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A Meta-Analysis of Factors Influencing Concentrations of Brominated Flame Retardants and Organophosphate Esters in Indoor Dust

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Abstract

Current assessments of human exposure to flame retardants (FRs) via dust ingestion rely on measurements of FR concentrations in dust samples collected at specific points in time and space. Such exposure assessments are rendered further uncertain by the possibility of within-room and within-building spatial and temporal variability, differences in dust particle size fraction analysed, as well as differences in dust sampling approach. A meta-analysis of peer-reviewed data was undertaken to evaluate the impact of these factors on reported concentrations of brominated flame retardants (BFRs) and organophosphate esters (OPEs) flame retardants in dust and subsequent human exposure estimates. Except for a few cases, concentrations of FRs in elevated surface dust (ESD) exceeded significantly those in floor dust (FD). The implications of this for exposure assessment are not entirely clear. However, they imply that analysing FD only will underestimate exposure for adults who likely rarely ingest floor dust, while analysing ESD only would overestimate exposure for toddlers who likely rarely ingest elevated surface dust. Considerable within-building spatial variability was observed with no specific trend between concentrations of either BFRs or OPEs in living rooms and bedrooms in the same homes, implying that exposure assessments based solely on sampling one room are uncertain. Substantial differences in FR concentrations were observed in different particle size fractions of dust. This is likely partly attributable to the presence of abraded polymer particles/fibres with high FR concentrations in larger particle size fractions. This has implications for exposure assessment as adherence to skin and subsequent FR uptake via ingestion and dermal sorption varies with particle size. Analysing dust samples obtained from a householder vacuum cleaner (HHVC) compared with researcher collected dust (RCD) will underestimate human exposure to the most of studied contaminants. This is likely due to the losses of volatile FRs from HHVC dust over the extended period such dust spends in the dust bag. Temporal variability in FR concentrations is apparent during month-to-month or seasonal monitoring, with such variability likely due more to changes in room contents rather than seasonal temperature variation.

Keywords

BFRs, OPEs, indoor dust, exposure assessment

Highlights

- FR concentrations vary significantly between floor and elevated surface dust
- FR contamination of floor dust can vary significantly by within-room and within-building spatial variability
- The most suitable dust size fraction for exposure assessment is <150 μm

1. Introduction

In recent decades, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) have been the most extensively used additive brominated flame retardants (BFRs) in consumer products such as polyurethane foam, electrical and electronic equipment, textiles, and construction materials. A large number of studies have indicated that PBDEs and HBCDD are persistent and bioaccumulative toxicants, which adversely affect human health (WHO, 2003; van der Ven et al., 2009; Noyes et al. 2010). Health concerns of PBDEs and HBCDD exposure include endocrine disruption (Johnson et al., 2013), impaired fertility (European Commission, 2012), neurotoxic effects (Schecter et al., 2012), and possible carcinogenicity (Li et al., 2014). As a consequence, the manufacture and new use of commercial mixtures of PBDEs (Penta-, Octa-, and Deca-BDE) and HBCDD have been subject to restrictions and listed under the Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP, 2008; 2012; 2019). Such actions have resulted in increased production and use of alternative FRs such as "novel" brominated flame retardants (NBFRs). NBFRs as relating to brominated flame retardants are defined here as those new to the market or recently observed in the environment such as BEH-TEBP (Bis(2-ethyl-1-hexyl) tetrabromophthalate), DBDPE (decabromodiphenylethane). In addition, organophosphate ester (OPEs) compounds which are extensively used as flame retardants and plasticizers. (Ven der Veen and de Boer, 2012; Greaves and Letcher, 2017; Blum et al., 2019). It is, however, expected that PBDE formulations may maintain a presence in new products due to the use of recycled materials, with old PBDE-containing products still in use constituting a further on-going emission source (USEPA, 2010; Pivnenko et al., 2017). The global production of FRs was estimated at 2.8 million tonnes in 2018, and to be growing by 4.6% per year (Yasin et al, 2016). In 2019, BFRs and OPEs represented 17% and 18% respectively of the global production of flame retardants, with Asia the largest market, followed by North America, Europe, Middle East, and Latin America respectively (IHS, 2020; Market Research Future, 2018).

Since BFRs and OPEs are semi-volatile organic compounds (SVOCs), they are – when used additively, i.e. non-chemically bound to the polymer - likely to be released to the environment via a combination of volatilisation, product abrasion, and direct contact with dust (Rauert et al., 2014a; Wei et al., 2015; Liagkouridis et al., 2017). A large number of published studies have reported concentrations of BFRs and OPEs in different environmental matrices (Abdallah et al., 2008; Yang et al., 2014; Harrad et al., 2019; Yadva et al., 2020) and human tissues

(Abdallah and Harrad, 2014; Liu et al., 2015; Alves et al., 2017), with increasing evidence of their toxicity and capacity for bioaccumulation (Ding et al., 2016; Blum et al., 2019).

Several studies have revealed a significant positive correlation between the concentrations of BFRs in indoor dust and human biological samples such as human milk (Toms et al., 2009; Kim et al., 2014) and serum (Stapleton et al., 2012; Ma et al., 2017) suggesting that indoor settled dust ingestion is a major pathway of exposure to such chemicals (Tue et al., 2013; de Boer et al., 2016). Currently, exposure to FRs via indoor dust ingestion is estimated as a multiple of the concentrations of these chemicals present in indoor dust and the rate at which such dust is ingested. Such methodology provides only an indication of the potential exposure via this pathway; with true exposure dependent on a variety of factors such as the amount of time an individual spends in the room from which dust was sampled, the number and extent of hand-to-mouth events that occur (widely assumed greater for toddlers than adults), as well as factors such as the dustiness of the room, whereby dust ingestion is likely greater in dustier rooms. Thus, it is important that indoor dust samples collected and analysed as part of such exposure assessments are truly representative of the dust to which humans are exposed. Moreover, assessment of human exposure to FRs via indoor dust is rendered uncertain because of a lack of knowledge about many potentially influencing factors. Besides the obvious uncertainty inherent in basing estimates of population-level exposure on limited sample sizes, how certain are we that a dust sample taken at a single point in space (e.g. a specific area of a living room) and time (e.g. winter) represents adequately the chronic exposure of a given individual? Specifically, if the FR concentration is measured in another dust sample taken 6 months later from another part of the same living room, or from a bedroom in the same house, how different would be the FR concentration and thus the estimated exposure? In addition, the sample collection method (e.g. participant-provided vacuum cleaner bags or samples collected by a researcher via a standard protocol) and dust particle size fraction analysed may also influence the FR concentrations measured in an indoor dust sample. Furthermore, human exposure estimates based only on floor dust may not reflect the actual exposure to FRs of adults. While toddlers and young children are likely more exposed to floor dust, adults likely have far greater contact with dust from elevated surfaces - e.g. desk and table tops. Hence, if dust concentrations vary significantly between elevated surfaces and the floor in the same room, one type of dust sample may not represent an acceptable estimate of exposure for *all* microenvironment occupants.

Against this backdrop, this paper critically reviews the available literature to evaluate the factors influencing the reported estimates of human exposure to BFRs and OPEs via indoor dust ingestion. We focus on four crucial factors linked to uncertainty in assessments of exposure to BFRs and OPEs via indoor dust ingestion, namely: (a) within-room and within-building spatial variability, (b) dust particle size, (c) sampling method, and (d) temporal/seasonal variability.

2. Methodology

The BFRs and OPEs targeted in this study comprise the following: BDE-47 (2,2',4,4'-tetrabromodiphenyl ether), BDE-99 (2,2',4,4',5-pentabromodiphenyl ether), BDE-153 (2,2',4,4',5,5'-hexabromodiphenyl ether), BDE-154 (2,2',4,4',5,6'-hexabromodiphenyl ether), BDE-183 (2,2',3,4,4',5',6-heptabromodiphenyl ether), BDE-209 (2,2',3,3',4,4',5,5',6,6'-decabromodiphenyl ether), HBCDD (hexabromocyclododecane), EH-TBB (2-ethylhexyl-2,3,4,5-tetrabromobenzoate), BTBPE (1,2-bis(2,4,6-tribromophenoxy)ethane), BEH-TEBP (Bis(2-ethyl-1-hexyl) tetrabromophthalate), DBDPE (decabromodiphenylethane), TNBP (tri-n-butyl phosphate), TCEP (tris (2-chloroethyl) phosphate), TCIPP (tris (2-chloroisopropyl) phosphate), TDCIPP (tris (1,3-dichloro-2-propyl) phosphate), TBOEP (tris (2–butoxyethyl) phosphate), TPHP (triphenyl phosphate), TMPP (tris(4-methylphenyl) phosphate) and EHDPP (2-ethylhexyl diphenyl phosphate). These are selected on the basis that they are widely used and the most commonly studied FRs and OPEs with respect to their presence in indoor dust.

