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1	A Meta-Analysis of Factors Influencing Concentrations of
2	Brominated Flame Retardants and Organophosphate Esters in
3	Indoor Dust
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29 Abstract

30 Current assessments of human exposure to flame retardants (FRs) via dust ingestion rely on 31 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-32 room and within-building spatial and temporal variability, differences in dust particle size 33 fraction analysed, as well as differences in dust sampling approach. A meta-analysis of peer-34 reviewed data was undertaken to evaluate the impact of these factors on reported concentrations 35 of brominated flame retardants (BFRs) and organophosphate esters (OPEs) flame retardants in 36 dust and subsequent human exposure estimates. Except for a few cases, concentrations of FRs 37 38 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 39 40 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 41 dust. Considerable within-building spatial variability was observed with no specific trend 42 between concentrations of either BFRs or OPEs in living rooms and bedrooms in the same 43 homes, implying that exposure assessments based solely on sampling one room are uncertain. 44 Substantial differences in FR concentrations were observed in different particle size fractions 45 46 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 47 assessment as adherence to skin and subsequent FR uptake via ingestion and dermal sorption 48 varies with particle size. Analysing dust samples obtained from a householder vacuum cleaner 49 (HHVC) compared with researcher collected dust (RCD) will underestimate human exposure 50 to the most of studied contaminants. This is likely due to the losses of volatile FRs from HHVC 51 dust over the extended period such dust spends in the dust bag. Temporal variability in FR 52 concentrations is apparent during month-to-month or seasonal monitoring, with such 53 54 variability likely due more to changes in room contents rather than seasonal temperature 55 variation.

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57 Keywords

58 BFRs, OPEs, indoor dust, exposure assessment

60 Highlights

61	• FR concentrations vary significantly between floor and elevated surface dust
62	• FR contamination of floor dust can vary significantly by within-room and within-
63	building spatial variability
64	• The most suitable dust size fraction for exposure assessment is $<150 \ \mu m$
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88 **1. Introduction**

In recent decades, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane 89 90 (HBCDD) have been the most extensively used additive brominated flame retardants (BFRs) in consumer products such as polyurethane foam, electrical and electronic equipment, textiles, 91 and construction materials. A large number of studies have indicated that PBDEs and HBCDD 92 are persistent and bioaccumulative toxicants, which adversely affect human health (WHO, 93 2003; van der Ven et al., 2009; Noyes et al. 2010). Health concerns of PBDEs and HBCDD 94 exposure include endocrine disruption (Johnson et al., 2013), impaired fertility (European 95 Commission, 2012), neurotoxic effects (Schecter et al., 2012), and possible carcinogenicity 96 97 (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 98 99 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 100 such as "novel" brominated flame retardants (NBFRs). NBFRs as relating to brominated flame 101 retardants are defined here as those new to the market or recently observed in the environment 102 such as **BEH-TEBP** (Bis(2-ethyl-1-hexyl) tetrabromophthalate), DBDPE 103 (decabromodiphenylethane). In addition, organophosphate ester (OPEs) compounds which are 104 extensively used as flame retardants and plasticizers. (Ven der Veen and de Boer, 2012; 105 Greaves and Letcher, 2017; Blum et al., 2019). It is, however, expected that PBDE 106 formulations may maintain a presence in new products due to the use of recycled materials, 107 108 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 2.8 109 million tonnes in 2018, and to be growing by 4.6% per year (Yasin et al, 2016). In 2019, BFRs 110 and OPEs represented 17% and 18% respectively of the global production of flame retardants, 111 with Asia the largest market, followed by North America, Europe, Middle East, and Latin 112 113 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 122 BFRs in indoor dust and human biological samples such as human milk (Toms et al., 2009; 123 Kim et al., 2014) and serum (Stapleton et al., 2012; Ma et al., 2017) suggesting that indoor 124 125 settled dust ingestion is a major pathway of exposure to such chemicals (Tue et al., 2013; de 126 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 127 such dust is ingested. Such methodology provides only an indication of the potential exposure 128 via this pathway; with true exposure dependent on a variety of factors such as the amount of 129 time an individual spends in the room from which dust was sampled, the number and extent 130 of hand-to-mouth events that occur (widely assumed greater for toddlers than adults), as well 131 as factors such as the dustiness of the room, whereby dust ingestion is likely greater in dustier 132 rooms. Thus, it is important that indoor dust samples collected and analysed as part of such 133 exposure assessments are truly representative of the dust to which humans are exposed. 134 135 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 136 uncertainty inherent in basing estimates of population-level exposure on limited sample sizes, 137 138 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 139 140 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, 141 142 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 143 144 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 145 exposure estimates based only on floor dust may not reflect the actual exposure to FRs of 146 adults. While toddlers and young children are likely more exposed to floor dust, adults likely 147 have far greater contact with dust from elevated surfaces - e.g. desk and table tops. Hence, if 148 dust concentrations vary significantly between elevated surfaces and the floor in the same 149 150 room, one type of dust sample may not represent an acceptable estimate of exposure for all 151 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 withinbuilding spatial variability, (b) dust particle size, (c) sampling method, and (d) temporal/seasonal variability.

