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Selected Organohalogenated Flame Retardants in Egyptian Indoor and Outdoor Environments: Levels, Sources and Implications for Human Exposure

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Highlights

- Indoor levels of flame retardants were significantly higher than outdoors.
- PBDEs>NFRs and BDE-47, 99, HBB, BTBPE and DDC-CO were the most abundant analytes.
- Significant log-linear $K_{dust\text{-}air}$-K_{OA} relationships indicated an equilibrium state.
- Low adverse health effects from inhalation/ingestion/dermal exposure pathways.

Abstract

There is scant information on the presence of the polybrominated diphenyl ethers (PBDEs) and other alternative flame-retardants (NFRs) in Africa. Hence, to investigate their levels, sources, and human exposure scenarios, elevated fine dust (EFD) samples from apartments ($n = 12$), working places ($n = 9$) and cars ($n = 12$), floor dust (FD) samples ($n = 5$) and outdoor dust samples ($n = 21$) were collected from Alexandria, Egypt during 2014. Gaseous concentrations were estimated using low density polyethylene sheets ($n = 33$ and 21 for indoor and outdoor sites respectively). Indoor gaseous and dust PBDE (7.0 – 300 pg/m^3; 4.0 – 770 ng/g), and NFR (0.40 – 48 pg/m^3; 0.50 – 8.5 ng/g) concentrations were significantly higher ($p = 0.004 – 0.02$) than outdoor concentrations (PBDEs: 3.0 – 41 pg/m^3, 1.5 – 195 ng/g; NFRs: 0.20 – 13 pg/m^3, 0.50 – 4.0 ng/g). Median PBDE concentration in cars (210 ng/g) was higher in apartments and working places (129 and 131 ng/g respectively). PBDE concentrations in FD were 7.0 – 14-folds lower than EFD concentrations. Outdoor PBDE concentrations were significantly higher ($p < 0.01$) at residential – industrial places with older buildings. All samples were dominated by BDE-47 and 99. HBB, BTBPE and DDC-CO were the most abundant NFRs in EFD samples. Profiles of PBDE and NFR in FD closely matched those of outdoor dust, indicating a possible carryover from the outdoor environment. Although factors such as number of electronics, construction year and floor type significantly correlated with the majority of PBDE congeners and some NFRs in apartments and working places, sources were not clearly identified for NFRs. Significant log-linear relationships were obtained between theoretical and calculated dust-air partitioning coefficients for all samples indicating an equilibrium state between dust and vapor. Low possibility of occurrence of adverse health effects was concluded, with the inhalation pathway (for adults) and dust ingestion (for children) acting as the most important exposure routes.

Keywords: PBDEs, NFRs, LDPE, Indoor, Outdoor, dust, health effects

1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been widely used worldwide since the 1970s (de Wit, 2002) to reduce fire risks. Although their production has phased out, they still exist in products and thus become released into the environment (Abbasi et al., 2015). Due to the phase out of PBDEs (Abbasi et al., 2015), alternative halogenated (NFRs) and organophosphate flame retardants (OPFRs) are now widely used in consumer products as substitutes (McDonough et al., 2016). PBDEs and NHRs can accumulate in humans (Leonetti et al., 2016; Liu et al., 2015) and thus numerous adverse health effects could occur (Costa and Giordano, 2007; Nakari and Huhtala, 2010).

PBDEs and NFRs have been detected in the indoor air (Al-Omran and Harrad, 2016; Cao et al., 2014; Fromme et al., 2014; Hassan and Shoeib, 2015; Kademoglou et al., 2017; Shoeib et al., 2012) and outdoor air and dust worldwide (Kurt-Karakus et al., 2017; Mahmood et al., 2015; McDonough et al., 2016; Salamova et al., 2014). The indoor environment with expected higher concentrations and longer exposure periods received special attention (Wilford et al., 2004).

The majority of the indoor studies concentrated on the floor dust (FD) (Ali et al., 2013; Cequier et al., 2014; Dodson et al., 2012; Fromme et al., 2014; Hassan and Shoeib, 2015; Kurt-Karakus et al., 2017; Shoeib et al., 2012). Few studies investigated (elevated) fine dust collected ~ 1 meter above the ground (Al-Omran and Harrad, 2016; Newton et al., 2015; Thuresson et al., 2012). In countries with high ambient particulate levels and dust content like Egypt, we hypothesized that floor dust will not properly represent the indoor environment due to the possible carryover of dust and dirt by the shoes from outdoors. However, although dust collected from elevated surfaces would be a better representative of exposure for adults, children are more exposed to the floor dust.

Low density polyethylene passive samplers (LDPE) have been widely used for monitoring hydrophobic organic compounds (HOCs) in the atmosphere (Bartkow et al., 2004; Kennedy et al., 2007; Khairy and Lohmann, 2014, 2013; McDonough et al., 2016). However, they have been rarely used to monitor indoor air pollution. HOCs accumulate in passive air samplers via diffusion and absorption into the sampler matrix, with a high enrichment in LDPE (Sacks and Lohmann, 2011) thus offering lower detection limits compared to conventional samplers. Accordingly, LPDE can be used as a very useful tool in screening level risk assessment studies.

Although PBDEs and NFRs were extensively investigated in the indoor dust and the ambient air worldwide, only few studies investigated their concentrations in the indoor dust (Abdallah and Covaci, 2014; Hassan and Shoeib, 2015) and ambient air (Arinaitwe et al., 2014; Pozo et al., 2009) collected from Africa. Additionally, none of the studies investigated their levels in the elevated fine dust (EFD), outdoor dust and/or their gaseous levels in the indoor and outdoor environments.

In the current study, we studied the levels of PBDEs and NFRs in the indoor and outdoor environments of Alexandria, Egypt. Vapor phase (using LDPE), in- and outdoor dust were collected to: i) determine concentrations, profiles and sources of PBDEs and NFRs in the indoor and outdoor air (gas phase + dust), ii) compare their concentrations among the different indoor microenvironments, and between floor dust and elevated fine dust, iii) assess the link between outdoor dust and indoor contamination, and iv) estimate the human intakes via inhalation and dust ingestion for adults and toddlers. To our knowledge, this is the first comprehensive study to measure PBDEs and NFRs in the indoor and outdoor environments, and to utilize LDPE as an indoor passive sampler.

2. Materials and methods

Detailed descriptions of the study area, LDPE passive sampling, extraction, cleanup, instrumental analysis, and estimation of the vapor phase concentrations from LDPE are provided in Appendix A, and are briefly summarized below.

