily exposure value				
		Infants	Toddlers	Children	Teenagers	Adults
Dust ingestion	Weight (kg) <sup>a</sup>	9.4	16.3	28.8	54.8	63.5
	Daily ingestion rate (g/day) <sup>a</sup>	0.06	0.10	0.103	0.086	0.05
	Indoor exposure fraction <sup>b</sup>	0.875	0.792	0.792	0.875	0.875
Dermal absorption	Weight (kg) <sup>a</sup>	9.4	16.3	28.8	54.8	63.5
	Total skin area (cm <sup>2</sup> ) <sup>a</sup>	4475	6940	11,500	16,100	17,000
	Body surface area (cm <sup>2</sup> /day) <sup>b</sup>	1119	1735	2875	4025	4250
	Dust adhered to skin (mg/cm <sup>2</sup> ) <sup>b</sup>	0.096	0.096	0.096	0.096	0.096
	NBFR fraction absorbed through the skin <sup>a</sup>	0.03	0.03	0.03	0.03	0.03
	Indoor exposure fraction <sup>b</sup>	0.875	0.792	0.792	0.875	0.875

<sup>a</sup> Data from MEEPRC [62]

<sup>b</sup> Data from USEPA [63]

**Table 6** Estimated daily exposure dose (DED) of NBFRs in house dust for five age groups in Shanghai, ng/kg bw/day

Dust type	DED <sub>i</sub>					DED <sub>da</sub>				
	Infants	Toddlers	Children	Teenagers	Adults	Infants	Toddlers	Children	Teenagers	Adults
FD										
Mean	2.75	2.39	1.39	0.68	0.34	0.15	0.12	0.11	0.09	0.08
Geomean	1.65	1.43	0.84	0.41	0.20	0.09	0.07	0.07	0.05	0.05
5th	0.24	0.21	0.12	0.06	0.03	0.01	0.01	<0.01	<0.01	<0.01
95th	9.10	7.92	4.62	2.24	1.12	0.49	0.40	0.37	0.30	0.27
ESD										
Mean	0.76	0.67	0.39	0.19	0.09	0.04	0.03	0.03	0.03	0.02
Geomean	0.66	0.57	0.33	0.16	0.08	0.04	0.03	0.03	0.02	0.02
5th	0.21	0.18	0.11	0.05	0.03	0.01	<0.01	<0.01	<0.01	<0.01
95th	2.26	1.97	1.15	0.56	0.28	0.12	0.10	0.09	0.07	0.07

file 1: Table S3). It is not surprising that the DED of NBFRs through dust ingestion made greater contribution to the cumulative DEDs, accounting for, on average, 90.4% of the total NBFR exposure. With age increasing, the proportion of DED<sub>i</sub> was becoming smaller, and dermal absorption for adults made greater contribution to total DEDs than that for younger groups. Adults showed the smallest percentage of DED<sub>i</sub> (80.9%), while the highest percentage was for the groups of infants and toddlers, both 95%. Younger infants and toddlers may be exposed more susceptibly to the risk of NBFRs from dust ingestion.

The DEDs of four main NBFRs (EHTBB, BEHTEBP, BTBPE, and DBDPE) via dust ingestion were also estimated for exposure risk from single NBFR compound (Additional file 1: Table S4). According to Hardy et al. [66], the oral reference dose (RfD) values for EHTBB, BEHTEBP, BTBPE, and DBDPE were 20,000, 20,000, 243,000, and 333,333 ng/kg bw/day. Compared with oral RfDs, the estimated DEDs of EHTBB, BEHTEBP, BTBPE, and DBDPE were at least  $2 \times 10^6$ ,  $10^6$ ,  $3 \times 10^6$ ,  $3.3 \times 10^6$  times lower for five age groups in this study, probably revealing that residents faced low exposure risks from EHTBB, BEHTEBP, BTBPE, and DBDPE in indoor house dust. We assumed 100% absorption efficiency of intake in accordance with the other studies [37, 67]. However, this assumption may result in the overestimation of human exposure to NBFRs. People may face lower risks if bioaccessibility of NBFRs is taken into account in future studies.