158 **2. Methodology**

The BFRs and OPEs targeted in this study comprise the following: BDE-47 (2,2',4,4'-159 tetrabromodiphenyl ether), BDE-99 (2,2',4,4',5-pentabromodiphenyl ether), BDE-153 160 (2,2',4,4',5,5'-hexabromodiphenyl ether), BDE-154 (2,2',4,4',5,6'-hexabromodiphenyl ether), 161 BDE-183 (2,2',3,4,4',5',6-heptabromodiphenyl ether), BDE-209 (2,2',3,3',4,4',5,5',6,6'-162 decabromodiphenyl ether), HBCDD (hexabromocyclododecane), EH-TBB (2-ethylhexyl-163 2,3,4,5-tetrabromobenzoate), BTBPE (1,2-bis(2,4,6-tribromophenoxy)ethane), BEH-TEBP 164 (Bis(2-ethyl-1-hexyl) tetrabromophthalate), DBDPE (decabromodiphenylethane), TNBP (tri-165 n-butyl phosphate), TCEP (tris (2-chloroethyl) phosphate), TCIPP (tris (2-chloroisopropyl) 166 phosphate), TDCIPP (tris (1,3-dichloro-2-propyl) phosphate), TBOEP (tris (2-butoxyethyl) 167 phosphate), TPHP (triphenyl phosphate), TMPP (tris(4-methylphenyl) phosphate) and EHDPP 168 (2-ethylhexyl diphenyl phosphate). These are selected on the basis that they are widely 169 used and the most commonly studied FRs and OPEs with respect to their presence in indoor 170 171 dust.

The data used for this meta-analysis is based on papers published in peer-reviewed journals 172 between 1st January 2000 and 1st April 2020, and one PhD thesis (Ortiz Carrizales, 2018). The 173 PhD thesis was included as it contains otherwise unpublished material. All datasets were 174 identified through targeted search terms via the most popular search engines such as Science 175 Direct, Scopus, and Google Scholar, using the terms "BFRs", "PFRs", "OPEs", "OPFRs", 176 "NBFRs", "PBDEs", "HBCDD", "HBCD", "indoor dust", "human exposure", "elevated 177 surface", "multi surface", "floor", "particle size", "sampling method", "sampling approach", 178 "spatial", living room", "bedroom", "kitchen", "temporal", "seasonal" and "variability". These 179 terms were classified into four groups depending on the investigated factor, which include: 180 spatial variability, particle size distribution, sampling method, and temporal variability of FR 181 concentrations in indoor dust. Figure S1 shows study selection flow diagram. All data included 182 in our evaluation are reported in the original publication. Comparison across some studies 183

required statistical analysis of the raw data which were requested from the authors or obtained 184 185 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. 186 187 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, 188 189 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 190 191 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 192 from the original papers. Data were excluded from the evaluation when detection frequency of 193 the target FR(s) was < 50%, to avoid bias caused by the method of substitution for non-detects 194 in the respective sample group. Table S1 lists the 45 original studies included in this meta-195 196 analysis.

197 **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, 206 2016a), USA (Allgood et al., 2017), UK (Al-Omran and Harrad 2018), Egypt (Khairy and 207 Lohmann, 2018), and China (Bu et al., 2019); median concentrations of most PBDEs and 208 NBFRs in ESD from the USA, UK, Iraq, and Egypt exceeded those in floor dust samples taken 209 210 in the same rooms by factors ranging between 1.1 and 19. A deviation from this overall trend 211 was the observation that median ESD:FD ratios in dust samples from Norwegian households 212 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 213 the different particle size of the two dust sample categories collected in this Norwegian study. 214 Specifically, FD was collected using a nozzle with a polyethylene grid (1–3 mm pore size) 215 while ESD was collected without the grid. This suggests that the floor dust collected may have 216

contained a greater proportion of fine particles preferentially contaminated with lower 217 brominated PBDEs. Similarly, in day care centres in China, median concentrations in ESD 218 exceeded those in FD for only 3 out of 7 BFRs (Bu et al., 2019). For OPEs, median ESD:FD 219 220 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; 221 222 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 223 224 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 225 the day care and apartment dust studied in China and Egypt respectively. In contrast to TPHP 226 and chlorinated OPEs, concentrations of TBOEP in FD exceeded those in ESD with median 227 ESD:FD ratios <1 (Cequier et al., 2014; Xu et a., 2016; Araki et al., 2014; Tajima et al., 2014; 228 Khairy and Lohmann, 2019). This may be due to the wide use of TBOEP in flooring and floor 229 polishing materials (Kajiwara et al., 2011; van der Veen and de Boer, 2012). Table 1 shows 230 the median ratios of the concentrations of FRs detected in ESD and FD collected from the 231 same rooms for various studies. Figure 1 shows the median concentrations of FRs present in 232 ESD and FD samples collected in the same studies. 233