2.1. Sampling

Indoor and outdoor samples were collected from Alexandria, Egypt (Figure A1) during the late autumn – winter (minimum ventilation effect) of 2014 (November – December). EFD was collected 1 m above the floor from homes (n = 12) and working places (n = 9) using an Electrolux JetMaxx Bag Canister Vacuum cleaner (Model EL4042A). Dust was collected in cellulose thimbles fitted between the crevice tool and the vacuum tube. The thimble was secured with a stainless-steel ring. At each sampled homes, dust was collected from the living area including the living room, dining room and the bedrooms, where people spend most of their indoor times (greatest exposure likelihood), and many products exist that may be sources of the target flame retardants. Prior to sampling, participants were asked not to vacuum or ventilate their places for a week. The crevice tool was passed on all the available surfaces in the rooms. After each of the 6 dust collection events at each location, the thimble was carefully removed, wrapped in a pre-combusted aluminum foil, sealed in plastic zip bag, and stored at −4 °C. After the final sampling event, thimbles were stored at -20 °C until analysis. Similarly, EFD from cars (n = 12) were obtained from all available surfaces except the floor (to prevent the effect of carryover from street dust). Participants were asked not to vacuum their vehicles at least two weeks before sampling and until the end of the sampling event (45 days). 15 indoor samples (other than what was analyzed in the current study) were excluded from further consideration because volunteers failed to follow our pre-sampling guidelines (see Appendix A).

FD was collected from selected homes (n = 5) using the same vacuum cleaner equipped with dust collection bag (Schreder and La Guardia, 2014). Information about the sampled indoor microenvironments is given in Table A1.

For outdoor samples (n = 21), settled dust on elevated smooth surfaces were collected using small plastic brushes and a small aluminum shovel from the same locations as the indoor samples, and stored in small amber glass jars at -20 °C until analysis.

For the outdoor vapor – phase samples, LDPE sheets were pre-cleaned, spiked with performance reference compounds (PRCs) according to Booij et al. (2002) and deployed in duplicates in the atmosphere for 2 months at the same sites as the outdoor dust (n = 42) in two inverted bowls as a shelter for protection against sunlight and precipitation (see Appendix A for more details).

Indoor LDPE sheets were deployed in duplicates for 2 months at each of the sampled rooms (n = 2 – 3) of the living area starting two weeks before the dust sampling campaign. Samplers were hung in protected places away from direct exposure to the light and direct contact with participants. The total number of deployed LDPE were 46 (23 x duplicates) at apartments, 24 at the cars, and 38 at working places (total = 108 samples). For the extraction/analysis step, all the LDPE sheets deployed at the different rooms within an apartment or working place were combined and analyzed together (n = 12, 9 and 12 for apartments, working places and cars respectively x duplicates).

2.2. Extraction, cleanup and instrumental analysis

Dust samples (indoor and outdoor) were sieved through a 3-inch, 100 mesh (150 µm) stainless steel sieve (Dodson et al., 2012), and ~ 0.5 g dry weight were sonicated with n-hexane/acetone (1:1, v:v) after spiking with surrogate standards (10 µL of a 2.0 ng/µL solution of $^{13}C_{12}$ BDE-28, 47, 99, 153 and 183 in nonane), purified and fractionated (fraction1 containing target analytes and fraction 2 containing organophosphate flame retardants, which is not discussed in the current manuscript) over silica gel and concentrated to a final volume of ~ 25 µL after the addition of the injection standards. LDPE samplers were spiked with surrogate standards and cold extracted twice with methylene chloride and n-hexane for 24 h each with no further cleanup.

Extracts were analyzed for 12 PBDE congeners (BDE-2, 8, 15, 28, 30, 47, 49, 99, 100, 153, 154 and 183) and 9 NFRs [tetrabromo-*p*-xylene (TBX), pentabromobenzene (PBB), pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), hexabromobenzene (HBB), hexachlorocyclo-pentadienyl-dibromocyclooctane (DBHCTD), 1,2-bis (2,4,6-tribromophenoxy) ethane (BTBPE), and anti- and syn-isomers of Dechlorane Plus (a-DDC-CO and s-DDC-CO)] on an Agilent 6890N gas chromatograph coupled to a Waters Quattro Micro mass spectrometer (GCMS/MS) in electron ionization mode (EI, 70 eV) using multiple reaction monitoring (MRM).

2.3. Quality assurance

Field blanks (Table A2), matrix spikes, and duplicate samples were included with each sample batch. Field blanks were composed of anhydrous sodium sulphate (3 g) contained in either a thimble, dust bag or a glass jar (based on the type of the sample). To prevent cross contamination between the sampling locations, cleaning of the vacuum cleaner was performed before and after ach sampling event (at each location) by using a detergent, warm water, methanol and air-drying.

Matrix spikes were prepared by spiking ground sea sand (1 g) contained in a thimble, dust bag or glass jar with 15 µL of a solution composed of all the target analytes at a concentration of 2.0 ng/µL in nonane, and performing the same analytical steps as the samples. All the LDPE were deployed and analyzed in duplicates. For the dust samples, 30 % of the total number of samples were analyzed in duplicates. Instrumental calibrations were checked by injection of the continuing calibration solution. The GC/MSMS calibration was verified before, during, and after each analytical sequence. Three calibration standards were continuously injected every 10 dust samples and 15 LDPE samples and the calibration check was maintained within ±15% for all analytes of interest. When an analyte was not detected, the noise was quantitated for the limit of detection (LOD) calculations. LODs were determined in the different matrices as the upper limit of the 95% confidence interval for the field blanks (Table A3). We used ½ the LOD for concentrations that were below the detection limit. Recoveries of the surrogate standards generally ranged from 74 - 92% and 71 – 96 % for the outdoor and indoor LDPE respectively, 63 – 86 % for the outdoor dust, 64 – 91 % for the indoor fine dust and 67 – 96 % for the floor dust. Matrix spikes recoveries (n = 5 for LDPEs and 6 for dust) ranged from 82 % (BDE 2) to 102 % (BDE 154) for the LDPE and from 76 % (BDE-2) to 103 % (BDE-100) for the dust (Table A4) with a relative standard deviation < 20%. Results of the replicate analysis of LDPE and the dust samples indicated that the reproducibility of the analysis ranged from 17.6 % – 25.3 %.