The data used for this meta-analysis is based on papers published in peer-reviewed journals between 1st January 2000 and 1st April 2020, and one PhD thesis (Ortiz Carrizales, 2018). The PhD thesis was included as it contains otherwise unpublished material. All datasets were identified through targeted search terms via the most popular search engines such as Science Direct, Scopus, and Google Scholar, using the terms ‘‘BFRs’’, “PFRs”, “OPEs”, “OPFRs”, “NBFRs”, “PBDEs”, “HBCDD”, “HBCD”, “indoor dust”, “human exposure”, “elevated surface”, “multi surface”, “floor”, “particle size”, “sampling method”, “sampling approach”, “spatial”, living room”, “bedroom”, “kitchen”, “temporal”, “seasonal” and “variability”. These terms were classified into four groups depending on the investigated factor, which include: spatial variability, particle size distribution, sampling method, and temporal variability of FR concentrations in indoor dust. Figure S1 shows study selection flow diagram. All data included in our evaluation are reported in the original publication. Comparison across some studies

required statistical analysis of the raw data which were requested from the authors or obtained from the Supporting Information data. Inspection of the raw data revealed it to be log-normally skewed, therefore all data were log-transformed prior to comparison of means and correlations. Student t-tests and ANOVA were applied for statistical comparison of means among different datasets and to establish the statistical significance of any observed differences. In addition, Pearson correlation was applied to test the relationship between 2 datasets, while maximum:minimum ratios and relative standard deviation (RSD) values were used to evaluate deviations from central tendencies and skewness of data. In a few studies where raw data were not available, geometric means and the outcome of a Wilcoxon rank test were taken directly from the original papers. Data were excluded from the evaluation when detection frequency of the target FR(s) was < 50%, to avoid bias caused by the method of substitution for non-detects in the respective sample group. Table S1 lists the 45 original studies included in this meta-analysis.

3. Results and discussion

3.1 Within-room spatial variability in BFR and PFR concentrations in indoor dust

3.1.1 Within-room spatial variability between floor and elevated surface dust

Several studies around the world that reported FR concentrations in elevated surface dust (ESD) and floor dust (FD), observed significant differences between the two types of dust. In the following sections, we compare the concentrations of FRs in ESD and FD from the same room taken at the same time, and discuss the causes of any such vertical spatial variability and its impact on assessments of exposure via indoor dust ingestion.

3.1.1.1 Comparison of FR concentrations in ESD and FD

For BFRs, out of six studies in Norway (Cequier et al., 2014), Iraq (Al-Omran and Harrad, 2016a), USA (Allgood et al., 2017), UK (Al-Omran and Harrad 2018), Egypt (Khairy and Lohmann, 2018), and China (Bu et al., 2019); median concentrations of most PBDEs and NBFRs in ESD from the USA, UK, Iraq, and Egypt exceeded those in floor dust samples taken in the same rooms by factors ranging between 1.1 and 19. A deviation from this overall trend was the observation that median ESD:FD ratios in dust samples from Norwegian households were <1 (0.5-0.7) for lower brominated PBDEs (tri-hexa-BDEs), although those for the other target PBDEs and NBFRs ranged between 1.1- 3.7 (Cequier et al., 2014). This may be due to the different particle size of the two dust sample categories collected in this Norwegian study. Specifically, FD was collected using a nozzle with a polyethylene grid (1−3 mm pore size) while ESD was collected without the grid. This suggests that the floor dust collected may have

contained a greater proportion of fine particles preferentially contaminated with lower brominated PBDEs. Similarly, in day care centres in China, median concentrations in ESD exceeded those in FD for only 3 out of 7 BFRs (Bu et al., 2019). For OPEs, median ESD:FD ratios for TPHP, TCEP, TCIPP, and TDCIPP from Japan (Araki et al., 2014; Tajima et al., 2014), USA (Allgood et al., 2017), UK (Ortiz Carrizales, 2018), Norway (Cequier et al., 2014; Xu et a., 2016), Egypt (Khairy and Lohmann, 2019), and China (Wu et al., 2016) ranged between 1.1- 3.9. Notable exceptions were TDCIPP and TPHP in China (Wu et al., 2016), and TPHP in Egypt (Khairy and Lohmann, 2019), for which median ratios were <1. The reason for this finding may be due to the small sample size (n=9) and low concentrations of OPEs in the day care and apartment dust studied in China and Egypt respectively. In contrast to TPHP and chlorinated OPEs, concentrations of TBOEP in FD exceeded those in ESD with median ESD:FD ratios <1 (Cequier et al., 2014; Xu et a., 2016; Araki et al., 2014; Tajima et al., 2014; Khairy and Lohmann, 2019). This may be due to the wide use of TBOEP in flooring and floor polishing materials (Kajiwara et al., 2011; van der Veen and de Boer, 2012). Table 1 shows the median ratios of the concentrations of FRs detected in ESD and FD collected from the same rooms for various studies. Figure 1 shows the median concentrations of FRs present in ESD and FD samples collected in the same studies.

Statistically significant differences were observed in concentrations of FRs between ESD and FD. For BFRs, a paired t-test showed that concentrations of BDE-209 in ESD exceeded significantly those in FD in all studies reviewed, with *p* values of 0.004, 0.002, 0.038, 0.007, and 0.017 in Norway (calculated from the raw data of Cequier et al., 2014), Iraq (Al-Omran and Harrad 2016a), USA (calculated from the raw data of Allgood et al., 2017), UK (calculated from the raw data of Al-Omran and Harrad, 2018), and China (Bu et al., 2019, using Wilcoxon rank test) respectively. In addition, concentrations of BDE-99 and BEH-TEBP in ESD exceeded significantly ($p < 0.05$) those in corresponding FD samples from Iraq (Al-Omran and Harrad, 2016a), UK (calculated from the raw data of Al-Omran and Harrad, 2018), and USA (collected from the raw data of Allgood et al., 2017). For OPEs, concentrations of chlorinated OPEs and TPHP in ESD exceeded significantly ($p < 0.05$) those in FD in 3 out of 5 studies. In contrast, TBOEP concentrations were significantly higher in FD than ESD in Norway (calculated from the raw data of Cequier et al., 2014) and Japan (Araki et al., 2014, using Wilcoxon rank test). Tables S2 and S3 show the outcomes of paired t-test comparisons of concentrations of BFRs and OPEs in ESD and FD respectively.

3.1.1.2 Relationship between concentrations of FRs in ESD and FD

Pearson correlation analysis revealed the concentrations of several FRs in ESD samples were significantly ($p < 0.05$) correlated with those in FD samples from the same room. For BFRs, significant positive linear correlation was found for BEH-TEBP and EH-TBB in Norway (calculated from the raw data of Cequier et al., 2014), Iraq (Al-Omran and Harrad, 2016a), USA (calculated from the raw data of Allgood et al., 2017) and UK (Al-Omran and Harrad, 2018) with respective correlation coefficient values of 0.673, 0.803, 0.875, and 0.952 for BEH-TEBP, and 0.686, 0.656, 0.827, and 0.639 for EH-TBB respectively. Concentrations of BDE-47 (in three studies), BDE-99, BDE-209 and DBDPE (in two studies) in ESD were significantly correlated with those in FD. In addition, chlorinated OPEs showed significant positive linear correlation between ESD and FD for TCEP in USA (calculated from the raw data of Allgood et al., 2017), UK (calculated from the raw data of Ortiz Carrizales, 2018), and China (Wu et al., 2016), as well as for TCIPP in Japan (Tajima et al., 2014) and China (Wu et al., 2016). This suggests that the sources of these contaminants in ESD and FD are similar. Tables S4 and S5 show the results of Pearson correlation analyses of concentrations of BFRs and OPEs in paired samples of ESD and FD reported in various studies.

3.1.1.3 Causes of variability in FR concentrations between ESD and FD

As SVOCs and additive chemicals, FRs can be released from the treated products via volatilisation and subsequent sorption to dust particles governed by the contaminant's octanol-air partition coefficient (K_{OA}) (Weschler and Nazaroff, 2010, Fromme, 2012). This combination of volatilisation and sorption seems more plausible for compounds with higher vapour pressures, such as tri-hexa-PBDEs (Weschler and Nazaroff, 2008; Al-Omran and Harrad 2016a), and TCEP (Xu et al., 2016; Wu et al., 2016). However, for lower vapour pressure compounds such as decabromodiphenyl ether (BDE-209), this explanation likely accounts for a lower proportion of the concentrations of such chemicals detected in settled dust. For such less volatile FRs, other mechanisms of source-to-dust transfer have been shown to make a greater contribution. These include: abrasion of particles/fibres of FR-treated materials and transfer via direct source:dust contact (Rauert and Harrad, 2015; Rauert et al., 2016). Table S6 shows physicochemical properties of our target FRs. In addition to differences between ESD and FD in dust particle size distribution, differences in the number and type of FR sources present on floors and elevated surfaces, are likely important factors influencing differences in concentrations of FRs between ESD and FD from the same microenvironment.

Building materials are an important source of FRs, especially floor materials (Kanazawa et al., 2010). Thus, the wide use of TBOEP as a plasticiser in floor wax and PVC floor coverings (Kajiwara et al., 2011; van der Veen and de Boer, 2012) likely explains the higher concentrations observed in floor as opposed to elevated surface dust. In contrast, wooden-structure houses displayed lower levels of TCIPP, TCEP, and TPHP in FD than other types of houses, such as those with reinforced concrete floors (Araki et al., 2014; Tajima et al., 2014). TPHP is commonly used in combination with halogenated OPEs added to polyurethane foam as well as sealing and heat-insulating materials (van der Veen and de Boer, 2012; Tajima et al., 2014). As shown in Figure 1, Table 1 and Table S3, the distribution profile of TPHP and chlorinated OPEs between ESD and FD are similar. Specifically, except for TCEP in Norway (for which the median ESD:FD ratio = 1); median ESD:FD ratios of TPHP (1.7, 2.5 and 1.2) were consistent (>1) with those of TCEP (1.0, 1.4 and 2.4), TCIPP (2.6, 3.0 and 1.5) and TDCIPP (2.8, 3.9 and 1.3) from the same dust sample in Norway, Japan and UK respectively.