## Conclusions

This study investigated the levels, temporal variation, and human exposure of seven target NBFRs (PBT, PBEB, HBB, EHTBB, BEHTEBP, BTBPE, and DBDPE) in indoor house dust from Shanghai. DBDPE was the predominant

compound in both FD and ESD, probably indicating the extensive used of DBDPE in indoor environment. Compared with the other studies, the concentrations of DBDPE in this study were generally moderate. DBDPE, BTBPE, and EHTBB in FD exceeded significantly those in ESD, revealing that different influencing factors (e.g., physicochemical property, emission or abrasion source, and particle size) may affect the distribution of these compounds in two dust categories. Significant correlations of PBT, HBB, EHTBB, BEHTEBP, and DBDPE were found between FD and ESD, which may be influenced by similar emission sources such as polymeric materials. The concentrations of  $\sum_7$ NBFRs in FD decreased in the order: summer > winter > autumn > spring, possibly due to more volatile emissions caused by higher room temperature. The estimated DEDs of  $\sum_7$ NBFRs for young infants and toddlers via dust ingestion and dermal absorption were higher than other age groups. The DEDs of EHTBB, BEHTEBP, BTBPE, and DBDPE were several orders of magnitude lower than their oral RfDs, showing people faced low risks of these four emerging compounds through dust ingestion. In the future, bioaccessibility of NBFRs should be fully considered for human exposure estimates.

## Additional file

[Additional file 1.](#) Additional tables and figures.

## Abbreviations

ANOVA: analysis of variance; BEHTEBP: bis(2-ethylhexyl) tetrabromophthalate; BFRs: brominated flame retardants; DEDs: daily exposure doses; DBDPE: decabromodiphenylethane; DEHP: di(2-ethylhexyl) phthalate; ESD: elevated surface dust; EPA: Environmental Protection Agency; FRs: flame retardants; FD: floor dust; GC/MS: gas chromatography/mass spectrometry; HBB: hexabromobenzene; HBCDs: hexabromocyclododecanes; LOD: limit of detection; LOQ: limit of quantification; NCI: negative chemical ionization; NBFRs: novel brominated

flame retardants; PBEB: pentabromoethylbenzene; PBT: pentabromotoluene; POPs: persistent organic pollutants; PBDEs: polybrominated diphenyl ethers; PTV: programmable temperature vaporizer; RfD: reference dose; SIM: selected ion monitoring; S/N: signal-to-noise ratio; SRM: standard reference material; BTBPE: 1,2-bis(2,4,6-tribromophenoxy) ethane; <sup>13</sup>C-BDE-209: <sup>13</sup>C-decabromodiphenylether; EHTBB: 2-ethylhexyl 2,3,4,5-tetrabromobenzoate; BDE-128: 2,2',3,3',4,4'-hexabromodiphenyl ether; BDE-138: 2,2',3,4,4',5-hexabromodiphenyl ether; BDE-77: 3,3',4,4'-tetrabromodiphenyl ether.

#### Authors' contributions

DN performed chemical and data analysis, and wrote the manuscript. YQ made the study design and is the PI of the NSFC project. LL made contributions to sampling design and helped with preparing the manuscript. XD and YZ performed data analysis and the revision of the manuscript. DY, ZL, LC, ZZ, JZ, and ÅB gave substantial suggestions on experimental design and data interpretation. All authors read and approved the final manuscript.

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#### Competing interests

The authors declare that they have no competing interests.

#### Availability of data and materials

Not applicable.

#### Consent for publication

Not applicable.

#### Ethics approval and consent to participate

Not applicable.