2.4. Human health risk assessment

To calculate the daily exposure doses from inhalation, ingestion and dermal exposure pathways, the following models were used according to the USPEA risk assessment guidelines (Means, 1989):

$$DD_{inh} = \frac{C_g\ .\ ET\ .\ InhR}{BW} \longrightarrow 1$$

$$DD_{ing} = \frac{C_{EFD}\ .\ ET\ .\ IngR}{BW} \longrightarrow 2$$

$$DD_{der} = \frac{C_{EFD}\ .\ ET\ .\ SA\ .\ AF_d\ .\ ABS}{BW} \longrightarrow 3$$

Where,

DD_{inh}, DD_{ing} and DD_{der} are the daily dose associated with the inhalation, ingestion and dermal contact (all in ng/ kg/d) from the different micrenvironments, C_g = the vapor phase concentration of each individual flame retardant (ng/m^3), ET = the exposure time (h/d), InhR = inhalation rate (m^3/d), BW = body weight (kg), C_{EFD} = the concentration of each flame retardant in the elevated fine dust (ng/g), IngR = ingestion rate (mg/d), ABS = dermal adsorption fraction (dimensionless), SA = skin surface area exposed (cm^2), AF_d = particle-to-skin adherence factor (g/cm^2/event).. Note that DD_{ing}, DD_{ing} and DD_{der} are the sum of inhalation, ingestion or dermal contact of vapor phase and elevated fine dust from homes, working places (for adults only), cars and schools.

The ET was obtained from the answers of the participants in the questionnaire. The average working hours were set as 8.0 and 6.0 hours/day for males and females respectively. For housewives, no working hours were included. For all adults and children, ET in cars was set at 2 hours/day. The remainder was the time spent at home. For toddlers, all the time was spent at home. DW was 70, 15 and 12 kg for adults, children and toddlers respectively (Means, 1989). Selected inhalation rates were 13.3 and 10.9 m^3/day for adults and children (toddlers) respectively (Means, 1989). In the best-case scenario, ingestion rates were set at 0.02 and 0.05 d/day for adults and children, whereas they were set at 0.05, 0.1 and 0.2 g/d for adults, children and toddlers respectively(Means, 1989). For the dermal exposure pathway, SA used were 8,620 and 4,970 cm^2 for adults and children(Means, 1989), AF_d (Means, 1989) was 0.000096 g/cm^2 and ABS was 0.03 (Cequier et al., 2014). Available reference dose values (RfD, mg/kg body weight/day) for BDE-47, 99, 153 and HBB were obtained from the Integrated Rias Assessment Information System (IRIS) of the USPEA (https://www.epa.gov/iris, accessed in 10/01/2017). For BTBPE, RfD was obtained from Ali et al. (2012).

2.5. Statistical analysis

Analysis of variance (ANOVA) and the student t-test were performed with SigmaPlot 11. Before applying the statistical tests, all numbers were log transformed for normality. Bivariate correlation was performed with IBM SPSS (V 23, USA).

3. Results and discussion

3.1. Indoor concentrations

3.1.1. PBDEs: ∑12 PBDE concentrations in the vapor phase ranged from 7.0 - 300 pg/m^3, 36 – 220 pg/m^3 and 13 – 303 pg/m^3 at apartments, working places and cars respectively (Table 1). In the EFD, concentrations ranged from 4.10 – 710, 80 – 540 and 3.0 – 770 ng/g respectively (Table 2). Concentrations of PBDEs (2.0 - 78 ng/g) in floor dust (Table A5) were lower than in EFD by a factor of 7.0 – 14 as was previously observed by Bjorklund et al. (2012) in Sweden. Working places (120 pg/m^3 compared to 66 – 85 pg/m^3 at the other microenvironments) and cars (210 ng/g compared to 129 – 131 ng/g for the other microenvironments) showed the highest median PBDE concentrations in the vapor phase and EFD samples respectively as was previously observed in the vapor phase samples of Sweden (Thuresson et al., 2012), and dust samples in Cairo, Egypt (Hassan and Shoeib, 2015), and in UK (Stuart et al., 2008). Vapor phase concentrations of BDE-2, 8 and 30 were significantly higher (One-Way Repeated Measures of

ANOVA; $p = 0.004 – 0.025$) at apartments probably due to limited ventilation at apartments compared to working places and cars.

Detected median vapor phase and EFD concentrations of $\sum_{12}$PBDE and the individual congeners were generally within the same range or lower than concentrations previously reported worldwide at working places and apartments/houses (Figure 1a,b and Table A6). In contrast, PBDE concentrations in the EFD samples of the cars were much higher than concentrations previously observed in Asia and in Cairo, Egypt (Figure 1c). PBDE concentrations in the FD samples were lower than previously detected concentrations in Europe, Asia and USA (Figure 1d) but close to what was previously detected in Cairo, Egypt (Hassan and Shoeib, 2015). However, it should be pointed out that the vapor phase sampling technique used in our study (LDPE passive samplers) is different from the active or passive sampling techniques (using PUFs) used in the other studies and this should be considered when comparing the results. Additionally, differences in the sampling periods, sampled cars (manufacturer, year and mileage), and conditions of the investigated apartments/houses are all significant factors and should be considered in this comparison. Accordingly, we tried to limit our comparisons (Figure 1) to studies with very close sampling periods whenever possible.

BDE-47 and BDE-99 were the dominant congeners in the vapor phase, EFD and FD samples (Figure 2) comprising on average 46 - 84% of the total concentrations. In the EFD samples, BDE-99 was the dominant congener compared to BDE-47 in the vapor phase and FD samples. The ratio of BDE-47 to BDE-99 for the vapor phase (1.0 – 1.7) and FD samples (1.3 – 1.9) were slightly higher than values observed for the penta-formulations (0.6 and 1.0, La Guardia et al., 2006), whereas values in the EFD samples were consistent with the penta-formulation (0.4 – 0.9), which is probably attributed to the slightly higher volatility of BDE-47 (log vapor pressure

= -3.61 pa) compared to BDE-99 (log vapor pressure = -4.30 pa) (Yue and Li, 2013). Our findings indicate the possible wide usage of the penta formulation in the imported consumer goods to Alexandria. Additionally, differences in the ratio values between EFD and FD samples suggest a different PBDE sources. BDE-183 (octa- formulation) showed minor contributions (< 10 %) (Figure 2) indicating probably a limited usage of the octa-BDE formulation, in-line with results reported for floor dust in Cairo (Hassan and Shoeib, 2015).