Statistically significant differences were observed in concentrations of FRs between ESD and 234 235 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, 236 237 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 238 239 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 240 241 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 242 243 (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 244 contrast, TBOEP concentrations were significantly higher in FD than ESD in Norway 245 (calculated from the raw data of Cequier et al., 2014) and Japan (Araki et al., 2014, using 246 Wilcoxon rank test). Tables S2 and S3 show the outcomes of paired t-test comparisons of 247 concentrations of BFRs and OPEs in ESD and FD respectively. 248

249 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 250 significantly (p < 0.05) correlated with those in FD samples from the same room. For BFRs, 251 252 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), 253 254 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-255 256 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 257 significantly correlated with those in FD. In addition, chlorinated OPEs showed significant 258 positive linear correlation between ESD and FD for TCEP in USA (calculated from the raw 259 data of Allgood et al., 2017), UK (calculated from the raw data of Ortiz Carrizales, 2018), and 260 China (Wu et al., 2016), as well as for TCIPP in Japan (Tajima et al., 2014) and China (Wu et 261 al., 2016). This suggests that the sources of these contaminants in ESD and FD are similar. 262 Tables S4 and S5 show the results of Pearson correlation analyses of concentrations of BFRs 263 and OPEs in paired samples of ESD and FD reported in various studies. 264

265 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 266 267 volatilisation and subsequent sorption to dust particles governed by the contaminant's octanolair partition coefficient (K_{OA}) (Weschler and Nazaroff, 2010, Fromme, 2012). This 268 269 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 270 Harrad 2016a), and TCEP (Xu et al., 2016; Wu et al., 2016). However, for lower vapour 271 272 pressure compounds such as decabromodiphenyl ether (BDE-209), this explanation likely 273 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 274 make a greater contribution. These include: abrasion of particles/fibres of FR-treated materials 275 and transfer via direct source:dust contact (Rauert and Harrad, 2015; Rauert et al., 2016). Table 276 S6 shows physicochemical properties of our target FRs. In addition to differences between 277 ESD and FD in dust particle size distribution, differences in the number and type of FR sources 278 present on floors and elevated surfaces, are likely important factors influencing differences in 279 concentrations of FRs between ESD and FD from the same microenvironment. 280

Building materials are an important source of FRs, especially floor materials (Kanazawa et al., 281 282 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 283 284 concentrations observed in floor as opposed to elevated surface dust. In contrast, woodenstructure houses displayed lower levels of TCIPP, TCEP, and TPHP in FD than other types of 285 286 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 polyure than foam 287 288 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 289 chlorinated OPEs between ESD and FD are similar. Specifically, except for TCEP in Norway 290 (for which the median ESD:FD ratio = 1); median ESD:FD ratios of TPHP (1.7, 2.5 and 1.2)291 were consistent (>1) with those of TCEP (1.0, 1.4 and 2.4), TCIPP (2.6, 3.0 and 1.5) and 292 293 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 294 air before settling on elevated surfaces (Wu et al., 2016). This may explain the presence of 295 lower vapour pressure compounds in ESD even where floor materials are the source. Also, as 296 finer particles are more atmospherically mobile, ESD likely contains a greater proportion of 297 fine particles containing chemicals that have accumulated via volatilisation and subsequent 298 deposition (Rauert and Harrad, 2015). Moreover, where ESD samples are collected from 299 300 surfaces that include putative sources (e.g. TVs and other electronics), a proportion of such 301 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 302 303 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 304 305 likely to dilute the concentration of pollutants in FD (Cao et al., 2014a). Conversely, the lowest 306 ESD:FD ratios of BFRs (Table 1) reported in dust samples taken from Iraq (Al-Omran and 307 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 308 309 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 310 between such dust types. The authors reported significantly (p < 0.05) higher proportions of fine 311 particles (<125 µm) in ESD samples and no substantial influence of OC on the observed 312

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

315 **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, 316 shelves, chairs, etc. exceeded significantly those in FD. However, the majority of assessments 317 of human exposure to FRs via indoor dust ingestion have to date focused on collection and 318 319 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, 320 321 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 322 323 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 324 BDE-99, BEH-TEBP, BDE-209, chlorinated OPEs, and TPHP. For example, such 325 underestimates for the adult population can be substantial e.g., reaching as high as factors of 326 5.0 and 19 for "typical" exposure rate of BDE-183 in USA (Allgood et al., 2017) and Egypt 327 (Khairy and Lohmann, 2018) respectively. 328

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330 3.1.2 Within-room spatial variability in floor dust from different areas of the same 331 microenvironment