Contaminated fine particles originating from the floor materials may be suspended into indoor air before settling on elevated surfaces (Wu et al., 2016). This may explain the presence of lower vapour pressure compounds in ESD even where floor materials are the source. Also, as finer particles are more atmospherically mobile, ESD likely contains a greater proportion of fine particles containing chemicals that have accumulated via volatilisation and subsequent deposition (Rauert and Harrad, 2015). Moreover, where ESD samples are collected from surfaces that include putative sources (e.g. TVs and other electronics), a proportion of such dust will have been in direct contact with these sources. Combined with the fact that elevated surfaces may be less frequently cleaned than floors, this provides another explanation for FR concentrations in ESD that exceed those in FD (Tajima et al, 2014; Rauert et al., 2014b). Furthermore, outdoor particles such as sand and soil tracked indoors with footwear are more likely to dilute the concentration of pollutants in FD (Cao et al., 2014a). Conversely, the lowest ESD:FD ratios of BFRs (Table 1) reported in dust samples taken from Iraq (Al-Omran and Harrad (2016a) is likely due to the deposition of ultrafine Sahara dust particles from outdoors, which may preferentially dilute ESD. Al-Omran and Harrad (2016a) examined the hypotheses that differences in concentrations of organic carbon (OC) and particle size distribution between ESD and FD contribute to observations of significant differences in concentrations of FRs between such dust types. The authors reported significantly ($p<0.05$) higher proportions of fine particles (<125 μm) in ESD samples and no substantial influence of OC on the observed

differences in concentrations between ESD and FD. To our knowledge, this is the only study addressing this topic; therefore, further research is needed to support these findings.

3.1.1.4 The influence of sampling surface on human exposure assessment

As discussed above, concentrations of several FRs in ESD samples collected from tables, shelves, chairs, etc. exceeded significantly those in FD. However, the majority of assessments of human exposure to FRs via indoor dust ingestion have to date focused on collection and analysis of floor dust samples. This is likely appropriate for young children and toddlers as they are far more likely to have contact with FD when playing close to the floor; however, adults, adolescents, and older children are more likely to come into contact with ESD. Coupled with observations of higher FR concentrations in ESD, this indicates that human exposure assessments based on FD alone will likely underestimate exposure via dust ingestion for adults. As shown in Tables 1, S2 and S3, and Figure 1, the extent of underestimation is greatest for BDE-99, BEH-TEBP, BDE-209, chlorinated OPEs, and TPHP. For example, such underestimates for the adult population can be substantial e.g., reaching as high as factors of 5.0 and 19 for "typical" exposure rate of BDE-183 in USA (Allgood et al., 2017) and Egypt (Khairy and Lohmann, 2018) respectively.

3.1.2 Within-room spatial variability in floor dust from different areas of the same microenvironment

Several studies report substantial within-room spatial variability in concentrations of FRs in floor dust taken at the same time from different areas of the same room. For BFRs, from five different microenvironments (3 homes and 2 offices), 5 floor dust samples were collected on the same day from different locations in each microenvironment (Harrad et al., 2008). This appears to be the first study of within-room spatial variability in contamination of dust with PBDEs, followed by a similar study of HBCDDs in six microenvironments in the UK (Harrad et al., 2009) and of PBDEs and NBFRs in one microenvironment in the Czech Republic (Melymuk et al., 2016). To differentiate spatial variability from analytical variability, the high RSD values of PBDEs (30-183%) and HBCDDs (36-93%) were compared with those calculated from replicate analyses of a dust standard reference material (SRM2585 and 2584), which were 7.8-67% for individual PBDEs and <22% for HBCDD (Harrad et al., 2008; 2009). High RSD values (60-116%) were also found for individual PBDEs and NBFRs in 4 dust samples (2 from exposed floor areas and 2 from hidden areas) collected from a bedroom in the Czech Republic (calculated from the raw data of Melymuk et al., 2016).

Other studies in the UK tested within-room spatial variability in concentrations of BFRs (Muenhor and Harrad, 2012; Al-Omran and Harrad 2018) and OPEs (Ortiz Carrizales, 2018) in floor dust samples (2-5) taken from different 1 m^2 areas within the same rooms (n=5-9). Significant differences ($p < 0.05$) were observed in concentrations of $\Sigma_7$tri-hepta-BDEs and ΣNBFRs in different areas of the same room for three out of nine rooms studied, but not for BDE-209 (calculated from the raw data of Al-Omran and Harrad 2018). For OPEs, significant differences ($p < 0.05$) were also found in two out of four rooms for TPHP and EHDPP and in one room for both TNBP and TDCIPP. No significant differences were observed for TCIPP and TCEP (Ortiz Carrizales, 2018). Concentrations of TCIPP and TCEP were strongly associated with floor materials such as tatami, tiles, and wall-to-wall carpet (Tajima et al., 2014). However, TCIPP displayed strong spatial variability in dust samples from the Czech Republic. Concentrations of TCIPP decreased with increasing distance from a sofa, implying the use of TCIPP in the foam filling of the sofa concerned (Jilkova et al., 2018).

Overall, the evidence suggests higher concentrations of FRs accumulate in dust samples located closest to potential emission sources, such as sofas, chairs, TVs, and laptops etc. Harrad et al., (2009) reported that concentrations of HBCDDs declined with increasing distance from a TV. In other words, within-room spatial variability is likely driven by varying distances from potential emission sources. Greater spatial variability was observed in large rooms and between bare floor and carpeted floor areas within the same room (Al-Omran and Harrad, 2018). In addition, in one office (1450 m^2) from China, higher concentrations of PBDEs were detected in some areas, compared to those in areas close to the entrance, probably due to the poor ventilation away from the entrance (Li et al., 2015). Table S7 reports the RSD values for BFRs in different microenvironments, while Figure S2 compares median concentrations of selected BFRs and OPEs in floor dust from two different areas of the same room located in the UK.

These results indicate that appreciable variation in FR contamination can be found, depending on where in a given room, floor dust samples are taken. This suggests that for accurate estimation of exposure, such "spot" samples may not be fully representative of the contamination of the room as a whole. For example, if in a given room (assuming the two floor areas are F1 and F2), the concentration of $\sum_7$tri-hepta-BDEs in F1 exceeds substantially that in dust samples from F2 with ratio of F1:F2=61:4 for one sampling event; in this room, the exposure estimate will vary by a factor of just over 15 depending on the area samples (Al-Omran and Harrad, 2018). Overall, while the substantial and sometimes significant variation in FR concentrations in floor dust from different areas of the same room, may imply that

sampling of the entire floor area is the best approach to estimating human exposure; the fact that doing so includes dust from rarely-frequented parts of the room, led Al-Omran and Harrad (2018) to conclude that floor dust samples should be taken from the most-frequented parts of the room in order to best reflect human exposure.

3.1.3 Within-building spatial variability

Within building spatial variability refers to whether concentrations of contaminants in dust vary between different rooms in the same building. Several studies have focused on within-home spatial variability between the living room and bedroom (Allen et al., 2008; Stapleton et al., 2008; Muenhor and Harrad 2012; Venier et al., 2016; Al-Omran and Harrad, 2017; 2018; Ortiz Carrizales, 2018). To our knowledge, only two studies have investigated within-home spatial variability between the kitchen and living area/bedroom (Kuang et al., 2016; Ortiz Carrizales, 2018), with two other studies reporting spatial variability between different rooms in academic microenvironments (Dodson et al., 2017; Jílkova et al. 2018).

In two studies from the USA (Allen et al., 2008; Stapleton et al., 2008), concentrations of Penta-BDE, Deca-BDE, HBCDD, EH-TBB, and BEH-TEBP were significantly (p <0.05) higher in the main living area compared to the bedroom. No significant differences were apparent for BTBPE and DBDPE (Stapleton et al., 2008). In UK house dust samples, median concentrations of individual PBDEs and NBFRs in the living room were noticeably higher than those in the corresponding bedrooms (statistically significant for BDE-183) (Al-Omran and Harrad, 2017). Kuang et al., (2016) also reported no significant differences (p >0.05) between BFR concentrations in the living room and bedroom in UK dust samples. In contrast, another study investigating PBDE and NBFR concentrations in living rooms and bedrooms in the same homes in three countries (USA, Canada, and Czech Republic) reported different patterns (Venier et al., 2016). Except for EH-TBB in the USA and BEH-TEBP in Canada, median concentrations of PBDEs and NBFRs in the bedrooms exceeded those in corresponding living rooms and were significantly (p <0.05) higher for BDE-153, BTBPE, and BDE-209 in USA, Canada, and the Czech Republic respectively (calculated from the raw data of Venier et al., 2016). In Australia, except for BDE-154 (for which concentrations were significantly higher in the living room), no significant differences were found between concentrations of PBDEs and NBFRs in living rooms and bedrooms from the same homes (calculated from the raw data of McGrath et al., 2018). Figure 2 illustrates the distribution profiles of the most common PBDEs and NBFRs in living rooms (LR) and bedrooms (BR) from Canada, Australia, USA,

411 and the UK. According to this figure, not only the concentrations, but also differences in the
412 profile of BFRs are apparent between living room and bedroom dust samples.