3.1.2. NFRs: Concentrations of NFRs ranged from 0.40 to 47 pg/m^3, 0.60 – 10 ng/g and 0.50 – 2.0 ng/g in the vapor phase, EFD ad floor dust samples respectively (Tables 1, 2 and A5). HBB displayed the highest concentrations (Figure 3) in the vapor phase samples, as was previously observed in Norwegian schools (Cequier et al., 2014). In all the microenvironments, PBEB, PBT and TBX were less abundant than HBB but showed significant contributions in the vapor phase (Figure 3a, c, f). BTBPE, DBHCTD, a-DDC-CO and s-DDC-CO were below LOD in all the samples probably due to their low vapor pressure (vapor pressure = -5.5 to -10.1 Pa) (McDonough et al., 2016) and accordingly, their preferential occurrence in the particulate phase as was observed in the current study (Figure 3b, d, f, g). Higher contribution of s-DDC-CO in the EFD of cars and working places was observed in the current study probably due to the usage of the technical DDC-CO as an additive in textile, automotive and in computers (Hassan and Shoeib, 2015).

Detected concentrations of NFRs in air and dust of the microenvironments were much lower than concentrations previously recorded worldwide (Table A7). However, EFD median concentrations of HBB (0.25 ng/g), BTBPE (1.0 ng/g), s-DDC-CO (2.0 ng/g) and a-DDC-CO

(0.9 ng/g) in cars of Alexandria were comparable with those observed in Cairo, Egypt (0.23, 1.3, 1.5 and 1.0 respectively) (Hassan and Shoeib, 2015). Additionally, median concentrations of HBB and BTBPE were very close to values reported for the FD samples of Cairo, Egypt (Hassan and Shoeib, 2015), whereas, the median concentrations of s- and a-DDC-CO in Cairo were 3.0 and 7.0 fold respectively higher than our reported values. Egypt does not manufacture flame retardants and any detected compounds are due to imported goods. Nevertheless, the lower observed concentrations in the current studies could be the reason of different materials imported, different manufacturing countries, ages and types of materials and electrical equipment used in the different studies, type, age, mileage and manufacturer of the vehicles and/or less strict fire safety standards in Egypt.

3.2. Outdoor concentrations

3.2.1. PBDEs: PBDE concentrations ranged from 3.0 - 41pg/m^3 and 1.5 - 200 ng/g in the vapor phase and dust samples respectively (Table 3). BDE-2 was below the LOD in all the outdoor samples. Vapor phase and dust concentrations of $\sum_{12}$PBDE, BDE-47, 49, 99, 100, 153 and 154 were significantly higher (t-test, $p < 0.05$) in the residential – industrial regions. This is probably attributed to the older age of the industrial buildings, ventilation systems and/or the number of items containing PBDEs in the industrial buildings. In all samples (Figures A3, A4), BDE-47 and BDE-99 dominated the profiles of vapor phase and dust samples in the outdoor environment as was previously observed for the indoor microenvironments (Figure 2). However, in the outdoor dust samples, BDE-47 showed higher concentrations that BDE-99 as was observed for the indoor floor dust samples (Figure 2g). Values of BDE-47/BDE-99 ratio (1.0 – 2.4) at all the locations were slightly higher than values of the penta-BDE formulations (La Guardia et al., 2006), EFD samples (0.4 – 0.9), but consistent with the FD samples (1.3 – 1.9) and the outdoor gaseous samples (1.0 – 1.9). The ratio values observed here were probably the result of the faster photodegradation of BDE-99 compared to BDE-47 (Dickhut et al., 2012). BDE- 28, 100 and 153 showed also significant contributions in the outdoor dust samples (8.0 – 15 % of the total concentrations).

Detected vapor phase concentrations in the current study were much lower than previously detected concentrations in USA (Batterman et al., 2009), China (Hong et al., 2016) and Turkey (Kurt-Karakus et al., 2017) (Table A6) but higher than values previously observed for air around the lower Great Lakes (0.10 – 14.5 pg/m^3) (McDonough et al., 2016) and Ottawa, Canada (<LOD – 4.4 pg/m^3) (Wilford et al., 2004).

3.2.2. NFRs**:** NFRs concentrations ranged from <LOD - 13 pg/m^3 and 0.40 – 4.0 ng/g in the outdoor vapor and dust samples respectively (Table 3), with no significant difference between the residential and residential-industrial areas as was previously observed for PBDEs. Concentrations of all the investigated NFRs were much lower than the indoor microenvironments (Table 2), and were also lower than PBDE concentrations (Table 3). Profiles of NFRs in the outdoor environment (Figures A5, A6) were similar to the profiles of NFRs in the vapor phase (dominance of HBB, PBB, PBEB and PBT) and dust samples (dominance of BTBPE, s-DDC-Co and a-DDC-CO) of the indoor microenvironments.

Detected concentrations of PBB, HBB, PBT and PBEB in the current study were much lower than concentrations previously observed in China (Table A7) (Hong et al., 2016; Lin et al., 2013), Stockholm, Sweden (for PBT: <LOD in the current study compared to 0.17 pg/m^3) (Newton et al., 2015), Istanbul, Turkey (for HBB and BTBPE; Table A7) and Great Lakes (for

BTBPE, DBHCTD, s-DDC-CO and a-DDC-CO) (McDonough et al., 2016). In contrast, the median concentration of HBB in the current study (0.10 pg/m^3) was higher than that in Sweden (<LOD), and the range of PBB, HBB and PBEB in the current study was also higher that concentrations observed at the lower Great Lakes. Again, much lower concentrations of NFRs demonstrate that these compounds have not (yet) replaced PBDEs as flame retardants in Alexandria.

3.3. Indoor vs Outdoor Organohalogenated Flame Retardants

The lack of correlations between detected vapor concentrations and their subcooled vapor pressures (P_L, Pa) imply that emissions of PBDEs and NFRs were dominated by various source strengths, reactivity but not their volatility. All the gaseous and EFD concentrations of $\sum_{12}$PBDEs and $\sum$NFRs in the indoor samples (excluding cars) were significantly higher (t-test, t = 2.95 – 7.13, $p < 0.006$) than the outdoor samples (Figure A7). This, in addition to the observed indoor - outdoor pattern variabilities and the between-samples variability in the indoor microenvironments suggests that detected indoor concentrations are mainly from indoor emissions with minimum contribution from the outdoor environment.