Several studies report substantial within-room spatial variability in concentrations of FRs in 332 floor dust taken at the same time from different areas of the same room. For BFRs, from five 333 different microenvironments (3 homes and 2 offices), 5 floor dust samples were collected on 334 335 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 336 337 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 338 (Melymuk et al., 2016). To differentiate spatial variability from analytical variability, the high 339 RSD values of PBDEs (30-183%) and HBCDDs (36-93%) were compared with those 340 341 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). 342 343 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 344 Czech Republic (calculated from the raw data of Melymuk et al., 2016). 345

Other studies in the UK tested within-room spatial variability in concentrations of BFRs 346 347 (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). 348 Significant differences (p <0.05) were observed in concentrations of Σ_7 tri-hepta-BDEs and 349 Σ NBFRs in different areas of the same room for three out of nine rooms studied, but not for 350 351 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 352 353 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 354 associated with floor materials such as tatami, tiles, and wall-to-wall carpet (Tajima et al., 355 2014). However, TCIPP displayed strong spatial variability in dust samples from the Czech 356 Republic. Concentrations of TCIPP decreased with increasing distance from a sofa, implying 357 the use of TCIPP in the foam filling of the sofa concerned (Jilkova et al., 2018). 358

Overall, the evidence suggests higher concentrations of FRs accumulate in dust samples located 359 360 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. 361 362 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 363 bare floor and carpeted floor areas within the same room (Al-Omran and Harrad, 2018). In 364 addition, in one office (1450 m²) from China, higher concentrations of PBDEs were detected 365 366 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 367 in different microenvironments, while Figure S2 compares median concentrations of selected 368 BFRs and OPEs in floor dust from two different areas of the same room located in the UK. 369

These results indicate that appreciable variation in FR contamination can be found, depending 370 371 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 372 contamination of the room as a whole. For example, if in a given room (assuming the two floor 373 areas are F1 and F2), the concentration of \sum_{7} tri-hepta-BDEs in F1 exceeds substantially that in 374 dust samples from F2 with ratio of F1:F2=61:4 for one sampling event; in this room, the 375 exposure estimate will vary by a factor of just over 15 depending on the area samples (Al-376 Omran and Harrad, 2018). Overall, while the substantial and sometimes significant variation 377 in FR concentrations in floor dust from different areas of the same room, may imply that 378

- 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.

383 **3.1.3 Within-building spatial variability**

384 Within building spatial variability refers to whether concentrations of contaminants in dust vary 385 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., 386 387 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 388 389 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 390 microenvironments (Dodson et al., 2017; Jílkova et al. 2018). 391

In two studies from the USA (Allen et al., 2008; Stapleton et al., 2008), concentrations of Penta-392 BDE, Deca-BDE, HBCDD, EH-TBB, and BEH-TEBP were significantly (p < 0.05) higher in 393 the main living area compared to the bedroom. No significant differences were apparent for 394 BTBPE and DBDPE (Stapleton et al., 2008). In UK house dust samples, median concentrations 395 396 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). 397 398 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 399 400 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 401 402 (Venier et al., 2016). Except for EH-TBB in the USA and BEH-TEBP in Canada, median 403 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, 404 Canada, and the Czech Republic respectively (calculated from the raw data of Venier et al., 405 2016). In Australia, except for BDE-154 (for which concentrations were significantly higher 406 in the living room), no significant differences were found between concentrations of PBDEs 407 408 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 409 410 PBDEs and NBFRs in living rooms (LR) and bedrooms (BR) from Canada, Australia, USA,

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

In addition to the above studies, three further UK studies (Muenhor and Harrad 2012; Al-413 Omran and Harrad 2018; Ortiz Carrizales, 2018) investigated within-home spatial variability 414 in concentrations of PBDEs, NBFRs, and OPEs in different rooms from a limited number (2-415 416 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 instance, while in one study, concentrations of BDE-47 and BDE-99 in the living room were 418 significantly higher (p<0.05) than those in the bedroom (Muenhor and Harrad, 2012), in 419 another they were significantly higher in the bedroom than the living room (Al-Omran and 420 Harrad, 2018). For OPEs, no significant differences were found between living rooms and 421 bedrooms (Ortiz Carrizales, 2018). 422

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

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

443 al., (2017) found concentrations of TCEP and TPHP in student dormitory rooms to exceed 444 significantly (p < 0.05) those in dust from common areas, while concentrations of BDE-209 in 445 common areas were significantly higher than in dormitory rooms (Dodson et al., (2017).