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#### References

- Ali N, Dirtu AC et al (2012) Occurrence of alternative flame retardants in indoor dust from New Zealand: indoor sources and human exposure assessment. *Chemosphere* 88(11):1276–1282
- Huang YM, Chen LG et al (2010) PBDEs in indoor dust in South-Central China: characteristics and implications. *Chemosphere* 78(2):169–174
- Bjorklund JA, Sellstrom U et al (2012) Comparisons of polybrominated diphenyl ether and hexabromocyclododecane concentrations in dust collected with two sampling methods and matched breast milk samples. *Indoor Air* 22(4):279–288
- Tay JH, Sellstrom U et al (2018) Assessment of dermal exposure to halogenated flame retardants: comparison using direct measurements from hand wipes with an indirect estimation from settled dust concentrations. *Environ Int* 115:285–294
- Stapleton HM, Eagle S et al (2011) Associations between Polybrominated Diphenyl Ether (PBDE) flame retardants, phenolic metabolites, and thyroid hormones during pregnancy. *Environ Health Persp* 119(10):1454–1459
- Holma-Suutari A, Ruokojarvi P et al (2016) Biomonitoring of selected persistent organic pollutants (PCDD/Fs, PCBs and PBDEs) in Finnish and Russian terrestrial and aquatic animal species. *Environ Sci Eur* 28:5
- UNEP (2017) The 16 new POPs under the Stockholm Convention. <http://chm.pops.int/TheConvention/ThePOPs/TheNewPOPs/tabid/2511/Default.aspx>. Accessed 15 Jan 2019
- Covaci A, Harrad S et al (2011) Novel brominated flame retardants: a review of their analysis, environmental fate and behaviour. *Environ Int* 37(2):532–556
- WHO (1997) Flame retardants: a general introduction. Environmental health criteria. <http://www.inchem.org/documents/ehc/ehc/ehc192.htm>. Accessed 15 Jan 2019
- Davis EF, Stapleton HM (2009) Photodegradation pathways of non-brominated diphenyl ethers, 2-ethylhexyltetrabromobenzoate and di(2-ethylhexyl)tetrabromophthalate: identifying potential markers of photodegradation. *Environ Sci Technol* 43(15):5739–5746
- Stapleton HM, Allen JG et al (2008) Alternate and new brominated flame retardants detected in U.S. house dust. *Environ Sci Technol* 42(18):6910–6916
- OSPAR (2013) OSPAR list of chemicals for priority action. <https://www.ospar.org/work-areas/hasec/chemicals/priority-action>. Accessed 15 Jan 2019
- McGrath TJ, Morrison PD et al (2018) Concentrations of legacy and novel brominated flame retardants in indoor dust in Melbourne, Australia: an assessment of human exposure. *Environ Int* 113:191–201
- Brits M, de Vos J et al (2016) Critical review of the analysis of brominated flame retardants and their environmental levels in Africa. *Chemosphere* 164:174–189
- Tomy GT, Palace VP et al (2007) Dietary exposure of juvenile rainbow trout (*Oncorhynchus mykiss*) to 1,2-bis(2,4,6-tribromophenoxy)ethane: bioaccumulation parameters, biochemical effects, and metabolism. *Environ Sci Technol* 41(14):4913–4918
- He MJ, Luo XJ et al (2012) Bioaccumulation of polybrominated diphenyl ethers and decabromodiphenyl ether in fish from a river system in a highly industrialized area, South China. *Sci Total Environ* 419:109–115
- Tao F, Abdallah MAE, Harrad S (2016) Emerging and legacy flame retardants in UK indoor air and dust: evidence for replacement of PBDEs by emerging flame retardants? *Environ Sci Technol* 50(23):13052–13061
- Noyes PD, Haggard DE et al (2015) Advanced morphological-behavioral test platform reveals neurodevelopmental defects in embryonic zebrafish exposed to comprehensive suite of halogenated and organophosphate flame retardants. *Toxicol Sci* 145(1):177–195
- Mankidy R, Ranjan B et al (2014) Effects of novel brominated flame retardants on steroidogenesis in primary porcine testicular cells. *Toxicol Lett* 224(1):141–146
- Saunders DMV, Higley EB et al (2013) In vitro endocrine disruption and TCDD-like effects of three novel brominated flame retardants: TBPH, TBB, & TBCO. *Toxicol Lett* 223(2):252–259
- Johnson PI, Stapleton HM et al (2013) Associations between brominated flame retardants in house dust and hormone levels in men. *Sci Total Environ* 445:177–184
- Egloff C, Crump D et al (2011) In vitro and in ovo effects of four brominated flame retardants on toxicity and hepatic mRNA expression in chicken embryos. *Toxicol Lett* 207(1):25–33
- Wang FX, Wang J et al (2010) Comparative tissue distribution, biotransformation and associated biological effects by decabromodiphenyl ether and decabrominated diphenyl ether in male rats after a 90-day oral exposure study. *Environ Sci Technol* 44(14):5655–5660
- Nakari T, Huhtala S (2010) In vivo and in vitro toxicity of decabromodiphenyl ether, a flame retardant. *Environ Toxicol* 25(4):333–338