In the current study, several findings were observed when we compare indoor and outdoor dust. First, different patterns of PBDE congeners (Figure A7c) were observed in the elevated fine versus floor dust samples (dominance of BDE-47 in FD compared to BDE-99 in the EFD), whereas for the NFRs (Figure A7d) similar patterns were observed in the EFD, FD and outdoor dust samples. Second, greater similarities in concentrations and profiles (Figure A7c, d) of PBDEs and NFRs were observed between floor and outdoor dust (dominance of BDE-47, BTBPE, HBB and s-DDC-CO and similar BDE-47/99 ratios). This finding is unusual considering the higher expected concentrations in the indoor samples, which is already observed for the gaseous and the elevated fine dust. Additionally, FD concentrations of HBB, BTBPE, DBHCTD and BDE-28 to 183 were more strongly correlated with the outdoor samples (r = 0.74 – 0.99, $p < 0.05$) than with elevated fine dust samples (r = -0.08 – 0.90), which were not significant ($p > 0.05$) in most cases. The exception were a-and s-DDC-CO, which displayed stronger correlation to elevated fine dust samples (r =0.80, 0.87 compared to r = 0.67 and 0.68 for outdoor samples), although they were statistically insignificant ($p > 0.05$). Third, elevated fine dust samples displayed much higher concentrations than floor and outdoor dust and slightly different profiles. Since all samples were sieved, we argue that different sources are responsible for the detected levels of PBDEs and NFRs in the EFD and FD. Floor dust is possibly contaminated by outdoor dust and dirt carried indoors by shoes, whereas this process is of minimum effect for the elevated fine dust (based on our requested pre-sampling and during-sampling ventilation conditions from volunteers).

3.4. Sources and relations between organohalogenated flame retardants in the samples

3.4.1. Indoor

To study the relations between detected flame retardants, a bivariate analysis was performed (correlations). Factors such as year of construction, number of foamed furniture, number of electronics, type of ventilation used and the number of vacuuming per week were included (Table A1). For cars, the make, year, type of seats and the ventilation system were included. All this information was obtained from the questionnaires given to the volunteers. For the indoor air (Table A8) and EFD (Table A9) samples, significant correlation between the tri- through hepta-brominated congeners (BDE- 28 to BDE- 183) was observed indicating possible common sources, the banned penta- and octa- formulations. Although a significant correlation was observed between all tri- through hepta- brominated congeners, we excluded the possibility of occurrence of debromination and conversion of the higher brominated congener (BDE-183) to lower brominated ones as this finding occurred both in the indoor and outdoor environments (see below). No significant correlations were observed between PBDE congeners in the gaseous and EFD samples and the number of electronics and the number of foamed furniture. However, some within variability was observed at working places. Concentration of PBDE congeners at the car upholstery shop (∑12 PBDE = 220 pg/m^3) was significantly higher (One-Way Repeated Measures of ANOVA; $p < 0.001$) than those recorded for the four sampled school offices (∑12 PBDE = 36 - 120 pg/m^3).

In the elevated fine dust samples (Table A9), all the PBDE congeners were negatively correlated with the construction year ($r = 0.43 – 0.62$, $p < 0.05$), probably due to the increased dependence on NFRs and OPFRs in recent years. Nevertheless, the majority of the investigated NFRs showed no significant correlations with the construction year. Most of the PBDE congeners showed significant correlations with the floor type (Table A9), where concentrations increased when floors were covered with carpets, probably relating to the use of PBDEs in carpet paddings (Stapleton et al., 2008).

For NFRs, only PBEB, s- and a-DDC-CO in the EFD samples showed significant correlations ($r = 0.47 – 0.68$, $p < 0.05$) with the number of electronics, probably due to their wide usage in electronics (Sverko et al., 2011). Although BTBPE was used as a replacement for the octa-BDE formulation by the end of 2004 (Kurt-Karakus et al., 2017), no correlation were observed with BDE-183 in the current study. Additionally, median concentrations of BTBPE in EFD of the indoor microenvironments (0.80 – 3.0 ng/g) and the outdoor dust (0.20 – 0.26 ng/g) were much lower than median concentrations of BDE-183 (2.2 – 25 ng/g and 0.50 – 2.50 ng/g respectively), which may be related to the limited usage of the octa-PBDE formulation in the imported goods in Egypt.

The detected concentrations of NFRs in the current study are more affected by the within-site variabilities and thus sources were not clearly identified for this class of flame-retardants. A similar finding was observed in Norway (Cequier et al., 2014).

As for cars, manufacturer was the key factor for the gaseous organohalogenated flame retardants (Table A14), where vapor phase concentrations of BDE-28 through BDE-154 in the Korean cars were significantly higher (One-Way Repeated Measures of ANOVA; $p < 0.004$) than the German and Japanese cars sampled here (Table 1). Higher concentrations of the same congeners were also observed in the EFD samples collected from the Korean cars compared to the German and Japanese cars but was statistically insignificant (Table 2). Like the apartments and working places, several significant positive correlations were observed for the different NFRs (Tables A10, A11) indicating common sources. Additionally, the significant correlations between the individual penta- BDE congeners were also observed for the air and dust in cars. HBB in air and dust showed significant positive correlation with the model year ($r = 0.677 – 0.74$, $p < 0.05$), whereas DBHCTD and a-DDC-CO in the dust samples were significantly higher ($r = 0.58 – 0.85$, $p < 0.05$) in the Korean cars.

3.4.2. Outdoor

No correlations were found with temperature, wind speed or wind direction (data not shown). Generally, some significant between-analyte correlations were observed for NFRs and for the individual penta-BDE congeners and BDE-183. As we mentioned earlier, industrial activities were only a major source of PBDEs (significantly higher concentrations at the residential –

industrial areas than the residential areas) in the outdoor environment, whereas this pattern was not observed for NFRs.

3.4. Partitioning of the Organohalogenated Flame Retardants in Air and Dust

To investigate the relationship between the detected concentrations in the dust and air samples, dust-air partitioning coefficients ($K_{dust\text{-}air}$; m^3/g) were calculated [concentrations in the dust (ng/g)/air concentrations (ng/m^3)]. For equilibrium conditions, $K_{dust\text{-}air}$ is expected to be directly proportional to the octanol-air partition coefficient (K_{OA}) (Cequier et al., 2014; Liagkouridis et al., 2017; Weschler and Nazaroff, 2010). In contrast, if the gaseous fraction was freshly released, or if the flame retardants in the air and dust were emitted from different sources, this relationship would be insignificant. Calculated log $K_{dust\text{-}air}$ values were compared with a theoretical $K_{dust\text{-}air}$ calculated based on K_{OA}, fraction of organic carbon (f_{OC}) and density of dust (ρ_{dust}, g/m^3) as follows:

$$K_{dust\text{-}air} = (f_{OC} \, . \, K_{OA}) / \rho_{dust} \qquad (4).$$

f_{OC} and ρ_{dust} were selected as 0.2 and 17 x 10^5 g/m^3 (Liagkouridis et al., 2017). Thus, the less volatile flame retardants (higher K_{OA}) will partition preferentially into the dust, thus resulting in higher $K_{dust\text{-}air}$.

