Brominated and phosphate flame retardants (FRs) in indoor dust from different microenvironments: Implications for human exposure via dust ingestion and dermal contact

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HIGHLIGHTS

- FRs were measured in different types of indoor dust in bedrooms and offices.
- AC filter dust and bed dust were main reservoirs of FRs in bedrooms.
- Dust from printer table surface had higher FR levels than other dust in offices.
- Dermal exposure to FRs via contact with beddings should be further investigated.

ABSTRACT

Indoor dust has been widely used to monitor flame retardants (FRs) in indoor environment, but most studies only focused on floor dust. In the present study, FRs were examined in indoor dust from different locations. Dust from air conditioner (AC) filters, beddings, floor, and windows in bedrooms, and dust from AC filters, printer table surface, computer table surface, floor, and windows in offices were collected, respectively. Polybrominated diphenyl ether congener 209 (BDE 209) and decabromodiphenyl ethane (DBDPE) were the most abundant brominated flame retardants (BFRs), and tris(chloroisopropyl) phosphate (TCIPP), tris(1,3-dichloroisopropyl) phosphate (TDCIPP), and triphenyl phosphate (TPHP) were the most abundant phosphate flame retardants (PFRs). In bedrooms, the AC filter dust had the highest median levels of BDE 209 (536 ng/g) and DBDPE (2720 ng/g), while bed dust had the highest median levels of ∑PFRs (2750 ng/g) among dust samples. In offices, printer table dust had higher median levels of BDE 209 (1330 ng/g), DBDPE (8470 ng/g), and ∑PFRs (11,000 ng/g) than those in other dust samples. The high dust ingestion values of BDE 209, DBDPE, and individual PFR were 0.28, 1.20, and <0.01–0.32 ng/kg bw/day and 7.37, 31.2, and <0.01–4.54 ng/kg bw/day for BDE 209, DBDPE, and individual PFR for adults and toddlers, respectively. The high dermal exposure values of individual PFR during sleeping were <0.01–0.23 and <0.01–0.36 ng/kg bw/day for adults and toddlers, respectively. More human exposure pathways other than dust ingestion should be considered, such as the dermal contact with beddings and furniture.

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1. Introduction

Flame retardants (FRs) are additives widely applied in textiles, foams, plastics, and also electronic products. Many flame retardants were found to be environmental pollutants in the past few decades...
(de Wit, 2002; Covaci et al., 2011; van der Veen and de Boer, 2012). The occurrence of these chemicals in indoor environment has become an important issue as people spend most of their daily time indoors (Abdallah et al., 2008; Ali et al., 2013; Stapleton et al., 2009). Polybrominated diphenyl ethers (PBDEs) used to be among the most commonly used FRs (de Wit, 2002). The Penta-BDE and Octa-BDE mixtures were banned because of their persistence, bioaccumulation, and toxicity in the environment and biota (UNEP, 2009). Consequently, several emerging FRs, such as deca-bromodiphenyl ethane (DBDPE) and phosphate flame retardants (PFRs) have replaced PBDEs (Covaci et al., 2011; van der Veen and de Boer, 2012). These emerging FRs are expected to be increasingly found in indoor environment. Some researchers have observed comparable or even higher concentrations of DBDPE and PFRs than PBDEs (Cao et al., 2014; Zheng et al., 2015). However, these emerging FRs could exhibit potential toxicity in organisms, which raised concern about potential adverse effects on wildlife and humans (Meeker et al., 2013; WHO, 1998).