446 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 447 448 rooms in the numbers and types of potential FR emission sources present. For example, higher 449 concentrations of PBDE congeners representative of the Penta- and Deca-BDE formulations in living rooms compared to be drooms may be attributed to the presence in the former, of specific 450 putative source items such as televisions and couches (Allen et al., 2008). Moreover, higher 451 concentrations of TCEP and TPHP in student dormitory rooms, compared to common areas, 452 may be related to the electronics and furnishings introduced to dormitories by students (Dodson 453 et al., 2017). Similar explanations were proposed by other studies (Stapleton et al., 2008; 454 Muenhor and Harrad, 2012; Al-Omran and Harrad, 2017; 2018; Jílkova et al., 2018; McGrath 455 et al., 2018), which is supported by the lack of strong correlations between FR concentrations 456 in rooms within the same building, as indicated by Table S8 that shows Pearson's correlation 457 458 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 459 bedrooms and living rooms in the same home suggests that emission sources are similar in both 460 461 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 462 463 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 464 465 subsequent deposition to dust; but is less plausible when other source-to-dust transfer 466 mechanisms dominate, such as source abrasion and direct source-dust contact.

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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 \pm SD = 3992 \pm 1906 ng/g) exceeded those in the living room (average \pm SD = 1811 \pm 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.

475 **3.2** The influence of dust particle size fraction

Dust from indoor microenvironments includes a wide range of particle sizes ranging from < 476 477 2.5 μ m to over 2000 μ m in diameter. Particles > 30 μ m may be classified as settled particles, which include "skin flakes, fragments of hair, microorganisms, such as fungal spores and 478 479 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 480 481 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), 482 483 $<100 \ \mu m$ (Kang et al., 2011), and $< 63 \ \mu m$ (Kopp et al., 2012). However, for assessment of exposure via dust ingestion, it is reasonable to assume that only those particles adhering to 484 human hands and that can subsequently be orally ingested via hand-to-mouth contact and/or 485 release FRs via dermal uptake are pertinent. The most recent recommendation by the USEPA 486 is that adherence to hands is greatest for dust particles <150 µm diameter (USEPA, 2016), 487 which is smaller than their previous recommendation to focus on particles <250 µm diameter 488 (USEPA, 2008). Moreover, increased FR bioaccessibility via dust ingestion was observed with 489 decreasing particle size (Yu et al., 2013; Fang and Stapleton, 2014). This section highlights the 490 extent to which analysing different dust fractions influences FR concentrations and consequent 491 492 exposure assessments.

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494 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 495 varied between different microenvironments. The first investigation of FRs in different particle 496 size fractions of indoor dust was reported by Wei et al., (2009) in the USA, concluding that 497 while in car dust, PBDE concentrations were inversely related to particle size, they were largely 498 comparable in four dust fractions (250- 420 μ m, 150-250 μ m, 75-150 μ m, and < 75 μ m) for 499 home dust. A comparison of dust from different microenvironments (offices, hotels, 500 kindergartens, and dormitories), showed that concentrations of DBDPE and HBCDD in hotel 501 dust were highest in fine particles (50-74 μ m and >50 μ m respectively), while in office dust 502 concentrations were highest in medium particles (100-200 µm) (Cao et al., 2014a; 2015). 503 504 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 505 strongly in both fine (43-63 µm) and finest (<43 µm) particles of office dust, while they were 506 present at higher concentrations in coarse particles (150-200 µm) in public microenvironments 507 (PMEs) (He et al., 2018). In addition, differences in distribution pattern of FRs between 508 different dust particle sizes were found between different countries from the same type of 509

microenvironment. While the concentration of \sum_7 OPEs was higher in particles 25-75 µm in homes from China, concentrations were higher in the finest particles (<25 µm) in Swedish homes (Li et al., 2019). It can be concluded that different microenvironments can show different distribution patterns of FRs in various particle size fractions with no specific trend of a given pollutant to accumulate in a specific size fraction. Figure S3 demonstrates the different distribution patterns of OPEs in different particle size fractions (<25-<500 µm) from different microenvironments and different countries.

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In other studies, the distribution pattern of FRs in different dust particle size fractions revealed 518 generally that concentrations of BFRs tend to increase with decreasing particle size, with some 519 significant differences. In three particle size fractions (106-150 μ m, 45-106 μ m, and <45 μ m) 520 of dust samples from 2 homes and 2 offices, concentrations of BDE-99 and BDE-153 increased 521 with decreasing particle size, while other PBDE congeners showed higher concentrations in 522 the fine (<45 µm) and medium particle size ranges (45-106 µm) (Kefeni and Okonkwo, 2014). 523 Elsewhere, in three size fractions of 10 dust samples from 5 homes in the UK, while 524 concentrations of Σ tri-hepta-BDEs and BEH-TEBP in the finest particles (<63 µm) exceeded 525 significantly those in medium (63 - $<125 \mu m$) and coarse particles ($<125-250 \mu m$); median 526 concentrations of BDE-209 in these three particle size fractions were comparable (Al-Omran 527 528 and Harrad 2016b). The same trend was found for PBDEs in four size-fractionated dust samples from 5 homes in Japan (Kajiwara and Takigami, 2016). Concentrations of PBDEs in dust 529 tended to increase with decreasing particle size, but concentrations in the finest particles (<50 530 µm) were not the highest. One study in 8 homes in Taiwan investigating PBDEs in three 531 particle size fractions (>149 μ m, 75-149 μ m, and <75 μ m) proposed that PBDE concentrations 532 may not be correlated with the particle size (Chao et al., 2014). For OPEs, in dust from 7 533 building material markets from Germany, most OPEs targeted showed no consistent trend with 534 particle size (Zhou and Puttmann, 2019). The exception to this was TCIPP, which was enriched 535 in medium size ($< 63-150 \mu m$) particles. From the above studies, it can be concluded that due 536 to the greater surface area to volume ratio of fine particles, the impact of atmospheric deposition 537 of FRs will be greater on fine indoor dust particles. 538