413 In addition to the above studies, three further UK studies (Muenhor and Harrad 2012; Al-
414 Omran and Harrad 2018; Ortiz Carrizales, 2018) investigated within-home spatial variability
415 in concentrations of PBDEs, NBFRs, and OPEs in different rooms from a limited number (2-
416 3) of homes over 8-12 months. No specific trend was evident for BFRs, with any between-
417 room variation dependent on differences in room contents with respect to putative sources. For
418 instance, while in one study, concentrations of BDE-47 and BDE-99 in the living room were
419 significantly higher ($p<0.05$) than those in the bedroom (Muenhor and Harrad, 2012), in
420 another they were significantly higher in the bedroom than the living room (Al-Omran and
421 Harrad, 2018). For OPEs, no significant differences were found between living rooms and
422 bedrooms (Ortiz Carrizales, 2018).

423 A comparative study of dust from UK kitchens and living rooms, revealed that concentrations
424 of individual PBDEs, NBFRs, and HBCDD in living rooms/bedrooms exceeded significantly
425 ($p \leq 0.05$) those in the corresponding kitchens; with the exception of DBDPE, for which no
426 significant differences were evident (Kuang et al., 2016). In a similar study conducted in the
427 UK for OPEs (Ortiz Carrizales, 2018), concentrations in kitchen floor dust were markedly
428 lower than those observed in the living room/bedroom. In 12 consecutive monthly dust samples
429 collected from one UK home, concentrations of TNBP and TDCIPP in the bedroom were
430 significantly higher than those in the corresponding kitchen with p values of 0.028 and 0.004,
431 respectively. Likewise, concentrations of TCIPP, EHDPP, and TPHP in the living room
432 exceeded significantly those in the kitchen with p values of 0.036, 0.001, and 0.003,
433 respectively (Ortiz Carrizales, 2018).

434 Within-building spatial variability was also studied in an academic microenvironment in the
435 Czech Republic between two rooms (CR-computer room and SR-seminar room) located one
436 floor apart with different furnishings and equipment but otherwise of identical size, shape,
437 location, and building material (Jílkova et al., 2018). While the average concentrations of
438 PBDEs, DBDPE, and EHDPP were comparable in both rooms, those of EH-TBB, BTBPE,
439 BEH-TEBP, TCIPP, and TDCIPP were higher in CR and significantly (p=0.044) higher for
440 EH-TBB. On the other hand, average concentrations of TNBP and TPHP were higher in SR
441 and significantly (p= 0.012) higher for TNBP (calculated from the raw data of Jílkova et al.,
442 2018). From another academic microenvironment (2 college campuses in the USA), Dodson et

al., (2017) found concentrations of TCEP and TPHP in student dormitory rooms to exceed significantly (p <0.05) those in dust from common areas, while concentrations of BDE-209 in common areas were significantly higher than in dormitory rooms (Dodson et al., (2017).

The above reveals meaningful differences in FR concentrations in dust between different microenvironments in the same building. This is considered a reflection of variability between rooms in the numbers and types of potential FR emission sources present. For example, higher concentrations of PBDE congeners representative of the Penta- and Deca-BDE formulations in living rooms compared to bedrooms may be attributed to the presence in the former, of specific putative source items such as televisions and couches (Allen et al., 2008). Moreover, higher concentrations of TCEP and TPHP in student dormitory rooms, compared to common areas, may be related to the electronics and furnishings introduced to dormitories by students (Dodson et al., 2017). Similar explanations were proposed by other studies (Stapleton et al., 2008; Muenhor and Harrad, 2012; Al-Omran and Harrad, 2017; 2018; Jílkova et al., 2018; McGrath et al., 2018), which is supported by the lack of strong correlations between FR concentrations in rooms within the same building, as indicated by Table S8 that shows Pearson's correlation coefficient (r) values between different rooms within the same building in various studies. Alternatively, the observed lack of significant differences in concentrations of FRs between bedrooms and living rooms in the same home suggests that emission sources are similar in both rooms (Kuang et al., (2016)). However, a similar lack of significant differences between rooms in the same house was attributed by Venier et al., (2016) to air exchange and concomitant transfer of FRs between rooms. This latter explanation likely applies when the main mechanism via which FRs enter dust in the building concerned, is volatilisation from sources with subsequent deposition to dust; but is less plausible when other source-to-dust transfer mechanisms dominate, such as source abrasion and direct source-dust contact.

Between-room differences in source types and numbers that lead to substantial within-building spatial variability means that exposure estimates based on measuring BFRs in dust from one room may not be entirely representative of exposure via dust ingestion to *all* individuals frequenting a given building. For example, in one home, concentrations of BEH-TEBP in the bedroom (average ± SD = 3992 ± 1906 ng/g) exceeded those in the living room (average ± SD = 1811 ± 1498 ng/g) (Al-Omran and Harrad 2018). This implies that exposure assessment in that home based on living room dust only may result in a substantial underestimate.

3.2 The influence of dust particle size fraction

Dust from indoor microenvironments includes a wide range of particle sizes ranging from < 2.5 μm to over 2000 μm in diameter. Particles > 30 μm may be classified as settled particles, which include "skin flakes, fragments of hair, microorganisms, such as fungal spores and pollen, food crumbs, abrasion of textiles and fittings, sand, loam, clay, and soot" (Morawska, 2004). Studies investigating human exposure to FRs via indoor dust ingestion have analysed a wide range of dust particle sizes, such as <2000 μm (Gevao et al., 2006), <500 μm (Abdallah and Covaci, 2014), <250 μm (Al-Omran and Harrad 2017), <150 μm (Shoeib et al., 2012), <100 μm (Kang et al., 2011), and < 63 μm (Kopp et al., 2012). However, for assessment of exposure via dust ingestion, it is reasonable to assume that only those particles adhering to human hands and that can subsequently be orally ingested via hand-to-mouth contact and/or release FRs via dermal uptake are pertinent. The most recent recommendation by the USEPA is that adherence to hands is greatest for dust particles <150 μm diameter (USEPA, 2016), which is smaller than their previous recommendation to focus on particles <250 μm diameter (USEPA, 2008). Moreover, increased FR bioaccessibility via dust ingestion was observed with decreasing particle size (Yu et al., 2013; Fang and Stapleton, 2014). This section highlights the extent to which analysing different dust fractions influences FR concentrations and consequent exposure assessments.

While in some studies, FR concentrations increased with decreasing dust particle size, other studies revealed that specific pollutants accumulated in specific particle size fractions and varied between different microenvironments. The first investigation of FRs in different particle size fractions of indoor dust was reported by Wei et al., (2009) in the USA, concluding that while in car dust, PBDE concentrations were inversely related to particle size, they were largely comparable in four dust fractions (250- 420 μm, 150-250 μm, 75-150 μm, and < 75 μm) for home dust. A comparison of dust from different microenvironments (offices, hotels, kindergartens, and dormitories), showed that concentrations of DBDPE and HBCDD in hotel dust were highest in fine particles (50-74 μm and >50 μm respectively), while in office dust concentrations were highest in medium particles (100–200 μm) (Cao et al., 2014a; 2015). These findings are consistent with other reported data from three different microenvironments in China. Among four particle size fractions, BDE-209 and BEH-TEBP accumulated most strongly in both fine (43-63 μm) and finest (<43 μm) particles of office dust, while they were present at higher concentrations in coarse particles (150-200 μm) in public microenvironments (PMEs) (He et al., 2018). In addition, differences in distribution pattern of FRs between different dust particle sizes were found between different countries from the same type of

510 microenvironment. While the concentration of $\sum_7$OPEs was higher in particles 25-75 μm in
511 homes from China, concentrations were higher in the finest particles (<25 μm) in Swedish
512 homes (Li et al., 2019). It can be concluded that different microenvironments can show
513 different distribution patterns of FRs in various particle size fractions with no specific trend of
514 a given pollutant to accumulate in a specific size fraction. Figure S3 demonstrates the different
515 distribution patterns of OPEs in different particle size fractions (<25- <500 μm) from different
516 microenvironments and different countries.
517
518 In other studies, the distribution pattern of FRs in different dust particle size fractions revealed
519 generally that concentrations of BFRs tend to increase with decreasing particle size, with some
520 significant differences. In three particle size fractions (106-150 μm, 45-106 μm, and <45 μm)
521 of dust samples from 2 homes and 2 offices, concentrations of BDE-99 and BDE-153 increased
522 with decreasing particle size, while other PBDE congeners showed higher concentrations in
523 the fine (<45 μm) and medium particle size ranges (45-106 μm) (Kefeni and Okonkwo, 2014).
524 Elsewhere, in three size fractions of 10 dust samples from 5 homes in the UK, while
525 concentrations of ∑tri-hepta-BDEs and BEH-TEBP in the finest particles (<63 μm) exceeded
526 significantly those in medium (63 - <125 μm) and coarse particles (<125-250 μm); median
527 concentrations of BDE-209 in these three particle size fractions were comparable (Al-Omran
528 and Harrad 2016b). The same trend was found for PBDEs in four size-fractionated dust samples
529 from 5 homes in Japan (Kajiwara and Takigami, 2016). Concentrations of PBDEs in dust
530 tended to increase with decreasing particle size, but concentrations in the finest particles (<50
531 μm) were not the highest. One study in 8 homes in Taiwan investigating PBDEs in three
532 particle size fractions (>149 μm, 75-149 μm, and <75 μm) proposed that PBDE concentrations
533 may not be correlated with the particle size (Chao et al., 2014). For OPEs, in dust from 7
534 building material markets from Germany, most OPEs targeted showed no consistent trend with
535 particle size (Zhou and Puttmann, 2019). The exception to this was TCIPP, which was enriched
536 in medium size (< 63-150 μm) particles. From the above studies, it can be concluded that due
537 to the greater surface area to volume ratio of fine particles, the impact of atmospheric deposition
538 of FRs will be greater on fine indoor dust particles.
539 Contaminants with relatively high vapour pressures (VP) are expected to migrate from sources
540 to the environment via evaporation and subsequent deposition to particles, depending on the
541 octanol-air partition coefficient (K_{OA}) of the contaminant and the organic carbon content of
542 indoor dust particles (Webster et al., 2009; Weschler and Nazaroff, 2010; Yu et al., 2013).
543 However, this does not seem to constitute a general rule. For instance, in 5 fractions (<43-200