Indoor dust could reflect the overall pollution status of pollutants in the indoor environment and has been frequently used in environmental monitoring studies (Cao et al., 2014; Dirtu et al., 2012; Harrad et al., 2008). In previous studies, floor dust was mostly swept or collected to explore the indoor contamination with FRs (Cao et al., 2014; Dirtu et al., 2012; Harrad et al., 2008). Some studies swept or vacuumed the surface of furniture and electronics, where FRs might be emitted from the inside of consumer products (Abbasi et al., 2016; Gallen et al., 2014; Liu et al., 2017). An old personal computer (PC) was found to influence the PBDE contamination in indoor air, as the PBDE concentrations decreased after the replacement of the old PC produced in 1998 with another PC produced in 2003 (Hazrati and Harrad, 2006). Aside from computers, other products, such as furniture, air conditioners (ACs) and printers can also serve as FR sources in indoor environment (Ali et al., 2016; He et al., 2016; Yu et al., 2013). He et al. (2016) detected FRs on AC filters, and found higher FR levels in AC filter dust than those in floor dust. The sources of FRs on AC filters have not been elucidated in previous studies (Ali et al., 2016; He et al., 2016; Yu et al., 2013). More studies on concentrations and composition patterns of FRs emitted from these commercial products would help to more accurately estimate the indoor FR contamination, and would also be useful in tracing the FR sources in indoor environment.

Dust ingestion has been recognized as an important pathway of human exposure to FRs (Ali et al., 2013; Dirtu et al., 2012; Zheng et al., 2015). Comparative levels of PFRs and PBDEs in indoor dust were reported (Zheng et al., 2015) and PFRs were proved to be less bioaccumulative than PBDEs in eggs (Zheng et al., 2016). Dust ingestion and food consumption are both important pathways of human exposure to FRs. In contrast, very few studies investigated the dermal exposure to FRs (Abdallah et al., 2015, 2016; Liu et al., 2017). Contact with indoor dust may lead to the adsorption of these chemicals to skin, which has been previously discussed in literature (Abdallah et al., 2015, 2016). Aside from the electronics and furniture, beddings including pillows and mattresses are also potential sources of FRs. TDCIPP was detected in polyurethane foam samples from mattress pad and pillow (Stapleton et al., 2009). We spend approximately 7–8 sleeping hours in beds every day, but the dermal absorption of FRs from beddings has been neglected so far.

In the present study, we measured FRs in indoor dust from different locations in bedrooms and offices. The aims of the present study were as follows: (i) to document the concentrations and profiles of FRs in different types of indoor dust, and to get more comprehensive knowledge of FR pollution in indoor environment; (ii) to investigate the difference in FR pollution and the potential sources of FRs in different indoor environments (bedrooms and offices); (iii) to estimate the human exposure of FRs via dust ingestion and dermal contact.

2. Materials and methods

2.1. Sample collection

Indoor dust samples from bedrooms and offices were collected in Guangzhou City located in South China in June 2015. Dust from AC filters (n = 8), beds (n = 9), floors (n = 9), and windows (n = 9) were collected in nine bedrooms, and dust from AC filters (n = 8), printer table surfaces (n = 8), PC table surfaces (n = 9), floors (n = 9), and windows (n = 9) were collected in nine offices, respectively (Supplementary Material). Most rooms were equipped with ACs as Guangzhou is located in the subtropical region. The sampling of dust followed the method in our previous studies (Wang et al., 2010; Zheng et al., 2015). The dust from AC filters, floors, and windows were collected using clean brushes, which were pre-cleaned with ethanol. Window dust samples were collected from the inner window sill. In offices, almost all computers were laptops, and printers were placed on another table in most cases. We moved the laptops and printers, and collected dust from the location of laptops and printers on table surface using pre-cleaned brushes. These dust samples were assigned as the PC table dust and printer table dust, respectively. Pillows, sheets, blankets, and quilts were vacuumed by a small cleaner, and the collected dust was named as bed dust. Individual dust sample was sieved through a stainless-steel sieve (2 mm), then stored under −20 °C until further analysis.

2.2. Sample preparation and analysis

The sample preparation was developed from our previous study with some modifications. After been spiked with internal standards (BDE 118, BDE 128, 13C-BDE 209, triphenyl phosphate-d15 (TPHP-d15), and tri-n-butyl phosphate-d27 (TnBP-d27)), samples were extracted by ultrasonication with 3 mL hexane/acetone (1/1, v/v). The extracts were fractioned on Supelclean™ ENV™-Florisil cartridges (500 mg, 3 mL, Supelco, Bellefonte, PA, USA); the first fraction was eluted with 8 mL hexane and the second fraction was eluted with 10 mL ethyl acetate. PBDEs, dechlorane plus (DPs), and DBDPE were present in the first fraction, while PFRs were present in the second fraction. After concentration under nitrogen, the first fraction was further cleaned by 2 mL concentrated sulfuric acid and the organic phase was transferred for further analysis. After evaporation to nearly dryness, both fractions were resolubilized in 100 μL of isooctane prior to GC-MS analysis.