In the current study, significant log-linear relationships between the calculated and theoretical $K_{dust\text{-}air}$ (R^2= 0.71 – 0.82 for EFD and FD, and 0.73 – 0.81 for the outdoor samples, $p < 0.001$) were observed for elevated fine and floor dust (Figure 4a) and the outdoor samples (Figure 4b). Additionally, slopes of the regression relationships (b = 0.53 – 0.65) were significantly different from zero ($p < 0.001$) indicating equilibrium distribution of the flame retardants between dust and vapor. Surprisingly, this relationship explained 75 % of the total variability for the elevated fine and floor dust samples, and the slopes were identical (b = 0.60) and insignificantly different from one ($p < 0.001$). Assuming that floor dust was contaminated by outdoor dust, a stronger relationship for EFD samples is expected. However, this was not observed probably due to the similarity between the profiles of PBDEs and NFRs in the vapor phase samples of the indoor and outdoor environments (Figure 2a, b), the similarities between profiles of NFRs in the EFD and FD samples and/or the possible ongoing emission of flame retardants in the indoor microenvironment, which affected the strength of the relationship.

3.5. Human Exposure Assessment

Daily intake of the flame retardants from dust ingestion (based on EFD samples), air inhalation (vapor phase concentrations) and dermal absorption was estimated in the current study for adult males and females (working and housewives), children (age more than 6 years) and toddlers. Calculated daily intakes are given in Table A12. Values generally ranged from 6.8×10^{-6} – 0.17, 2.0×10^{-5} – 0.60, 7.0×10^{-8} – 0.03 and 1.0×10^{-5} – 0.04 ng/kg body weight/day for the ingestion pathway (best case and worst-case scenarios respectively), inhalation and dermal exposure pathways. Calculated daily dose for the three exposure routes were generally higher for PBDEs than NFRs (Figure 5). Toddlers showed the highest ingestion (2.9×10^{-3} – 1.0×10^{-2}), inhalation (1.4×10^{-3} – 5.5×10^{-3}) and dermal (2.1×10^{-4} – 1.3×10^{-3}) daily dose values compared to the other groups. Ingestion of NFRs was the most important exposure route (higher calculated daily dose values; Table A12) for toddlers, children, adult males and housewives (Figure 5a, 6), and for all the groups with respect to PBDEs. In contrast, inhalation (especially of HBB and TBX) was more important for adult working females. For most of the NFRs, comparable daily dose values (from all exposure routes) were observed for adult males and working females, which were higher than values calculated for housewives (Figure 5a, c, e). This observed pattern is probably attributed to the higher observed concentrations of most of the NFRs at the working places. In contrast, the pattern was not observed for PBDEs (Figure 5b, d, f). Generally, inhalation pathway was more important for the more volatile flame retardants (TBX, PBB, HBB, PBT, PBEB, BDE-2, 8 and 15), whereas the ingestion pathway was the most important exposure route for BTBPE, DBHCTD, s-DDC-CO, BDE-28, 47, 99, 100 and 153 (Figure 5). Nevertheless, calculated values for BDE-47, 99, 153 and HBB were at least three orders of magnitude lower than their corresponding oral reference doses (RfD) (Table A12).

Conclusions

To our knowledge, this is the first study reporting levels of PBDEs and NFRs in the air and dust samples from the indoor and outdoor environments in Alexandria, Egypt. Results show that PDBEs are present everywhere at elevated concentrations relative to NFRs targeted here. The exposure assessment study indicated that inhalation of volatile flame retardants is a more important exposure route for adults than dust ingestion. Accordingly, gaseous concentrations of flame retardants should be included in future studies. For that purpose, LDPE could be used as an effective indoor sampling technique in human exposure studies. Inclusion of the outdoor samples indicated that: (i) indoor flame retardants are mainly emitted from the existing indoor sources (higher concentrations in the indoor samples) and (ii) elevated fine dust samples are less influenced by contamination from outdoor dust compared to the floor dust samples. Thus, outdoor samples should be considered in future studies especially at locations lacking emission inventory database. Concentrations of flame retardants in the elevated fine dust were higher than those for the floor dust samples, which could result in underestimated exposures when relying on floor dust. Nevertheless, estimated daily doses from all exposure routes for some of the target analytes were much lower than available reference doses. However, the small number of floor dust samples analyzed in the current study is limiting conclusions. Our results could represent an important baseline database for developing response actions to limit the exposure to the organohalogenated flame retardants in the atmospheric environment of Alexandria, Egypt

Supplementary data

Details on the sampling, chemical and statistical analysis, estimation of gaseous concentrations and human exposure assessment are given in Appendix A.

References

Abbasi, G., Buser, A.M., Soehl, A., Murray, M.W., Diamond, M.L., 2015. Stocks and Flows of PBDEs in Products from Use to Waste in the U.S. and Canada from 1970 to 2020. Environ. Sci. Technol. 49, 1521–1528. https://doi.org/10.1021/es504007v

Abdallah, M.A.-E., Covaci, A., 2014. Organophosphate Flame Retardants in Indoor Dust from Egypt: Implications for Human Exposure. Environ. Sci. Technol. 48, 4782–4789. https://doi.org/10.1021/es501078s

Al-Omran, L.S., 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. Emerg. Contam. 2, 7–13. https://doi.org/10.1016/j.emcon.2015.10.001

Ali, N., Ali, L., Mehdi, T., Dirtu, A.C., Al-Shammari, F., Neels, H., Covaci, A., 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. https://doi.org/10.1016/j.envint.2013.02.001

Ali, N., Dirtu, A.C., Eede, N. Van den, Goosey, E., Harrad, S., Neels, H., ’t Mannetje, A., Coakley, J., Douwes, J., Covaci, A., 2012. Occurrence of alternative flame retardants in indoor dust from New Zealand: Indoor sources and human exposure assessment. Chemosphere 88, 1276–1282. https://doi.org/https://doi.org/10.1016/j.chemosphere.2012.03.100