μm) of dust samples from three different microenvironments, the highest concentrations of TCEP (VP=$5.2x10^{-2}$ Pa) were found in the finest (<43 μm) particles with maximum:minimum ratios of 2.2, 1.4 and 1.3 in office, public microenvironment and car dust samples respectively. However, concentrations of TCIPP (VP=$7.4x10^{-3}$ Pa), close to that of TCEP) were higher in the coarse fraction (100-200 μm). In contrast, the concentrations of DBDPE and BDE-209 (VP=$2.6x10^{-11}$ and $9.3x10^{-9}$ Pa respectively), were higher in the finest particles of the same samples (He et al., 2018; Cao et al., 2013). These findings are consistent with other studies, e.g. while concentrations of the more volatile tri-hepta BDEs were higher in medium sized (125–212 μm) particles in a pooled sample of dust from air conditioner filters in dining halls, the concentration of BDE-209 was higher in the fine particles (<43-63 μm) (Yu et al., 2013). The above examples suggest that while in many scenarios, the predominant source-to-dust transfer mechanism is volatilisation followed by deposition to finer particles with greater specific surface area; in some situations, FRs in indoor dust particles might arise predominantly from a source weathering or abrasion process rather than volatilisation/deposition. Thus, in such situations, irrespective of the vapour pressure, the size of the abraded, FR-rich particles is likely to determine the fraction with the highest FR concentration. Overall, regardless of the predominant source-to-dust transfer mechanism, our meta-analysis suggests that significant differences do exist between concentrations of FRs in different particle size fractions. This supports focusing on analysis of those particle sizes shown to adhere to hands as the most relevant for human exposure via unintentional dust ingestion. Current USEPA recommendations are that these are particles <150 μm diameter (USEPA, 2016).

3.3 Evaluation of dust sampling approaches

Dust sampling approaches can be divided into passive and active techniques. The passive technique includes collecting dust via natural accumulation of suspended particles onto collection receptacles for a given period (Butte and Heinzow, 2002; Niu et al., 2018; 2019). This method is rarely used due to the time required to collect sufficient dust for analysis (Mercier et al., 2011). Active techniques involve several approaches including wiping (Abbasi et al., 2016; Xu et al; 2019), sweeping (Qi et al., 2014; Zeng et al., 2018; Iwegbue et al., 2019), and vacuuming (Fromme et al., 2014; Tao et al., 2016). The wiping method was originally used to collect settled dust by rubbing hard surfaces with alcohol moistened paper towels (USEPA, 1995). Although this method is not recommended for collection of surface dust due to poor collection efficiency of fine particles (USEPA, 2008), it is widely applied for collection of

hand-adhered dust to e.g. test associations between indoor dust and pollutant body burdens and to evaluate the significance of dermal exposure (Hoffman et al., 2015; Cowell et al., 2017; Larsson et al., 2018).

The most widely used sampling technique is vacuuming, most commonly with conventional household vacuum cleaners as inexpensive alternatives to the standard HVS3 (High Volume Small Surface Sampler) developed by USEPA (Colt et al., 2008). Vacuum cleaner methods may be split into two categories; (a) researcher collected dust (RCD), where dust is collected by researchers using specific accessories and standardised methods; and (b) householder vacuum dust (HHVD), where householders provide researchers with their vacuum cleaner contents (Shen et al., 2015; Ali et al., 2016). The most common accessories that have been used for RCD are nylon "socks" (Harrad et al., 2016), filters (Newton et al., 2015), and Soxhlet thimbles (Stapleton et al., 2012), all deployed to collect dust before it enters the vacuum cleaner dust bag/receptacle. However, for assessment of human exposure to indoor pollutants via dust ingestion, all of the above approaches have associated uncertainties. This section discusses to what extent the sampling method used influences FR concentrations in the dust collected and the consequent exposure assessment.

To date, a small number of studies have compared concentrations of FRs in RCD and HHVD. Two early studies in the USA found that concentrations of Penta-BDE and Deca-BDE congeners in RCD (obtained using cellulose extraction thimbles inserted in the vacuum cleaner sampling hose) collected from living rooms and those of Penta-BDE congeners in bedrooms exceeded significantly ($p<0.05$) those in HHVD. In contrast, geometric mean concentrations in HHVD were 2 times higher for HBCDD and comparable for BDE-209 compared to RCD collected in bedrooms. In general, concentrations of PBDEs were weakly to moderately correlated between dust collected via the two sampling methods (Allen et al., 2008; Stapleton et al., 2008). Another study in Sweden examined BFRs in RCD (collected using cellulose filters inserted in the vacuum cleaner hose) and HHVD samples, observing that the median concentrations of PBDEs in RCD were 2-3 times higher than those in HHVD and significantly ($P<0.05$) higher for ∑PentaBDE, ∑OctaBDE, and ∑DecaBDE. In contrast, median concentrations of HBCDD were 10 times higher in HHVD than RCD. Statistically significant correlations were found for Octa- and Deca-BDE between the two sampling methods, while no correlation was found for ∑PentaBDE (Björklund et al., 2012). In this study, RCD samples were collected from elevated surfaces only. Assuming that the HHVD samples were collected

611 from the floor, then the elevated levels of BFRs in RCD may be attributed to the different
612 sampling surfaces. In the UK, concentrations of ∑tri-hexa-BDEs and to a moderate extent those
613 of BEH-TEBP in RCD (obtained using nylon socks) exceeded significantly those in HHVD
614 with *p* values of 0.012 and 0.077 respectively. Moreover, in living rooms, median
615 concentrations of BDE-209 and EH-TBB in RCD exceeded those in HHVD, while in contrast,
616 DBDPE concentrations were comparable in both RCD and HHVD samples (Al-Omran and
617 Harrad, 2017). This similarity between RCD and HHVD for DBDPE differs from the
618 observation of 3.5-3.9 times higher concentrations in RCD in the USA (Stapleton et al., 2008).
619 Concentrations of ∑tri-hexa-BDEs, BDE-209, and BEH-TEBP were moderately correlated
620 between dust collected via the two sampling approaches (Al-Omran and Harrad, 2017). In
621 contrast with these studies, in South Africa (Brits et al., 2019), median concentrations of lower
622 brominated PBDEs (BDE-47, -99, -153, and -154) were comparable in dust collected via both
623 sampling methods, while for BDE-209 the concentration in RCD (obtained via a cone-shaped
624 folded filter paper placed in the vacuum cleaner sampling hose) exceeded that in HHVD by a
625 factor of 3.2. In the same study, the RCD sample analysed included both ESD and FD and the
626 analysed particles were <150 μm, which may contribute to the observed high levels of BDE-
627 209. For OPEs, the same study found that median concentrations of TBOEP and TMPP were
628 comparable in both RCD and HHVD samples, while TNBP was higher in HHVD. Another
629 study from Canada (Fan et al., 2014) investigated the two sampling techniques and found that
630 with the exception of EHDPP, concentrations of chlorinated OPEs, TPHP and TMPP were
631 higher in RCD. Except for EHDPP (moderate correlation), strong correlations were found
632 between the two sampling approaches. Figure 3 compares concentrations of ∑tetra-hepta-
633 BDEs, BDE-209, and BEH-TEBP in RCD and HHVD from different countries. According to
634 this, it can be concluded that concentrations of lower brominated compounds, BEH-TEBP,
635 TPHP and chlorinated OPEs are substantially higher in RCD, while HBCDD concentrations
636 are higher in HHVD. Several reasons may contribute to this trend for some FRs in RCD
637 compared with HHVD. RCD samples were collected from a specific room – e.g. bedroom or
638 living room – containing several sources of FRs - whereas the contents of the participant's
639 home vacuum cleaner bag likely represents dust sampled to varying degrees from all rooms
640 including kitchen, bathroom, hallways and sometimes outside of the home, all of which likely
641 contain fewer FR treated products than the bedroom or living room. In addition, there is more
642 opportunity for losses of volatile FRs from HHVD over the extended period such dust spends
643 in the dust bag. Furthermore, fine particles that contain higher concentrations of some FRs may

have been lost from the vacuum cleaner bags during the process of sample collection and transfer to the lab.