PBDEs, DPs, and DBDPE were measured by a 7890 Agilent (Santa Clara, California, USA) gas chromatography (GC), coupled with a 5975 mass spectrometer (MS) operated in electron capture negative ionization and the selected ion monitoring mode. A DB-5HT capillary column (15 m × 250 μm i.d. × 0.10 μm film thickness; J&W Scientific, CA) was used to separate the PBDEs (BDEs 28, 47, 99, 100, 153, 184, and 209), DPs, and DBDPE. Details of the instrumental conditions were published elsewhere (Zheng et al., 2012). Ions m/z 79 and 81 were monitored for tri-to nona-BDEs and DBDPE. Ions m/z 486.7 and 488.7, 653.8 and 655.8, 494.7, and 496.7 were monitored for BDE 209, DPs, and 13C-BDE 209, respectively. Eleven PFRs (triethyl phosphate (TEP), tri-n-propyl phosphate (TTP), triisopropyl phosphate (TIPP), tri(2-chloroethyl) phosphate (TCEP), tris[chloroisopropyl] phosphate (TCIPP), tris(1,3-dichloroisopropyl) phosphate (TDICP), tri-n-butyl phosphate (TNBP), tris(2-butoxyethyl) phosphate (TBOEP), TPHP, 2-ethylhexyl diphenyl phosphate (EHDPP), and tri(2-ethylhexyl) phosphate (TEHP)) were quantified using a Shimadzu 2010 gas chromatograph.
coupled to a mass spectrometer (GC/MS) with an electron impact ion source. A DB-5MS capillary column (30 m × 0.25 mm × 0.25 μm, Agilent, USA) column was used, and the MS was operated in SIM mode with 2 characteristic ions acquired for each compound. Detailed information about analytical parameters was provided in our previous study (He et al., 2015).

2.3. Quality control

The method for quality control (QC) was performed by regular analysis of blank blanks (sodium sulfate prepared during sampling), procedural blanks (sodium sulfate prepared before sample extraction), and spiked blanks and floor dust in every batch. Spiked mixtures consisted of 20 ng of PBDEs (BDEs 28, 47, 99, 100, 153, 154, and 183), DBDPE, and DPs, 50 ng of BDE 209, and 100 ng of individual PFR. Procedure blanks contained traces of target chemicals, including BDE 153, BDE 209, TNBP, TCEP, TCIPP, and EHDPP with average values of 0.22, 0.13, 0.78, 1.61, 2.69, and 2.17 ng, respectively. The average values of FRs in blanks were subtracted from the values in dust samples. The recoveries in spiked blanks and dust samples were provided in our previous study (He et al., 2015). The concentrations of PBDEs in AC filter dust from South China (median: 477 ng/g) (Ni et al., 2011), Philippines (mean: 2172 ng/g) (Pulong and Espino, 2013), Greece (median: 1092 ng/g) (Besis et al., 2014), and Saudi Arabia (mean: 350 ng/g) (Ali et al., 2016) were in the same range with our results (536 and 812 ng/g in AC filter dust from bedrooms and offices, respectively). The median level of ΣPFRs (15,400 ng/g) in AC filters from Saudi Arabia (Ali et al., 2016) was higher than those (1440 and 4020 ng/g in AC filter dust from bedrooms and offices, respectively) in the present study. In the study of He et al. (2016), the ΣPFR concentrations in AC filter dust ranked in the order of dorms (300 ng/g) < houses (3120 ng/g) < office (5940 ng/g) < public environments (11,600 ng/g) (He et al., 2016). The lower ΣPFR concentrations in dorms were attributed to the less furnishing and electronics in dorms compared with offices and public environments (He et al., 2016). We also observed the elevated levels of most FRs in AC filter dust, floor dust, and window dust from offices than from those bedrooms.