Ali, N., Harrad, S., Goosey, E., Neels, H., Covaci, A., 2011. “Novel” brominated flame retardants in Belgian and UK indoor dust: Implications for human exposure. Chemosphere 83, 1360–1365. https://doi.org/10.1016/j.chemosphere.2011.02.078

Arinaitwe, K., Muir, D.C.G., Kiremire, B.T., Fellin, P., Li, H., Teixeira, C., 2014. Polybrominated Diphenyl Ethers and Alternative Flame Retardants in Air and Precipitation Samples from the Northern Lake Victoria Region, East Africa. Environ. Sci. Technol. 48, 1458–1466. https://doi.org/10.1021/es403600a

Bartkow, M.E., Hawker, D.W., Kennedy, K.E., Müller, J.F., 2004. Characterizing Uptake Kinetics of PAHs from the Air Using Polyethylene-Based Passive Air Samplers of Multiple Surface Area-to-Volume Ratios. Environ. Sci. Technol. 38, 2701–2706. https://doi.org/10.1021/es0348849

Batterman, S.A., Chernyak, S., Jia, C., Godwin, C., Charles, S., 2009. Concentrations and Emissions of Polybrominated Diphenyl Ethers from U.S. Houses and Garages. Environ. Sci. Technol. 43, 2693–2700. https://doi.org/10.1021/es8029957

Björklund, J.A., Sellström, U., de Wit, C.A., Aune, M., Lignell, S., Darnerud, P.O., 2012. Comparisons of polybrominated diphenyl ether and hexabromocyclododecane concentrations in dust collected with two sampling methods and matched breast milk samples. Indoor Air 22, 279–288. https://doi.org/10.1111/j.1600-0668.2011.00765.x

Booij, K., Smedes, F., van Weerlee, E.M., 2002. Spiking of performance reference compounds in low density polyethylene and silicone passive water samplers. Chemosphere 46, 1157–1161. https://doi.org/http://dx.doi.org/10.1016/S0045-6535(01)00200-4

Cao, Z., Xu, F., Covaci, A., Wu, M., Wang, H., Yu, G., Wang, B., Deng, S., Huang, J., Wang, X., 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, 8839–8846. https://doi.org/10.1021/es501224b

Cequier, E., Ionas, A.C., Covaci, A., Marcé, R.M., Becher, G., Thomsen, C., 2014. Occurrence of a Broad Range of Legacy and Emerging Flame Retardants in Indoor Environments in Norway. Environ. Sci. Technol. 48, 6827–6835. https://doi.org/10.1021/es500516u

Civan, M.Y., Kara, U.M., 2016. Risk assessment of PBDEs and PAHs in house dust in Kocaeli, Turkey: levels and sources. Environ. Sci. Pollut. Res. 23, 23369–23384. https://doi.org/10.1007/s11356-016-7512-5

de Wit, C.A., 2002. An overview of brominated flame retardants in the environment. Chemosphere 46, 583–624. https://doi.org/http://dx.doi.org/10.1016/S0045-6535(01)00225-9

Dickhut, R.M., Cincinelli, A., Cochran, M., Kylin, H., 2012. Aerosol-mediated transport and deposition of brominated diphenyl ethers to Antarctica. Environ. Sci. Technol. 46, 3135–40. https://doi.org/10.1021/es204375p

Dodson, R.E., Perovich, L.J., Covaci, A., Van den Eede, N., Ionas, A.C., Dirtu, A.C., Brody, J.G., Rudel, R.A., 2012. After the PBDE Phase-Out: A Broad Suite of Flame Retardants in Repeat House Dust Samples from California. Environ. Sci. Technol. 46, 13056–13066. https://doi.org/10.1021/es303879n

Fromme, H., Hilger, B., Kopp, E., Miserok, M., Völkel, W., 2014. Polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and “novel” brominated flame retardants in house dust in Germany. Environ. Int. 64, 61–68. https://doi.org/10.1016/j.envint.2013.11.017

Hassan, Y., Shoeib, T., 2015. Levels of polybrominated diphenyl ethers and novel flame retardants in microenvironment dust from Egypt: An assessment of human exposure. Sci. Total Environ. 505, 47–55. https://doi.org/10.1016/j.scitotenv.2014.09.080

Hoh, E., Zhu, Hites, R.A., 2005. Novel Flame Retardants, 1,2-Bis(2,4,6-tribromophenoxy)ethane and 2,3,4,5,6-Pentabromoethylbenzene, in United States’ Environmental Samples. Environ. Sci. Technol. 39, 2472–2477. https://doi.org/10.1021/es048508f

Hong, W.-J., Jia, H., Ding, Y., Li, W.-L., Li, Y.-F., 2016. Polychlorinated biphenyls (PCBs) and halogenated flame retardants (HFRs) in multi-matrices from an electronic waste (e-waste) recycling site in Northern China. J. Mater. Cycles Waste Manag. https://doi.org/10.1007/s10163-016-0550-8

Kademoglou, K., Xu, F., Padilla-Sanchez, J.A., Haug, L.S., Covaci, A., Collins, C.D., 2017. Legacy and alternative flame retardants in Norwegian and UK indoor environment:

Implications of human exposure via dust ingestion. Environ. Int. 102, 48–56. https://doi.org/10.1016/j.envint.2016.12.012

Kennedy, K.E., Hawker, D.W., Müller, J.F., Bartkow, M.E., Truss, R.W., 2007. A field comparison of ethylene vinyl acetate and low-density polyethylene thin films for equilibrium phase passive air sampling of polycyclic aromatic hydrocarbons. Atmos. Environ. 41, 5778–5787. https://doi.org/https://doi.org/10.1016/j.atmosenv.2007.02.033

Khairy, M.A., Lohmann, R., 2014. Field calibration of low density polyethylene passive samplers for gaseous POPs. Environ. Sci. Process. Impacts 16, 414–421.

Khairy, M.A., Lohmann, R., 2013. Feasibility of using low density polyethylene sheets to detect atmospheric organochlorine pesticides in Alexandria, Egypt. Environ. Pollut. 181, 151–158.