To evaluate the extent to which exposure to FRs via dust ingestion is influenced by the type of dust sampling method, we compared the median concentrations of various FRs in dust collected via the two sampling approaches. Table S9 lists ratios of median concentrations of BFRs between RCD and HHVD samples collected from the same homes. According to these results, assessments based on FR concentrations detected in HHVD rather than RCD will underestimate exposure for lower brominated BFRs, BEH-TEBP, TPHP, and chlorinated OPEs by factors ranging between 1.1 and 5.5. This indicates that the HHVD method is a less acceptable method of assessing exposure to such pollutants.

3.4 Temporal and seasonal variability

Several studies have investigated temporal variability in concentrations of FRs in indoor dust, including PBDEs (Allen et al., 2008; Harrad et al., 2008; Batterman et al. 2009; Yu et al., 2012; Muenhor and Harrad 2012; Al-Omran and Harrad, 2018; Niu et al., 2018), NBFRs (Cao et al., 2014b, Al-Omran and Harrad 2018, Niu et al., 2019), HBCDD (Harrad et al., 2009; Cao et al., 2015), and OPEs (Cao et al., 2014b; Ortiz Carrizales, 2018). While some of these studies presented their data on a month-to-month basis, others presented it by season (spring, summer, autumn, and winter) or classified into colder and warmer periods. However, results are insufficient to elucidate seasonal trends of FRs when based on discontinuous (i.e. incomplete calendar year) campaigns, although these results can be used for attribution of emission source(s). This section highlights the extent to which assessments of human exposure to BFRs and OPEs via indoor dust ingestion may be influenced by temporal and seasonal variability. The relative standard deviation of concentrations (RSD) of individual FRs and maximum:minimum concentration ratios were applied to measure the extent of variability over the studied time period.

The first investigation of temporal variability in concentrations of PBDEs in dust samples from US homes, revealed no significant differences in samples taken from the same rooms 8 months apart, likely because home furnishings changed little over that time (Allen et al., 2008). However, for no obvious reason, only those congeners associated with Octa-BDE in this study, were significantly higher in dust samples collected in autumn compared with samples collected in winter. In contrast, another study in the USA (Batterman et al. (2009) showed little consistent

678 trend in PBDE concentrations in dust samples collected in two different seasons from homes
679 and garages, which is in line with the temporal variation observed in several UK buildings. In
680 the UK, four studies (Harrad et al., 2008; 2009; Muenhor and Harrad 2012; Al-Omran and
681 Harrad, 2018) reported substantial variability in BFR concentrations in floor dust collected
682 from several microenvironments at monthly intervals over 8-10 months. For example, RSDs
683 of concentrations of ∑tri-hexa-BDEs (52-156%), BDE-209 (58-166%), and HBCDDs (27–
684 190%) exceeded those observed for within-room spatial variability (Harrad et al., 2008a; 2009).
685 This is likely attributable to changes over time in room contents with respect to potential
686 sources of FRs. For instance, when a new rug was introduced, concentrations of BDE-209 rose
687 substantially from 3152 to 19802 ng/g (Al-Omran and Harrad, 2018).
688
689 As SVOCs, it is expected that temperature changes between the four seasons or between colder
690 and warmer months will exert an influence on the transfer of FRs from sources to indoor dust
691 via volatilisation/deposition. Yu et al., (2012) reported that average concentrations of PBDEs
692 in indoor dust from China followed the order summer> winter> spring> autumn (Yu et al.,
693 2012). This is consistent with recent studies (Niu et al., 2018; 2019) comparing concentrations
694 of BFRs in different seasons, that found concentrations of Σ_7NBFRs were significantly ($P <$
695 0.05) higher in August than in other months. The geometric mean concentrations followed the
696 order: summer> winter> autumn> spring, indicating that elevated temperatures in summer
697 increased emissions of FRs from their sources via volatilisation. However, these seasonal
698 variations conflict to some extent with the results reported by Cao et al. (2014b) for dust
699 samples collected from three offices for 9 consecutive months (for which there was no apparent
700 addition or removal of FR containing products). While ΣPBDE and ΣNBFR concentrations
701 remained relatively stable throughout, PFR concentrations changed in contrast to the trend of
702 temperature, with higher concentrations in later winter and early spring and lower levels in
703 summer. This temporal variation was attributed to the greater volatility of the OPEs studied
704 relative to the BFRs (Cao et al., 2014b). Regarding OPEs, similar observations about temporal
705 variability to those reported by Cao et al. (2014b) were reported in a study of house dust
706 samples from the UK at monthly intervals for one year (Ortiz Carrizales, 2018). The
707 concentrations of OPEs were higher in later winter and early spring, and lowest in autumn and
708 early winter. Combined, these two studies revealed concentrations of OPEs in indoor dust to
709 be sensitive to temperature, although RSD and maximum/minimum concentration ratios were
710 lower than observed in other studies for BFRs. Tables S10 and S11 illustrate temporal

variability in concentrations of FRs in indoor dust observed in different studies conducted in a range of microenvironments.

In summary, these considerable temporal and seasonal variations indicate the uncertainty associated with basing exposure assessments via dust ingestion on a single grab sample taken from a given area at a given point in time. The high RSD values of FRs in Tables S10 and S11 imply that human exposure to such chemicals in the rooms studied via contact with dust would vary to the same extent. Moreover, the high maximum:minimum ratios in Table S10 imply that exposure could be overestimated or underestimated by factors equal to these ratios, which can be substantial e.g. reaching as high as 440 for BDE-209 and 350 for EH-TBB.

4. Conclusions

Differences in FR concentrations and consequent human exposure assessments were observed due to spatial and temporal variability, the dust particle size fraction analysed, and the sampling method. Significant differences were observed in concentrations of BFRs and OPEs between floor dust and elevated surface dust. Such differences are attributable to a variety of factors, including the presence of sources on the surfaces sampled (e.g. flooring material), and the higher proportion of fine particles found on elevated surfaces. In the majority of studies, FR concentrations in elevated surface dust exceed those detected in floor dust from the same room. Thus, for most FRs, exposure assessments will be underestimated when based on floor dust only, particularly for lower brominated compounds, BEH-TEBP, BDE-209, chlorinated OPEs, and TPHP. Meanwhile, sampling dust from one specific area of the floor within a room may not reflect contamination of the entire room due to spatial variations in distance from putative sources. Within-building variability in FR concentrations between different rooms is likely due to between-room differences in putative sources, implying exposure estimates based on one room may not be entirely representative for all people living or working in that building. While differences are observed in the distribution of FRs between different dust particle size fractions; these differences did not follow a definitive trend. Specifically, while in some studies, FR concentrations increased in line with increases in particle specific surface area associated with decreasing particle size; this appears to be overridden in situations where the principal source of FRs in a dust sample is abrasion of source material. In such situations, FR concentrations are highest in the size range of the abraded particles. With respect to the sampling method deployed, concentrations of lower brominated BFRs, BEH-TEBP, TPHP, and chlorinated OPEs are generally reported to be higher in RCD than in HHVD, implying that exposure

assessments based on HHVD will be underestimated. Moreover, temporal variations in FR concentrations in dust revealed uncertainty in exposure assessments based on a single dust sample taken from a given area at a given point in time. This is likely due to month-to-month changes in room contents with respect to putative emission sources. While some seasonal variation in FR concentrations in indoor dust has been observed, it is less marked than those observed in outdoor air. This is likely attributable to the temperature difference between warmer and colder seasons being less marked indoors than outdoors, leading to a less marked increase indoors in emissions from sources due to volatilisation in warmer compared to colder months, with any such increased indoor emissions in warmer months likely further offset by increased ventilation.

In conclusion, the above factors should be taken into account when designing a study to assess human exposure to FRs and OPEs via ingestion of indoor dust. For example, the choice of whether to collect and analyse ESD or FD may be influenced by whether exposure is being estimated for young children in daycare centres or adults in offices. In the former case, FD is recommended, while ESD is advised in the latter scenario. Moreover, the observed within-room and within-building variability in FR and OPE concentrations suggests that rather than sampling all surfaces in a room, dust should be collected and analysed only from the most-frequented areas of that room. While ideally multiple rooms in a dwelling would be sampled to fully reflect exposure of its inhabitants, where resources permit sampling of only 1 room, it is recommended that this be the main living room or area as it is frequented by all inhabitants for a substantial proportion of the day. Furthermore, given the temporal variations in concentrations of FRs and OPEs in dust observed in the same rooms in some studies, exposure should ideally be estimated based on at least 2 dust samples taken several months apart. Finally, variations in FR and OPE concentrations between different indoor dust particle size fractions means that for the purposes of assessing exposure via dust ingestion, dust analysed should be confined to the size fraction that adheres to hands.