2.4. Statistical analysis

Statistical calculations were performed using the SPSS 22 software for windows (SPSS). Statistical analysis was performed for FRs with detection frequencies higher than 50%. The concentrations of FRs were log-transformed to follow normal distribution. The values of undetected samples were replaced with LOQs/sqrt(2) before statistics. The correlations between chemical levels in different types of dust samples were determined by Spearman rank correlation analysis. The level of significance was set at p = 0.05 throughout the study. Principal component analysis (PCA) was conducted to compare the profiles of contaminants in different types of indoor dust from bedrooms and offices, respectively. The first three principal components (PCs) were considered to account for a significant contribution to the total variance according to the latent root criterion.

3. Results and discussion

3.1. Concentrations of FRs

Most of FRs, including PBDEs, DPs, DBDPE, and PFRs, were detected in more than 50% of dust samples from bedrooms (Tables 1 and 2). AC filter dust had the highest median concentrations of PBDEs (536 ng/g) and DBDPE (2720 ng/g), while bed dust had the highest median concentrations of ΣPFRs (sum of concentrations of eleven PFRs, 2750 ng/g) in all types of dust in bedrooms (Table 1). Among dust samples from offices, printer table dust had the highest median concentrations of PBDEs (1335 ng/g), DBDPE (8470 ng/g), and PFRs (11,000 ng/g) (Table 2). In most cases, the predominant FRs, including BDE 209, DBDPE, TCEP, TDCIPP, and TPHP, exhibited lower or similar concentrations in floor dust compared with those in dust from AC filters, printer tables, and PC tables, which were consistent with the results from He et al. (2016). Our results of PBDEs, DBDPE, and ΣPFRs in floor dust were similar with the data in literature (Ali et al., 2013; Dirtu et al., 2012; Van den Eede et al., 2011), and up to two orders of magnitudes lower than our previous measurements on FRs in indoor dust from electronic waste recycling sites (Zheng et al., 2015). The concentrations of PBDEs in AC filter dust from South China (median: 477 ng/g) (Ni et al., 2011), Philippines (mean: 2172 ng/g) (Pulong and Espino, 2013), Greece (median: 1092 ng/g) (Besis et al., 2014), and Saudi Arabia (mean: 350 ng/g) (Ali et al., 2016) were in the same range with our results (536 and 812 ng/g in AC filter dust from bedrooms and offices, respectively). The median level of ΣPFRs (15,400 ng/g) in AC filters from Saudi Arabia (Ali et al., 2016) was higher than those (1440 and 4020 ng/g in AC filter dust from bedrooms and offices, respectively) in the present study. In the study of He et al. (2016), the ΣPFR concentrations in AC filter dust ranked in the order of dorms (300 ng/g) < houses (3120 ng/g) < office (5940 ng/g) < public environments (11,600 ng/g) (He et al., 2016). The lower ΣPFR concentrations in dorms were attributed to the less furnishing and electronics in dorms compared with offices and public environments (He et al., 2016). We also observed the elevated levels of most FRs in AC filter dust, floor dust, and window dust from offices than from those bedrooms.