Contaminants with relatively high vapour pressures (VP) are expected to migrate from sources to the environment via evaporation and subsequent deposition to particles, depending on the octanol-air partition coefficient (K_{OA}) of the contaminant and the organic carbon content of indoor dust particles (Webster et al., 2009; Weschler and Nazaroff, 2010; Yu et al., 2013). 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 544 TCEP (VP= 5.2×10^{-2} Pa) were found in the finest (<43 µm) particles with maximum:minimum 545 ratios of 2.2, 1.4 and 1.3 in office, public microenvironment and car dust samples respectively. 546 However, concentrations of TCIPP (VP=7.4x10⁻³ Pa), close to that of TCEP) were higher in 547 the coarse fraction (100-200 µm). In contrast, the concentrations of DBDPE and BDE-209 548 $(VP=2.6x10^{-11} \text{ and } 9.3x10^{-9} \text{ Pa respectively})$, were higher in the finest particles of the same 549 samples (He et al., 2018; Cao et al., 2013). These findings are consistent with other studies, 550 551 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, 552 the concentration of BDE-209 was higher in the fine particles ($<43-63 \mu m$) (Yu et al., 2013). 553 The above examples suggest that while in many scenarios, the predominant source-to-dust 554 transfer mechanism is volatilisation followed by deposition to finer particles with greater 555 specific surface area; in some situations, FRs in indoor dust particles might arise predominantly 556 from a source weathering or abrasion process rather than volatilisation/deposition. Thus, in 557 such situations, irrespective of the vapour pressure, the size of the abraded, FR-rich particles is 558 likely to determine the fraction with the highest FR concentration. Overall, regardless of the 559 predominant source-to-dust transfer mechanism, our meta-analysis suggests that significant 560 differences do exist between concentrations of FRs in different particle size fractions. This 561 supports focusing on analysis of those particle sizes shown to adhere to hands as the most 562 relevant for human exposure via unintentional dust ingestion. Current USEPA 563 564 recommendations are that these are particles <150 µm diameter (USEPA, 2016).

565

566 **3.3 Evaluation of dust sampling approaches**

Dust sampling approaches can be divided into passive and active techniques. The passive 567 technique includes collecting dust via natural accumulation of suspended particles onto 568 collection receptacles for a given period (Butte and Heinzow, 2002; Niu et al., 2018; 2019). 569 This method is rarely used due to the time required to collect sufficient dust for analysis 570 (Mercier et al., 2011). Active techniques involve several approaches including wiping (Abbasi 571 et al., 2016; Xu et al; 2019), sweeping (Qi et al., 2014; Zeng et al., 2018; Iwegbue et al., 2019), 572 and vacuuming (Fromme et al., 2014; Tao et al., 2016). The wiping method was originally used 573 to collect settled dust by rubbing hard surfaces with alcohol moistened paper towels (USEPA, 574 1995). Although this method is not recommended for collection of surface dust due to poor 575 collection efficiency of fine particles (USEPA, 2008), it is widely applied for collection of 576

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).

580

The most widely used sampling technique is vacuuming, most commonly with conventional 581 582 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 583 584 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 585 vacuum dust (HHVD), where householders provide researchers with their vacuum cleaner 586 contents (Shen et al., 2015; Ali et al., 2016). The most common accessories that have been used 587 for RCD are nylon "socks" (Harrad et al., 2016), filters (Newton et al., 2015), and Soxhlet 588 589 thimbles (Stapleton et al., 2012), all deployed to collect dust before it enters the vacuum cleaner 590 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 591 what extent the sampling method used influences FR concentrations in the dust collected and 592 593 the consequent exposure assessment.