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Table 1. Median ESD:FD ratios of BFRs and OPEs in indoor dust from Norway (Cequier et al., 2014; Xu et al., 2016*), Iraq (Al-Omran and Harrad, 2016a), USA (Allgood et al., 2017), UK (Al-Omran and Harrad 2018; Ortiz Carrizales, 2018), Egypt (Khairy and Lohmann, 2018; 2019), China (Bu et al., 2019; Wu et al., 2016) and Japan (Araki et al., 2014; Tajima et al., 2014)**

BFRs						
Target BFR	**Norway**	**Iraq**	**USA**	**UK**	**Egypt**	**China**
BDE-47	0.5	**1.8**	**2.8**	**3.5**	**3.4**	0.8
BDE-99	0.5	**1.9**	**2.9**	**2.6**	**13**	0.9
BDE-153	0.7	**1.4**	**3.0**	**1.2**	**11**	-
BDE-154	0.6	**1.2**	**3.3**	**1.5**	**7.0**	-
BDE-183	**1.1**	0.9	**5.0**	**1.5**	**19**	-
BDE-209	**3.7**	**1.4**	**4.3**	**2.5**	na	**2.2**
EH-TBB	**2.2**	**1.2**	**1.9**	**1.9**	na	-
BTBPE	**1.1**	**1.1**	-	**2.1**	-	**1.3**
BEH-TEBP	**1.9**	**1.3**	**1.2**	**2.0**	na	0.5
DBDPE	**1.5**	**1.5**	**2.2**	0.5	na	**3.4**
HBCDD	na	na	**1.2**	na	na	na
OPEs						
Target PFR	**Norway (*)**	**Japan (**)**	**China**	**Egypt**	**USA**	**UK**
TNBP	0.5 (-)	**1.1 (**-)	-	-	na	**3.2**
TCEP	**2.3 (**1.0**)**	**1.4** (-)	0.9	-	**1.4**	**2.4**
TCIPP	1.0 (**2.6**)	**3.0 (3.0)**	**2.8**	**1.5**	**1.4**	**1.5**
TDCIPP	**1.1 (2.8)**	**3.9 (-)**	0.7	**2.2**	**2.7**	**1.3**
TBOEP	0.6 (0.8)	0.2 (0.9)	**1.4**	0.3	na	**-**
TPHP	**1.3 (1.7)**	**2.5 (3.6)**	0.3	0.8	na	**1.2**
TMPP	**1.3 (1.9)**	**- (-)**	0.3	-	na	-
EHDPP	0.9 (**1.5**)	na (na)	na	-	na	0.6

na = not applied, - = not detected or detection frequencies <50%, bold values= median ESD:FD ratios >1, (*) = Xu et al., 2016, (**) =Tajima et al., 2014

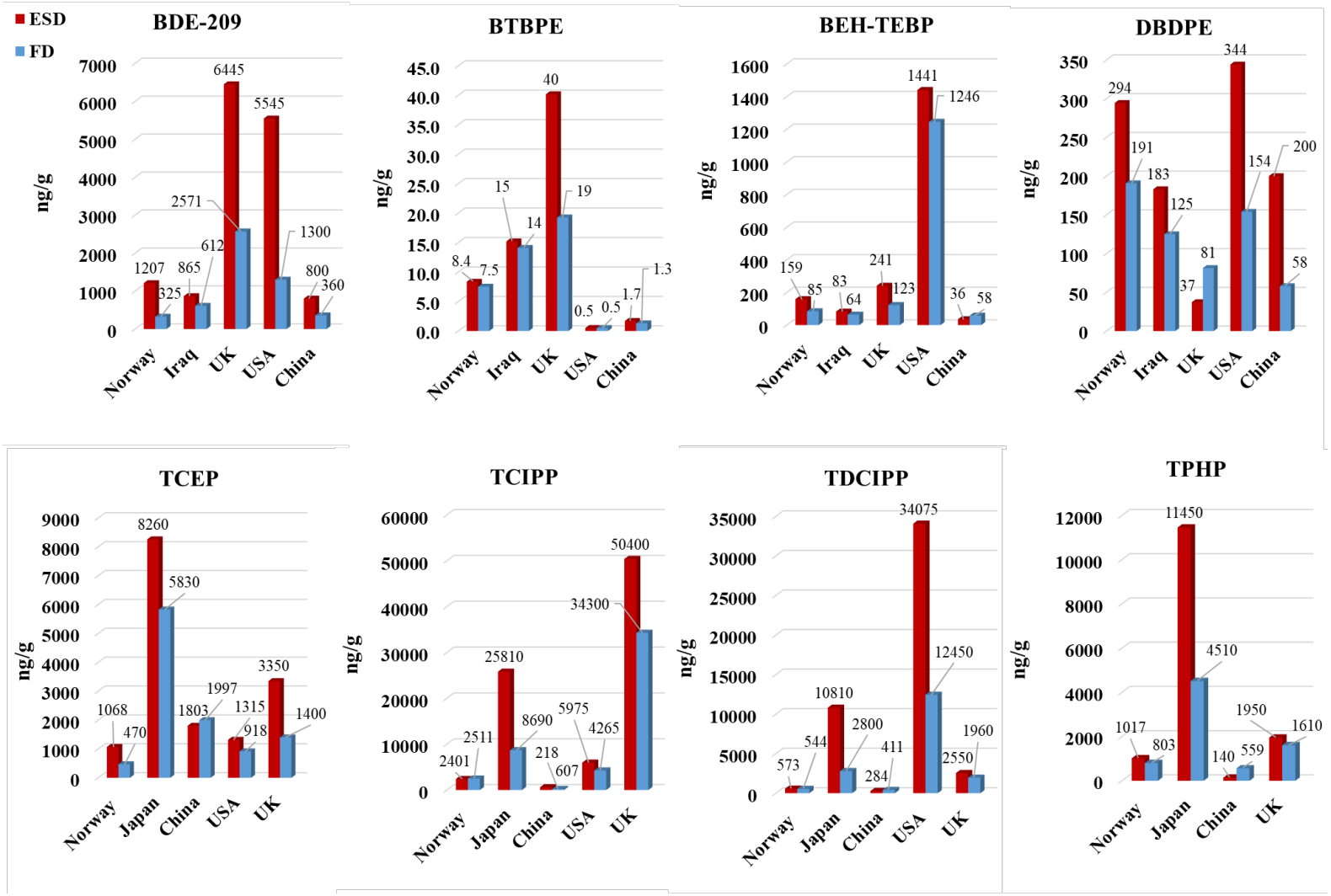

Figure 1. Comparison of median concentrations of the most abundant BFRs and OPEs in elevated surface dust (ESD) vs. floor dust (FD) from different regions and countries (Cequier et al., 2014 (Norway); Araki et al., 2014 (Japan); Al-Omran and Harrad, 2016a (Iraq); 2018 (UK); Wu et al., 2016 (China); Allgood et al., 2017a (USA); Ortiz Carrizales, 2018 (UK); Bu et al., 2019 (China)

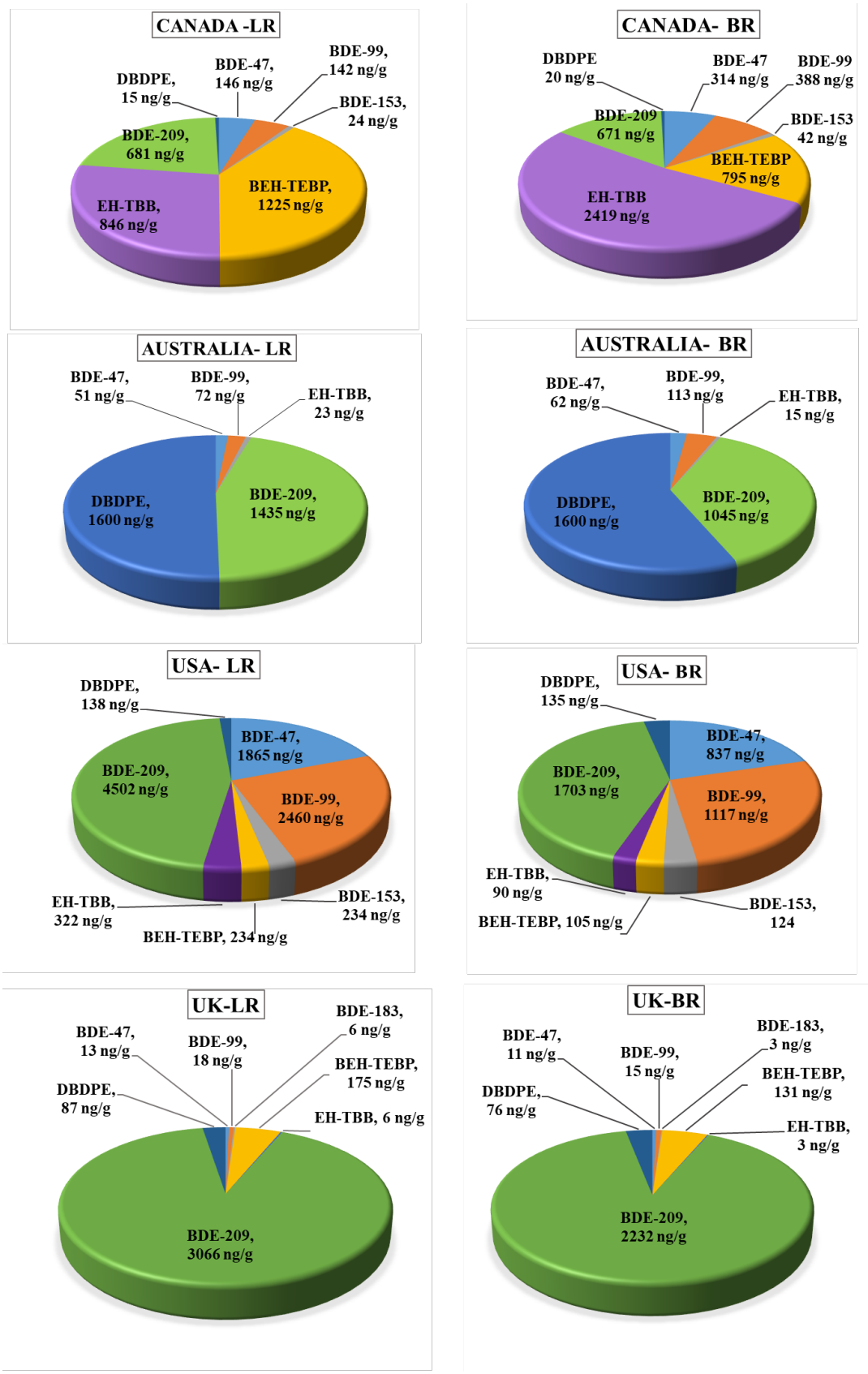

Figure 2. Relative abundance and median concentrations of PBDEs and NBFRs in floor dust from living rooms (LR) and bedrooms (BR) from Canada (Venier et al., 2016), Australia (McGrath et al., 2018), USA (Allen et al., 2008; Stapleton et al., 2008) and UK (Al-Omran and Harrad (2017).