25. Brown FR, Whitehead TP et al (2014) Levels of non-polybrominated diphenyl ether brominated flame retardants in residential house dust samples and fire station dust samples in California. *Environ Res* 135:9–14
26. Dodson RE, Perovich LJ et al (2012) After the PBDE phase-out: a broad suite of flame retardants in repeat house dust samples from California. *Environ Sci Technol* 46(24):13056–13066
27. Fan XH, Kubwabo C et al (2016) Non-PBDE halogenated flame retardants in Canadian indoor house dust: sampling, analysis, and occurrence. *Environ Sci Pollut R* 23(8):7998–8007
28. Al-Omran LS, Harrad S (2018) Within-room and within-home spatial and temporal variability in concentrations of legacy and “novel” brominated flame retardants in indoor dust. *Chemosphere* 193:1105–1112
29. Al-Omran LS, Harrad S (2017) Influence of sampling approach on concentrations of legacy and “novel” brominated flame retardants in indoor dust. *Chemosphere* 178:51–58
30. Ali N, Harrad S et al (2011) “Novel” brominated flame retardants in Belgian and UK indoor dust: implications for human exposure. *Chemosphere* 83(10):1360–1365
31. Tay JH, Sellstrom U et al (2017) Human exposure to legacy and emerging halogenated flame retardants via inhalation and dust ingestion in a Norwegian Cohort. *Environ Sci Technol* 51(14):8176–8184
32. Cequier E, Jonas AC et al (2014) Occurrence of a broad range of legacy and emerging flame retardants in indoor environments in Norway. *Environ Sci Technol* 48(12):6827–6835
33. Cristale J, Hurtado A et al (2016) Occurrence and sources of brominated and organophosphorus flame retardants in dust from different indoor environments in Barcelona, Spain. *Environ Res* 149:66–76
34. Sahlstrom LMO, Sellstrom U et al (2015) Estimated intakes of brominated flame retardants via diet and dust compared to internal concentrations in a Swedish mother-toddler cohort. *Int J Hyg Envir Heal* 218(4):422–432
35. Newton S, Sellstrom U, de Wit CA (2015) Emerging flame retardants, PBDEs, and HBCDDs in indoor and outdoor media in Stockholm, Sweden. *Environ Sci Technol* 49(5):2912–2920
36. Peng CF, Tan HL et al (2017) Emerging and legacy flame retardants in indoor dust from East China. *Chemosphere* 186:635–643
37. Zheng XB, Xu FC et al (2015) Flame retardants and organochlorines in indoor dust from several e-waste recycling sites in South China: composition variations and implications for human exposure. *Environ Int* 78:1–7
38. Cao ZG, Xu FC et al (2014) Distribution patterns of brominated, chlorinated, and phosphorus flame retardants with particle size in indoor and outdoor dust and implications for human exposure. *Environ Sci Technol* 48(15):8839–8846
39. Ali N, Ali L et al (2013) Levels and profiles of organochlorines and flame retardants in car and house dust from Kuwait and Pakistan: implication for human exposure via dust ingestion. *Environ Int* 55:62–70
40. Nkabinde SN, Okonkwo JO et al (2018) Determination of legacy and novel brominated flame retardants in dust from end of life office equipment and furniture from Pretoria, South Africa. *Sci Total Environ* 622:275–281
41. Cao ZG, Xu FC et al (2014) Differences in the seasonal variation of brominated and phosphorus flame retardants in office dust. *Environ Int* 65:100–106
42. Johnson-Restrepo B, Kannan K (2009) An assessment of sources and pathways of human exposure to polybrominated diphenyl ethers in the United States. *Chemosphere* 76(4):542–548
43. Hoffman K, Webster TF et al (2017) Toddler’s behavior and its impacts on exposure to polybrominated diphenyl ethers. *J Expo Sci Env Epid* 27(2):193–197
44. Du XY, Yuan B et al (2018) Short-, medium-, and long-chain chlorinated paraffins in wildlife from paddy fields in the Yangtze River Delta. *Environ Sci Technol* 52(3):1072–1080
45. Ali N, Harrad S et al (2011) Analytical characteristics and determination of major novel brominated flame retardants (NBFRs) in indoor dust. *Anal Bioanal Chem* 400(9):3073–3083