3.2. Composition profiles of FRs

Among the eight PBDE congeners, BDE 99 and BDE 209 were detected in more than 50% of dust samples. BDE 209 contributed more than 97% of PBDEs in all dust samples. Penta- and Octa-BDEs have been listed in the persistent organic pollutants (POPs) list since 2009 by Stockholm Convention (UNEP, 2009). Deca-BDE was proposed for listing in POPs list under Stockholm Convention, but has not been added yet (Stockholm Convention, 2013). The predominance of BDE 209 in PBDEs in indoor dust was also reported in numerous studies (Ali et al., 2013; Cao et al., 2014; Dirtu et al., 2012). TCIPP and TDCIPP were main PFRs in bedrooms (Fig. 1A). TEP, TPP, TNBP, TPP, and TBOEP were found in less than 50% of samples. In offices, PFRs in dust were dominated by TCIPP and TPHP (Fig. S1B, Supplementary Material). TCIPP is widely applied in polyurethane foam, and TPHP is used in hydraulic fluids, polyvinyl chloride, and electronic equipment (WHO, 1997; van der Veen and de Boer, 2012), which may lead to the relative high concentrations of TCIPP and TPHP in offices. The composition profiles of FRs varied in different types of indoor dust. In dust from bedrooms, DBDPE, TCIPP, and TDCIPP were the three main pollutants (Fig. 1A). Dust from AC filters (proportion: 50%) and windows (53%) had higher proportions of DBDPE than other chemicals, while bed dust had higher proportions of TCIPP (31%, 1005 ng/g) and TDCIPP (26%, 1050 ng/g) than other chemicals (Fig. 1A). DBDPE is widely applied in electronics as replacement of BDE 209 (Covaci et al., 2011). Bed dust mixed used to be widely used in textiles, and PFRs are widely used in textiles and foams as replacements for phased-out Penta-BDEs. TCEP and TDCIPP have been used in textiles. TCIPP, TDCIPP, TEP, and TEHP are applied in polyurethane foam, while TEP and TEHP are also used as plasticizers in polyvinyl chloride. (WHO, 1997; van der Veen and de Boer, 2012). Bed dust had lower fractions of PBDEs and DBDPE, and higher fractions of most PFRs compared with those in AC filter dust and floor dust, indicating beds as important reservoirs of PFRs in bedrooms. FRs in AC filter dust could derive from both the emission of FRs in AC components and the deposition of FRs on filters along with the small suspended particles or dust during ventilation (Ali et al., 2016; He et al., 2016; Yu et al., 2013). Thus, AC filter could be either source or reservoir of FRs. It is interesting to find higher proportion of TCEP (21%, 433 ng/g) in AC filter dust, while TCEP only
Besides, TCEP had higher vapor pressure (1.1 \text{mm Hg at 25 \degree C}) than TCIPP (1.9 \text{mm Hg at 25 \degree C}), TDCIPP (7.4 \text{mm Hg at 25 \degree C}), and TPHP (1.2 \text{mm Hg at 25 \degree C}) (van der Veen and de Boer, 2012). TCEP might be more volatile, and TCEP could be from the circuit board and resins in ACs (WHO, 1997). TCEP might be more volatile, and TCEP could be from the circuit board and resins in ACs (WHO, 1997).