794 **Table 2. Maximum:Minimum concentration ratios of selected BFRs and OPEs in different particle size fractions of indoor dust**

BFRs										
Microenvironment	**Fraction (n)**	**BDE-47**	**BDE-99**	**BDE-153**	**BDE-154**	**BDE-183**	**BDE-209**	**BEH-TEBP**	**DBDPE**	**Reference**
Dining hall, China	<43- 200 μm (5)	1.6	1	1.3	2	2.4	2.2	na	na	Yu et al., 2013
Laboratory, China	<43- 200 μm (5)	26	5	6.1	6	1.7	2.7	na	na	Yu et al., 2013
Office, South Africa	<45-150 μm (3)	3.3	4	2.6	2	1.5	4.2	na	na	Kefeni and Okonkwo, 2014
Home, South Africa	<45-150 μm (3)	2.2	3	3.2	nd	nd	1.6	na	na	Kefeni and Okonkwo, 2014
Office, China	<50-500 μm (7)	na	na	na	na	na	5.1	6	5.9	Cao et al., 2014a
Hotel, China	<50-500 μm (7)	na	na	na	na	na	15	2	6	Cao et al., 2014a
Home, UK	<63-250 μm (3)	1.2	2	1.3	1	1.7	1.1	1	1.3	Al-Omran and Harrad 2016b
Home, Japan	<53-500 μm (3)	1.4	2	na	na	1.3	2.1	na	na	Kajiwara and Takigami, 2016
Office, China	<43- 200 μm (5)	nd	nd	1.8	1	1.7	1.4	5	1.6	He et al., 2018
OPEs										
Microenvironment	**Fraction (n)**	**-**	**TNBP**	**TCEP**	**TCIPP**	**TDCIPP**	**TPHP**	**TMPP**	**EHDPP**	
Office, China	<50-500 μm (7)	-	189	na	an	12	11	5	2.9	Cao et al., 2014a
Hotels, China	<50-500 μm (7)	-	43	na	na	5	4	3	4	Cao et al., 2014a
Office, China	<43- 200 μm (5)	-	na	2.2	1	2.9	na	na	4.1	He et al., 2018
Home, China	>25-36 μm (4)	-	na	2.6	3	1.8	1.7	2	na	Li et al., 2019
Home, Stockholm	>25-36 μm (4)	-	na	2.2	2	1.5	2.1	2	na	Li et al., 2019

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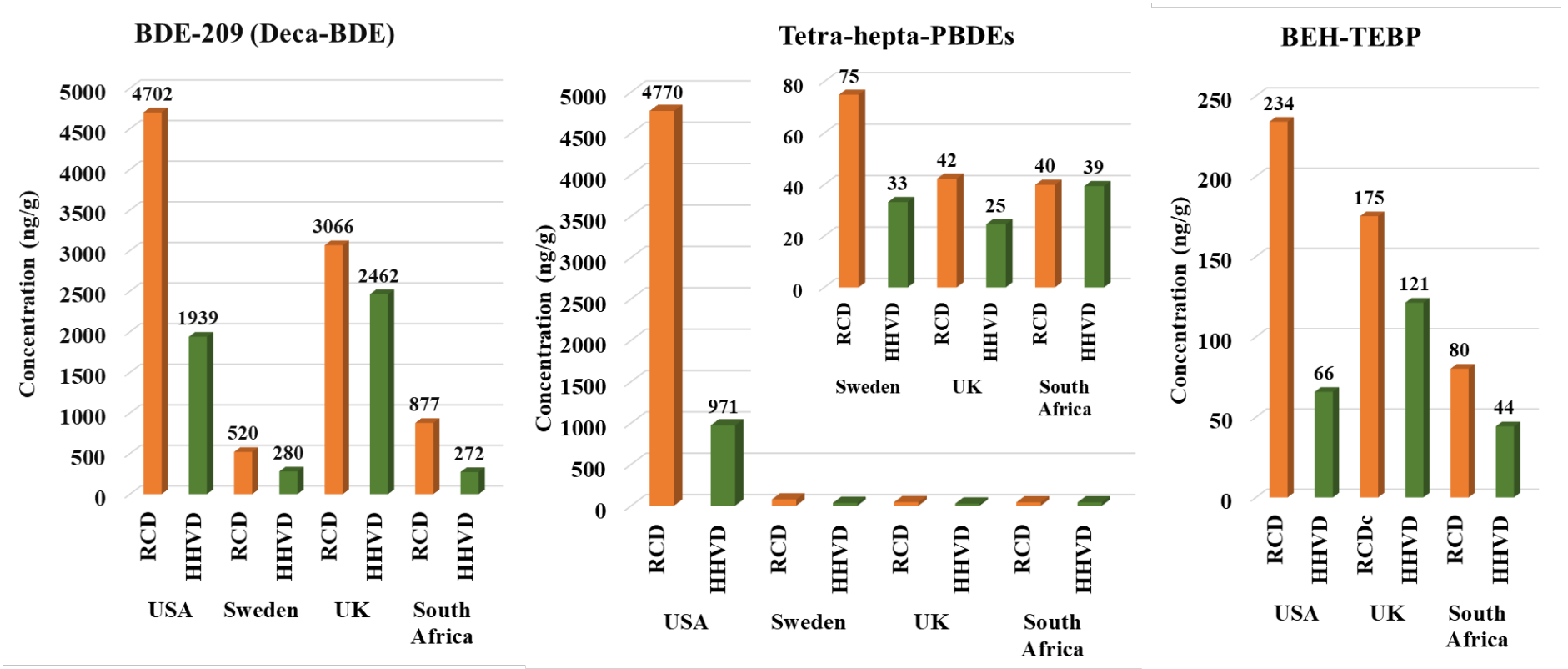

Figure 3: Median (GM for USA) concentrations (ng/g) of selected BFRs in RCD (researcher collected dust) and HHVD (household vacuum dust) from USA (Allen et al., 2008 and Stapleton et al., 2008), Sweden (Björklund et al., 2012), UK (Al-Omran and Harrad, 2017) and South Africa (Brits et al., 2019)

List of ABBREVIATIONS

ANOVA, Analysis of variance

BDE-153, (2,2',4,4',5,5'-hexabromodiphenyl ether

BDE-154, (2,2',4,4',5,6'-hexabromodiphenyl ether

BDE-183, (2,2',3,4,4',5',6-heptabromodiphenyl ether

BDE-209, (2,2',3,3',4,4',5,5',6,6'-decabromodiphenyl ether

BDE-47, (2,2',4,4'-tetrabromodiphenyl ether

BDE-99, (2,2',4,4',5-pentabromodiphenyl ether

BEH-TEBP, Bis(2-ethyl-1-hexyl)tetrabromophthalate

BFR, brominated flame retardant

BTBPE, 1,2-bis(2,4,6-tribromophenoxy)ethane

DBDPE, Decabromodiphenylethane

Deca-BDE, decabromodiphenyl ether

EHDPP, 2-Ethylhexyl diphenyl phosphate

EH-TBB, 2-ethylhexyl-2,3,4,5-tetrabromobenzoate

FRs, flame retardants

HBCDD, hexabromocyclododecane

K_{OA}, octanol-air partition coefficient

NBFRs, “Novel” brominated flame retardants

OC, organic carbon contents content

Octa-BDE, octabromodiphenyl ethers

Pa, Pascal

PBDE, polybrominated diphenyl ether

Penta-BDE, pentabromodiphenyl ether

OPEs, organophosphate esters

POPs, persistent organic pollutants

RSD, relative standard deviation

SRM2585 and 2584, dust standard reference material

SVOCs, semi-volatile organic compounds

TBOEP, tris (2–butoxyethyl) phosphate

TCEP, tri(2-chloroethyl) phosphate

TCIPP, Tris(2-chloroisopropyl) phosphate

TDCIPP, (Tris (1,3-dichloro-2-propyl) phosphate

TMPP, Tris(4-methylphenyl) phosphate

TNBP, tri-n-butyl phosphate

TPHP, triphenyl phosphate

UNEP, United Nations Environment Programme

VP, vapour pressure

WHO, World Health Organization

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