594

595 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 596 congeners in RCD (obtained using cellulose extraction thimbles inserted in the vacuum cleaner 597 sampling hose) collected from living rooms and those of Penta-BDE congeners in bedrooms 598 599 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 600 collected in bedrooms. In general, concentrations of PBDEs were weakly to moderately 601 correlated between dust collected via the two sampling methods (Allen et al., 2008; Stapleton 602 et al., 2008). Another study in Sweden examined BFRs in RCD (collected using cellulose filters 603 inserted in the vacuum cleaner hose) and HHVD samples, observing that the median 604 concentrations of PBDEs in RCD were 2-3 times higher than those in HHVD and significantly 605 (P<0.05) higher for Σ PentaBDE, Σ OctaBDE, and Σ DecaBDE. In contrast, median 606 concentrations of HBCDD were 10 times higher in HHVD than RCD. Statistically significant 607 correlations were found for Octa- and Deca-BDE between the two sampling methods, while no 608 correlation was found for Σ PentaBDE (Björklund et al., 2012). In this study, RCD samples 609 were collected from elevated surfaces only. Assuming that the HHVD samples were collected 610

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

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

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647 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 648 649 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, 650 651 assessments based on FR concentrations detected in HHVD rather than RCD will underestimate exposure for lower brominated BFRs, BEH-TEBP, TPHP, and chlorinated OPEs 652 by factors ranging between 1.1 and 5.5. This indicates that the HHVD method is a less 653 acceptable method of assessing exposure to such pollutants. 654

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656 **3.4 Temporal and seasonal variability**

Several studies have investigated temporal variability in concentrations of FRs in indoor dust, 657 including PBDEs (Allen et al., 2008; Harrad et al., 2008; Batterman et al. 2009; Yu et al., 2012; 658 Muenhor and Harrad 2012; Al-Omran and Harrad, 2018; Niu et al., 2018), NBFRs (Cao et al., 659 2014b, Al-Omran and Harrad 2018, Niu et al., 2019), HBCDD (Harrad et al., 2009; Cao et al., 660 2015), and OPEs (Cao et al., 2014b; Ortiz Carrizales, 2018). While some of these studies 661 presented their data on a month-to-month basis, others presented it by season (spring, summer, 662 autumn, and winter) or classified into colder and warmer periods. However, results are 663 664 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 665 666 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. 667 The relative standard deviation of concentrations (RSD) of individual FRs and 668 maximum:minimum concentration ratios were applied to measure the extent of variability over 669 670 the studied time period.

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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

trend in PBDE concentrations in dust samples collected in two different seasons from homes 678 679 and garages, which is in line with the temporal variation observed in several UK buildings. In the UK, four studies (Harrad et al., 2008; 2009; Muenhor and Harrad 2012; Al-Omran and 680 681 Harrad, 2018) reported substantial variability in BFR concentrations in floor dust collected from several microenvironments at monthly intervals over 8-10 months. For example, RSDs 682 683 of concentrations of ∑tri-hexa-BDEs (52-156%), BDE-209 (58-166%), and HBCDDs (27-190%) exceeded those observed for within-room spatial variability (Harrad et al., 2008a; 2009). 684 685 This is likely attributable to changes over time in room contents with respect to potential sources of FRs. For instance, when a new rug was introduced, concentrations of BDE-209 rose 686 substantially from 3152 to 19802 ng/g (Al-Omran and Harrad, 2018). 687

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As SVOCs, it is expected that temperature changes between the four seasons or between colder 689 and warmer months will exert an influence on the transfer of FRs from sources to indoor dust 690 691 via volatilisation/deposition. Yu et al., (2012) reported that average concentrations of PBDEs in indoor dust from China followed the order summer> winter> spring> autumn (Yu et al., 692 2012). This is consistent with recent studies (Niu et al., 2018; 2019) comparing concentrations 693 of BFRs in different seasons, that found concentrations of Σ_7 NBFRs were significantly (P < 694 0.05) higher in August than in other months. The geometric mean concentrations followed the 695 order: summer> winter> autumn> spring, indicating that elevated temperatures in summer 696 697 increased emissions of FRs from their sources via volatilisation. However, these seasonal variations conflict to some extent with the results reported by Cao et al. (2014b) for dust 698 samples collected from three offices for 9 consecutive months (for which there was no apparent 699 addition or removal of FR containing products). While Σ PBDE and Σ NBFR concentrations 700 remained relatively stable throughout, PFR concentrations changed in contrast to the trend of 701 temperature, with higher concentrations in later winter and early spring and lower levels in 702 summer. This temporal variation was attributed to the greater volatility of the OPEs studied 703 relative to the BFRs (Cao et al., 2014b). Regarding OPEs, similar observations about temporal 704 variability to those reported by Cao et al. (2014b) were reported in a study of house dust 705 706 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 early winter. Combined, these two studies revealed concentrations of OPEs in indoor dust to 708 709 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 arange of microenvironments.

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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.