Table 1

<table>
<thead>
<tr>
<th></th>
<th>AC dust (n = 8)</th>
<th>Bed dust (n = 9)</th>
<th>Floor dust (n = 9)</th>
<th>Window dust (n = 9)</th>
</tr>
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<tbody>
<tr>
<td>BDE 47</td>
<td>nd</td>
<td>nd</td>
<td>nd-1.25</td>
<td>nd</td>
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<tr>
<td>BDE 99</td>
<td>2.74 \text{(nd-8.35)}</td>
<td>2.67 \text{(0.76–7.48)}</td>
<td>3.63 \text{(0.73–27.5)}</td>
<td>2.00 \text{(nd-3.27)}</td>
</tr>
<tr>
<td>BDE 100</td>
<td>nd</td>
<td>nd-0.99</td>
<td>nd-4.23</td>
<td>nd-1.68</td>
</tr>
<tr>
<td>BDE 153</td>
<td>nd</td>
<td>nd</td>
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<td>nd</td>
</tr>
<tr>
<td>BDE 154</td>
<td>nd-12.65</td>
<td>nd</td>
<td>nd-0.04</td>
<td>nd</td>
</tr>
<tr>
<td>BDE 183</td>
<td>nd-5.39</td>
<td>nd</td>
<td>nd-38.8</td>
<td>nd</td>
</tr>
<tr>
<td>BDE 209</td>
<td>533 \text{(284–1230)}</td>
<td>138 \text{(35.3–364)}</td>
<td>351 \text{(87.8–1250)}</td>
<td>520 \text{(103–1140)}</td>
</tr>
<tr>
<td>PBDEs</td>
<td>536 \text{(286–1240)}</td>
<td>141 \text{(34.3–369)}</td>
<td>355 \text{(88.6–1250)}</td>
<td>522 \text{(103–1140)}</td>
</tr>
<tr>
<td>syn-TP</td>
<td>0.97 \text{(nd-3.18)}</td>
<td>0.08 \text{(nd-1.69)}</td>
<td>0.44 \text{(0.16–3.91)}</td>
<td>0.91 \text{(nd-4.21)}</td>
</tr>
<tr>
<td>anti-TP</td>
<td>3.70 \text{(nd-10.4)}</td>
<td>1.53 \text{(nd-3.91)}</td>
<td>2.35 \text{(1.29–14.1)}</td>
<td>3.66 \text{(nd-12.8)}</td>
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<tr>
<td>DPs</td>
<td>5.20 \text{(nd-13.5)}</td>
<td>2.15 \text{(nd-4.54)}</td>
<td>2.91 \text{(1.52–18.0)}</td>
<td>4.05 \text{(nd-17.0)}</td>
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<tr>
<td>(f_{\text{anti}})</td>
<td>0.83 \text{(0.63–0.90)}</td>
<td>0.93 \text{(0.47–0.98)}</td>
<td>0.85 \text{(0.73–0.91)}</td>
<td>0.80 \text{(0.74–0.94)}</td>
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<tr>
<td>DBDPE</td>
<td>2720 \text{(629–20,010)}</td>
<td>516 \text{(73.8–5630)}</td>
<td>1030 \text{(272–3720)}</td>
<td>1940 \text{(1140–3700)}</td>
</tr>
<tr>
<td>TEP</td>
<td>nd-28.9</td>
<td>nd-30.8</td>
<td>nd-6.36</td>
<td>nd-2.26</td>
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<td>TPP</td>
<td>nd</td>
<td>nd-57.5</td>
<td>nd-24.2</td>
<td>nd-103</td>
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<tr>
<td>TiPP</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd-188</td>
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<tr>
<td>TNBP</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
</tr>
<tr>
<td>TCEP</td>
<td>433 \text{(81.8–19200)}</td>
<td>65.6 \text{(46.7–261)}</td>
<td>106 \text{(46.5–690)}</td>
<td>167 \text{(58.9–717)}</td>
</tr>
<tr>
<td>TCIPP</td>
<td>272 \text{(87.5–4250)}</td>
<td>1005 \text{(538–5850)}</td>
<td>251 \text{(11.2–3420)}</td>
<td>339 \text{(73.4–2260)}</td>
</tr>
<tr>
<td>TDCIPP</td>
<td>217 \text{(nd-196–5580)}</td>
<td>1050 \text{(196–5580)}</td>
<td>327 \text{(77.7–9640)}</td>
<td>95.7 \text{(nd-158)}</td>
</tr>
<tr>
<td>TBOEP</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
<td>nd</td>
</tr>
<tr>
<td>TPHP</td>
<td>160 \text{(42.8–339)}</td>
<td>172 \text{(78.8–267)}</td>
<td>281 \text{(193–2550)}</td>
<td>199 \text{(44.7–397)}</td>
</tr>
<tr>
<td>EHDPP</td>
<td>210 \text{(nd-803)}</td>
<td>86.9 \text{(nd-295)}</td>
<td>180 \text{(nd-5220)}</td>
<td>140 \text{(nd-707)}</td>
</tr>
<tr>
<td>TEPH</td>
<td>143 \text{(nd-301)}</td>
<td>368 \text{(nd-862)}</td>
<td>194 \text{(nd-6570)}</td>
<td>nd-104</td>
</tr>
<tr>
<td>(\Sigma)PFRs</td>
<td>1440 \text{(887–19,400)}</td>
<td>2750 \text{(1560–12,600)}</td>
<td>1340 \text{(670–18,300)}</td>
<td>1480 \text{(177–3040)}</td>
</tr>
</tbody>
</table>

Printer table dust had higher proportion of TPHP (38\%, 5780 ng/g) than other chemicals, while AC filter dust, window dust, and floor dust had higher fractions of TCIPP (25–27\%, 954–1970 ng/g) than other chemicals. The three types of electric products, namely printers, PCs, and ACs, are all important reservoirs of DBDPE in offices. TCIPP and TPHP seem to be easily retained in dust from ACs and printers, respectively. Concentrations of FRs in floor dust were influenced by complex indoor sources, and may represent the integrated contamination of FRs in beds and offices.