46. Van den Eede N, Dirtu AC et al (2012) Multi-residue method for the determination of brominated and organophosphate flame retardants in indoor dust. *Talanta* 89:292–300
47. Sahlstrom L, Sellstrom U, de Wit CA (2012) Clean-up method for determination of established and emerging brominated flame retardants in dust. *Anal Bioanal Chem* 404(2):459–466
48. Cristale J, Lacorte S (2013) Development and validation of a multiresidue method for the analysis of polybrominated diphenyl ethers, new brominated and organophosphorus flame retardants in sediment, sludge and dust. *J Chromatogr A* 1305:267–275
49. Sun JQ, Xu Y et al (2018) Levels, occurrence and human exposure to novel brominated flame retardants (NBFRs) and Dechlorane Plus (DP) in dust from different indoor environments in Hangzhou, China. *Sci Total Environ* 631–632:1212–1220
50. Qi H, Li WL et al (2014) Levels, distribution and human exposure of new non-BDE brominated flame retardants in the indoor dust of China. *Environ Pollut* 195:1–8
51. Niu D, Qiu Y et al (2018) Occurrence of polybrominated diphenyl ethers in floor and elevated surface house dust from Shanghai, China. *Environ Sci Pollut Res Int*
52. NFPA (2018) List of NFPA Codes & Standards. <https://www.nfpa.org/Codes-and-Standards/All-Codes-and-Standards/List-of-Codes-and-Standards>. Accessed 15 Jan 2019
53. Parliament, UK (1988) The Furniture and Furnishings (Fire) (Safety) Regulations. <http://www.legislation.gov.uk/ukSI/1988/1324/resources>. Accessed 15 Jan 2019
54. Ma YN, Venier M, Hites RA (2012) 2-Ethylhexyl tetrabromobenzoate and Bis(2-ethylhexyl) tetrabromophthalate flame retardants in the Great Lakes atmosphere. *Environ Sci Technol* 46(1):204–208
55. Gouteux B, Alaei M et al (2008) Polymeric brominated flame retardants: are they a relevant source of emerging brominated aromatic compounds in the environment? *Environ Sci Technol* 42(24):9039–9044
56. Kuang JM, Ma YN, Harrad S (2016) Concentrations of “legacy” and novel brominated flame retardants in matched samples of UK kitchen and living room/bedroom dust. *Chemosphere* 149:224–230
57. Harrad S, Ibarra C et al (2008) Concentrations of brominated flame retardants in dust from United Kingdom cars, homes, and offices: causes of variability and implications for human exposure. *Environ Int* 34(8):1170–1175
58. Al-Omran LS, Harrad S (2016) Polybrominated diphenyl ethers and “novel” brominated flame retardants in floor and elevated surface house dust from Iraq: implications for human exposure assessment. *Emerging Contaminants* 2:7–13
59. Yu YX, Pang YP et al (2012) Concentrations and seasonal variations of polybrominated diphenyl ethers (PBDEs) in in- and out-house dust and human daily intake via dust ingestion corrected with bioaccessibility of PBDEs. *Environ Int* 42:124–131
60. USEPA (2008) Integrated Risk Information System (IRIS). National Center for Environmental Assessment, Washington, DC. <http://www.epa.gov/iris>. Accessed 15 Jan 2019
61. MEEPRC (2016) Exposure Factors Handbook of Chinese Population. China Environmental Science Press: Chapter 10 (Chinese Edition)
62. MEEPRC (2016) Exposure Factors Handbook of Chinese Population (Children 0–5 years). China Environmental Science Press: Chapter 5/11 (Chinese Edition)
63. USEPA (2011) Exposure Factors Handbook: 2011. Edition, National Centre for Environmental Assessment, Washington, DC. EPA/600/R-09/052F
64. Stapleton HM, Dodder NG (2008) Photodegradation of decabromodiphenyl ether in house dust by natural sunlight. *Environ Toxicol Chem* 27(2):306–312
65. Lorber M (2008) Exposure of Americans to polybrominated diphenyl ethers. *J Expo Sci Env Epid* 18(1):2–19
66. Hardy M, Biesemeier J et al (2008) Comment on “Alternate and new brominated flame retardants detected in U.S. house dust”. *Environ Sci Technol* 42(24):9453–9454 (**author reply 9455–9456**)
67. Ali N, Van den Eede N et al (2012) Assessment of human exposure to indoor organic contaminants via dust ingestion in Pakistan. *Indoor Air* 22(3):200–211