Kurt-Karakus, P.B., Alegria, H., Jantunen, L., Birgul, A., Topcu, A., Jones, K.C., Turgut, C., 2017. Polybrominated diphenyl ethers (PBDEs) and alternative flame retardants (NFRs) in indoor and outdoor air and indoor dust from Istanbul-Turkey: Levels and an assessment of human exposure. Atmos. Pollut. Res. 8, 801–815. https://doi.org/https://doi.org/10.1016/j.apr.2017.01.010

La Guardia, M.J., Hale, R.C., Harvey, E., 2006. Detailed Polybrominated Diphenyl Ether (PBDE) Congener Composition of the Widely Used Penta-, Octa-, and Deca-PBDE Technical Flame-retardant Mixtures. Environ. Sci. Technol. 40, 6247–6254. https://doi.org/10.1021/es060630m

Leonetti, C., Butt, C.M., Hoffman, K., Miranda, M.L., Stapleton, H.M., 2016. Concentrations of polybrominated diphenyl ethers (PBDEs) and 2,4,6-tribromophenol in human placental tissues. Environ. Int. 88, 23–29. https://doi.org/http://dx.doi.org/10.1016/j.envint.2015.12.002

Liagkouridis, I., Lazarov, B., Giovanoulis, G., Cousins, I.T., 2017. Mass transfer of an organophosphate flame retardant between product source and dust in direct contact. Emerg. Contam. 3, 115–120. https://doi.org/https://doi.org/10.1016/j.emcon.2017.09.002

Lin, Y., Qiu, X., Zhao, Y., Ma, J., Yang, Q., Zhu, T., 2013. Polybromobenzene Pollutants in the Atmosphere of North China: Levels, Distribution, and Sources. Environ. Sci. Technol. 47, 12761–12767. https://doi.org/10.1021/es403854d

Liu, L.-Y., Salamova, A., He, K., Hites, R.A., 2015. Analysis of polybrominated diphenyl ethers and emerging halogenated and organophosphate flame retardants in human hair and nails. J. Chromatogr. A 1406, 251–257. https://doi.org/http://dx.doi.org/10.1016/j.chroma.2015.06.003

Mahmood, A., Malik, R.N., Syed, J.H., Li, J., Zhang, G., 2015. Dietary exposure and screening-level risk assessment of polybrominated diphenyl ethers (PBDEs) and dechloran plus (DP) in wheat, rice, soil and air along two tributaries of the River Chenab, Pakistan. Chemosphere 118, 57–64. https://doi.org/http://dx.doi.org/10.1016/j.chemosphere.2014.05.071

McDonough, C.A., Puggioni, G., Helm, P.A., Muir, D., Lohmann, R., 2016. Spatial Distribution and Air–Water Exchange of Organic Flame Retardants in the Lower Great Lakes. Environ. Sci. Technol. 50, 9133–9141. https://doi.org/10.1021/acs.est.6b02496

Means, B., 1989. Risk-assessment guidance for Superfund. Volume 1. Human Health Evaluation Manual. Part A. Interim report (Final). United States.

Nakari, T., Huhtala, S., 2010. In vivo and in vitro toxicity of decabromodiphenyl ethane, a flame retardant. Environ. Toxicol. 25, 333–338. https://doi.org/10.1002/tox.20499

Newton, S., Sellstrom, U., de Wit, C.A., 2015. Emerging Flame Retardants, PBDEs, and

HBCDDs in Indoor and Outdoor Media in Stockholm, Sweden. Environ. Sci. Technol. 49, 2912–2920. https://doi.org/10.1021/es505946e

Pozo, K., Harner, T., Lee, S.C., Wania, F., Muir, D.C.G., Jones, K.C., 2009. Seasonally Resolved Concentrations of Persistent Organic Pollutants in the Global Atmosphere from the First Year of the GAPS Study. Environ. Sci. Technol. 43, 796–803. https://doi.org/10.1021/es802106a

Sacks, V.P., Lohmann, R., 2011. Development and use of polyethylene passive samplers to detect triclosans and alkylphenols in an urban estuary. Environ. Sci. Technol. 45, 2270–7. https://doi.org/10.1021/es1040865

Salamova, A., Ma, Y., Venier, M., Hites, R.A., 2014. High Levels of Organophosphate Flame Retardants in the Great Lakes Atmosphere. Environ. Sci. Technol. Lett. 1, 8–14. https://doi.org/10.1021/ez400034n

Schreder, E.D., La Guardia, M.J., 2014. Flame Retardant Transfers from U.S. Households (Dust and Laundry Wastewater) to the Aquatic Environment. Environ. Sci. Technol. 48, 11575–11583. https://doi.org/10.1021/es502227h

Shoeib, M., Harner, T., Webster, G.M., Sverko, E., Cheng, Y., 2012. Legacy and current-use flame retardants in house dust from Vancouver, Canada. Environ. Pollut. 169, 175–182. https://doi.org/10.1016/j.envpol.2012.01.043

Stapleton, H.M., Sjödin, A., Jones, R.S., Niehüser, S., Zhang, Y., Patterson, D.G., 2008. Serum Levels of Polybrominated Diphenyl Ethers (PBDEs) in Foam Recyclers and Carpet Installers Working in the United States. Environ. Sci. Technol. 42, 3453–3458. https://doi.org/10.1021/es7028813

Stuart, H., Ibarra, C., Abdallah, M.A.-E., Boon, R., Neels, H., Covaci, A., 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, 1170–1175.

Sverko, E., Tomy, G.T., Reiner, E.J., Li, Y.-F., McCarry, B.E., Arnot, J.A., Law, R.J., Hites, R.A., 2011. Dechlorane Plus and Related Compounds in the Environment: A Review. Environ. Sci. Technol. 45, 5088–5098. https://doi.org/10.1021/es2003028

Thuresson, K., Bjorklund, J.A., de Wit, C.A., 2012. Tri-decabrominated diphenyl ethers and hexabromocyclododecane in indoor air and dust from Stockholm microenvironments 1: Levels and profiles. Sci. Total Environ. 414, 713–721. https://doi.org/10.1016/j.scitotenv.2011.11.016

Weschler, C.J., Nazaroff, W.W., 2010. SVOC partitioning between the gas phase and settled dust indoors. Atmos. Environ. 44, 3609–3620. https://doi.org/https://doi.org/10.1016/j.atmosenv.2010.06.029

Wilford, B.H., Harner, T., Zhu, J., Shoeib, M., Jones, K.C., 2004. Passive Sampling Survey of Polybrominated Diphenyl Ether Flame Retardants in Indoor and Outdoor Air in Ottawa, Canada: Implications for Sources and Exposure. Environ. Sci. Technol. 38, 5312–5318. https://doi.org/10.1021/es049260x

Yue, C., Li, L.Y., 2013. Filling the gap: Estimating physicochemical properties of the full array of polybrominated diphenyl ethers (PBDEs). Environ. Pollut. 180, 312–323. https://doi.org/http://dx.doi.org/10.1016/j.envpol.2013.05.029