3.3. PCA results of FRs in indoor dust

To further assess the pollution patterns and sources of FRs in...
indoor environment, PCA was performed using FR data from bedroom dust (Fig. 2A) and of floor dust (Fig. 2B), separately (Supplementary Material, Table S1). The first three factors included in the analysis accounted for 62% and 61% of the total variances for dust samples from bedrooms and of floors, respectively.

In bedrooms, PC1 was heavily weighted by BDE 209, anti-DP, syn-DP, and DBDPE, PC2 was mainly contributed by TCIPP and TDCIPP, while PC3 was mainly comprised of BDE 99 and TPHP (Fig. 2A). AC filter dust had the highest levels of BDE 209 and DBDPE among all types of samples, while TCIPP and TDCIPP were the most abundant chemicals in bed dust. In Fig. 2A, AC filter dust and window dust had higher scores in PC1 than other matrices, and bed dust had the highest scores in PC2. The PCA results indicate that ACs in bedrooms are likely to be the main source or reservoir of BDE 209, DBDPE and DP isomers. Beds are probably the main source of PFRs, especially TCIPP and TDCIPP. Floor dust had the highest score in PC3, which was mainly influenced by BDE 99 and TPHP. BDE 99 and TPHP might be from other indoor FR sources, such as furnishing and coatings in bedrooms. The concentrations of three main PFRs in bed dust, namely TCEP, TCIPP, TDCIPP, were significantly correlated with each other in bed dust (p < 0.01) (Supplementary Material, Table S2), but not in other types of dust (p > 0.05) (Supplementary Material, Tables S3–S5), which also supported that beds are the main source of PFRs in bedrooms.

In offices, PC1 mainly consisted of TCEP, TDCIPP, EHDPP, and TEHP (Fig. 2B). PC2 had higher loadings in BDE 209, TCIPP, and DP isomers, while PC3 was more contributed by BDE 209 and DBDPE. Printer table dust and AC filter dust had the highest scores in PC1 and 2, respectively. PC3 exhibited higher scores in both printer table dust and PC table dust. Printer table dust seems to be a source for a broad suite of FRs, including several PFRs, BDE 209, and DBDPE. PC table dust and AC filter dust were also important sources of BDE 209, DBDPE, and TCIPP. With the limitation in BDE 209 usage, DBDPE was probably the main substitute for BDE 209 in computers and ACs. Floor dust exhibited different score with other matrix, implying floor dust as the composite reservoir of FRs from various sources. Concentrations of TCIPP (p < 0.05) and EHDPP (p < 0.05) in AC filter dust were significantly correlated with those in floor dust (Supplementary Material, Tables S6–S11). Concentrations of TCIPP (p < 0.05), EHDPP (p < 0.05), and TEHP (p < 0.01) in PC table dust were significantly correlated with those in floor dust (Supplementary Material, Tables S6–S11). The migration of FRs from different sources to windows and floors still warrants more details.

### 3.4. Human exposure assessment

Human exposure to FRs via dust ingestion was estimated using the FR concentrations in floor dust from bedrooms and offices. Floor dust was most frequently used in estimation of human exposure to FRs via dust ingestion in literature. AC dust was also...
used to estimate human exposure in He et al. (2016). Therefore, we used the average values of FR concentrations in floor dust and AC dust to be comparable with previous studies. The daily intake of contaminants via dust ingestion was estimated as:

$$E - \text{ingestion} = \sum (C \times EF \times DI) / BM$$

where E-ingestion is the exposure value via dust ingestion (ng/kg bw/day), C is the average concentration of FR in floor dust and AC dust (ng/g), EF is the fraction of time spent in bedrooms or offices, DI is the dust ingestion rate (mg/day), and BM is the average body weight (kg). The EFs were 63.8% and 22.3% for home and of furniture or in offices. The average and high DIs were assumed as 20 and 50 mg/day for adults, and 50 and 200 mg/day for toddlers, respectively (Jones-Otazo et al., 2005). The average body weights were assumed as 70 kg for adults and 12 kg for toddlers.