721 **4. Conclusions**

Differences in FR concentrations and consequent human exposure assessments were observed 722 723 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 724 floor dust and elevated surface dust. Such differences are attributable to a variety of factors, 725 including the presence of sources on the surfaces sampled (e.g. flooring material), and the 726 higher proportion of fine particles found on elevated surfaces. In the majority of studies, FR 727 concentrations in elevated surface dust exceed those detected in floor dust from the same room. 728 729 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, 730 and TPHP. Meanwhile, sampling dust from one specific area of the floor within a room may 731 not reflect contamination of the entire room due to spatial variations in distance from putative 732 733 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 734 room may not be entirely representative for all people living or working in that building. While 735 differences are observed in the distribution of FRs between different dust particle size fractions; 736 these differences did not follow a definitive trend. Specifically, while in some studies, FR 737 concentrations increased in line with increases in particle specific surface area associated with 738 decreasing particle size; this appears to be overridden in situations where the principal source 739 of FRs in a dust sample is abrasion of source material. In such situations, FR concentrations 740 are highest in the size range of the abraded particles. With respect to the sampling method 741 deployed, concentrations of lower brominated BFRs, BEH-TEBP, TPHP, and chlorinated 742 OPEs are generally reported to be higher in RCD than in HHVD, implying that exposure 743

assessments based on HHVD will be underestimated. Moreover, temporal variations in FR 744 745 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 746 747 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 748 749 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 750 751 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 752 increased ventilation. 753

In conclusion, the above factors should be taken into account when designing a study to assess 754 human exposure to FRs and OPEs via ingestion of indoor dust. For example, the choice of 755 whether to collect and analyse ESD or FD may be influenced by whether exposure is being 756 estimated for young children in daycare centres or adults in offices. In the former case, FD is 757 recommended, while ESD is advised in the latter scenario. Moreover, the observed within-758 room and within-building variability in FR and OPE concentrations suggests that rather than 759 sampling all surfaces in a room, dust should be collected and analysed only from the most-760 frequented areas of that room. While ideally multiple rooms in a dwelling would be sampled 761 762 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 763 for a substantial proportion of the day. Furthermore, given the temporal variations in 764 concentrations of FRs and OPEs in dust observed in the same rooms in some studies, exposure 765 766 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 767 means that for the purposes of assessing exposure via dust ingestion, dust analysed should be 768 confined to the size fraction that adheres to hands. 769

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778 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

780 Lohmann, 2018; 2019), China (Bu et al., 2019; Wu et al., 2016) and Japan (Araki et al.,

781 **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	-			
ТРНР	1.3 (1.7)	2.5 (3.6)	0.3	0.8	na	1.2			
ТМРР	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

783 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).

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	ТРНР	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

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



797 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)

800 List of ABBREVIATIONS

- 801 ANOVA, Analysis of variance
- 802 BDE-153, (2,2',4,4',5,5'-hexabromodiphenyl ether
- 803 BDE-154, (2,2',4,4',5,6'-hexabromodiphenyl ether
- 804 BDE-183, (2,2',3,4,4',5',6-heptabromodiphenyl ether
- 805 BDE-209, (2,2',3,3',4,4',5,5',6,6'-decabromodiphenyl ether
- 806 BDE-47, (2,2',4,4'-tetrabromodiphenyl ether
- 807 BDE-99, (2,2',4,4',5-pentabromodiphenyl ether
- 808 BEH-TEBP, Bis(2-ethyl-1-hexyl)tetrabromophthalate
- 809 BFR, brominated flame retardant
- 810 BTBPE, 1,2-bis(2,4,6-tribromophenoxy)ethane
- 811 DBDPE, Decabromodiphenylethane
- 812 Deca-BDE, decabromodiphenyl ether
- 813 EHDPP, 2-Ethylhexyl diphenyl phosphate
- 814 EH-TBB, 2-ethylhexyl-2,3,4,5-tetrabromobenzoate
- 815 FRs, flame retardants
- 816 HBCDD, hexabromocyclododecane
- 817 K_{OA}, octanol-air partition coefficient
- 818 NBFRs, "Novel" brominated flame retardants
- 819 OC, organic carbon contents content
- 820 Octa-BDE, octabromodiphenyl ethers
- 821 Pa, Pascal
- 822 PBDE, polybrominated diphenyl ether
- 823 Penta-BDE, pentabromodiphenyl ether
- 824 OPEs, organophosphate esters
- 825 POPs, persistent organic pollutants
- 826 RSD, relative standard deviation
- 827 SRM2585 and 2584, dust standard reference material
- 828 SVOCs, semi-volatile organic compounds
- 829 TBOEP, tris (2–butoxyethyl) phosphate
- 830 TCEP, tri(2-chloroethyl) phosphate
- 831 TCIPP, Tris(2-chloroisopropyl) phosphate

- 832 TDCIPP, (Tris (1,3-dichloro-2-propyl) phosphate
- 833 TMPP, Tris(4-methylphenyl) phosphate
- 834 TNBP, tri-n-butyl phosphate
- 835 TPHP, triphenyl phosphate
- 836 UNEP, United Nations Environment Programme
- 837 VP, vapour pressure
- 838 WHO, World Health Organization
- 839

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