The dermal contact with beddings should be given attention and be further investigated. The dermal exposure values of FRs were up to 0.04 ng/kg bw/day (Table 3), which were much lower than FR intake via dust ingestion. The dermal exposure values were lower than FR intake via dust ingestion. The dermal exposure values during sleeping were estimated as 0.02, 0.23, 0.12, 0.04, 0.02, and 0.07 ng/kg bw/day for TCIPP, TDCIPP, TPHP, and TEHP, respectively, with the assumption that all body surface was in contact with beddings (Table 3). The dermal exposure values during sleeping were generally lower than dust ingestion values for adults and toddlers. Notably, comparable exposure was observed for AC dust, the dermal exposure values of FRs and the PCA results showed that ACs and beds are the main reservoirs of BFRs and PFRs in bedrooms, respectively. FRs in window dust and AC filters, printer tables, PC tables, and beddings were calculated as:

$$E - \text{dermal} = C \times SA \times DAS \times F \times EF / BW$$

where E-dermal is the dermal exposure value (ng/kg bw/day), C is the concentration of contaminant in dust from PC table, printer table, or bed (ng/g), SA is the skin surface area exposed (cm²), DAS is the dust amount adhered to skin (g/cm²), F is the fraction absorbed by the skin (unitless), EF is the fraction of time spent in offices or in beds (unitless), BW is the average body weight (kg). EF and BW were the same with those in the calculation of dust ingestion. The other parameters were described in detail in Supplementary Material.

The high exposure values via dust ingestion for adults and toddlers were 0.28, 1.20, 0.18, 0.02, 0.13, 0.11, and 0.01 ng/kg bw/day and 7.37, 31.2, 4.49, 4.36, 4.54, 3.68, 3.25, and 2.80 ng/kg bw/day for BDE 209, DBDPE, TCEP, TDCIPP, TPHP, EHDPP, and TEHP, respectively (Table 3). If assumed people contact with printer or PC tables only in offices, the dermal exposure values of FRs were up to 0.04 ng/kg bw/day (Table 3), which were much lower than FR intake via dust ingestion. The dermal exposure values during sleeping were estimated as 0.02, 0.23, 0.12, 0.04, 0.02, and 0.07 ng/kg bw/day for TCIPP, TDCIPP, TPHP, EHDPP, and TEHP, respectively, with the assumption that all body surface was in contact with beddings (Table 3). The dermal exposure values during sleeping were generally lower than dust ingestion values for adults and toddlers. Notably, comparable exposure was observed for AC dust, the dermal exposure values of FRs and the PCA results showed that ACs and beds are the main reservoirs of BFRs and PFRs in bedrooms, respectively. FRs in window dust and AC filter dust are likely from the same sources in bedrooms. In offices, FRs in dust and AC filter dust are likely from the same sources in bedrooms. In offices, the printers appear to be the most important source of most FRs in offices. Floor dust seems to be reservoir of FRs from various sources in both bedrooms and offices. The dermal contact with beddings and furniture would bring significant exposure risks for TCIPP, TDCIPP, and TEHP, indicating dermal exposure should be involved in human exposure assessment.

4. Conclusions

Dust from different locations in bedrooms (AC filters, beddings, floors, and windows) and offices (AC filters, printer tables, PC tables, floors, and windows) had different levels and composition profiles of FRs. The composition profiles of FRs and the PCA results showed that ACs and beds are the main reservoirs of BFRs and PFRs in bedrooms, respectively. FRs in window dust and AC filter dust are likely from the same sources in bedrooms. In offices, the printers appear to be the most important source of most FRs in offices. Floor dust seems to be reservoir of FRs from various sources in both bedrooms and offices. The dermal contact with beddings and furniture would bring significant exposure risks for TCIPP, TDCIPP, and TEHP, indicating dermal exposure should be involved in human exposure assessment.

Acknowledgments

This study was financially supported by the National Science Foundation of China (No. 41603085, 41230639, 41573088, and 21307037) and Natural Science Foundation of Guangdong Province.
Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.05.167.

References


