

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

56

57 **Keywords**

58 BFRs, OPEs, indoor dust, exposure assessment

59

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 µm

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88 **1. Introduction**

89 In recent decades, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane
90 (HBCDD) have been the most extensively used additive brominated flame retardants (BFRs)
91 in consumer products such as polyurethane foam, electrical and electronic equipment, textiles,
92 and construction materials. A large number of studies have indicated that PBDEs and HBCDD
93 are persistent and bioaccumulative toxicants, which adversely affect human health (WHO,
94 2003; van der Ven et al., 2009; Noyes et al. 2010). Health concerns of PBDEs and HBCDD
95 exposure include endocrine disruption (Johnson et al., 2013), impaired fertility (European
96 Commission, 2012), neurotoxic effects (Schechter et al., 2012), and possible carcinogenicity
97 (Li et al., 2014). As a consequence, the manufacture and new use of commercial mixtures of
98 PBDEs (Penta-, Octa-, and Deca-BDE) and HBCDD have been subject to restrictions and
99 listed under the Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP, 2008;
100 2012; 2019). Such actions have resulted in increased production and use of alternative FRs
101 such as “novel” brominated flame retardants (NBFRs). NBFRs as relating to brominated flame
102 retardants are defined here as those new to the market or recently observed in the environment
103 such as BEH-TEBP (Bis(2-ethyl-1-hexyl) tetrabromophthalate), DBDPE
104 (decabromodiphenylethane). In addition, organophosphate ester (OPEs) compounds which are
105 extensively used as flame retardants and plasticizers. (Ven der Veen and de Boer, 2012;
106 Greaves and Letcher, 2017; Blum et al., 2019). It is, however, expected that PBDE
107 formulations may maintain a presence in new products due to the use of recycled materials,
108 with old PBDE-containing products still in use constituting a further on-going emission source
109 (USEPA, 2010; Pivnenko et al., 2017). The global production of FRs was estimated at 2.8
110 million tonnes in 2018, and to be growing by 4.6% per year (Yasin et al, 2016). In 2019, BFRs
111 and OPEs represented 17% and 18% respectively of the global production of flame retardants,
112 with Asia the largest market, followed by North America, Europe, Middle East, and Latin
113 America respectively (IHS, 2020; Market Research Future, 2018).

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

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

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

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

158 **2. Methodology**

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

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

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

197 **3. Results and discussion**

198 **3.1 Within-room spatial variability in BFR and PFR concentrations in indoor dust**

199 **3.1.1 Within-room spatial variability between floor and elevated surface dust**

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

205 **3.1.1.1 Comparison of FR concentrations in ESD and FD**

206 For BFRs, out of six studies in Norway (Cequier et al., 2014), Iraq (Al-Omran and Harrad,
207 2016a), USA (Allgood et al., 2017), UK (Al-Omran and Harrad 2018), Egypt (Khairy and
208 Lohmann, 2018), and China (Bu et al., 2019); median concentrations of most PBDEs and
209 NBFRs in ESD from the USA, UK, Iraq, and Egypt exceeded those in floor dust samples taken
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
213 target PBDEs and NBFRs ranged between 1.1- 3.7 (Cequier et al., 2014). This may be due to
214 the different particle size of the two dust sample categories collected in this Norwegian study.
215 Specifically, FD was collected using a nozzle with a polyethylene grid (1–3 mm pore size)
216 while ESD was collected without the grid. This suggests that the floor dust collected may have

217 contained a greater proportion of fine particles preferentially contaminated with lower
218 brominated PBDEs. Similarly, in day care centres in China, median concentrations in ESD
219 exceeded those in FD for only 3 out of 7 BFRs (Bu et al., 2019). For OPEs, median ESD:FD
220 ratios for TPHP, TCEP, TCIPP, and TDCIPP from Japan (Araki et al., 2014; Tajima et al.,
221 2014), USA (Allgood et al., 2017), UK (Ortiz Carrizales, 2018), Norway (Cequier et al., 2014;
222 Xu et al., 2016), Egypt (Khairy and Lohmann, 2019), and China (Wu et al., 2016) ranged
223 between 1.1- 3.9. Notable exceptions were TDCIPP and TPHP in China (Wu et al., 2016), and
224 TPHP in Egypt (Khairy and Lohmann, 2019), for which median ratios were <1. The reason
225 for this finding may be due to the small sample size (n=9) and low concentrations of OPEs in
226 the day care and apartment dust studied in China and Egypt respectively. In contrast to TPHP
227 and chlorinated OPEs, concentrations of TBOEP in FD exceeded those in ESD with median
228 ESD:FD ratios <1 (Cequier et al., 2014; Xu et al., 2016; Araki et al., 2014; Tajima et al., 2014;
229 Khairy and Lohmann, 2019). This may be due to the wide use of TBOEP in flooring and floor
230 polishing materials (Kajiwara et al., 2011; van der Veen and de Boer, 2012). Table 1 shows
231 the median ratios of the concentrations of FRs detected in ESD and FD collected from the
232 same rooms for various studies. Figure 1 shows the median concentrations of FRs present in
233 ESD and FD samples collected in the same studies.

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

249 **3.1.1.2 Relationship between concentrations of FRs in ESD and FD**

250 Pearson correlation analysis revealed the concentrations of several FRs in ESD samples were
251 significantly ($p < 0.05$) correlated with those in FD samples from the same room. For BFRs,
252 significant positive linear correlation was found for BEH-TEBP and EH-TBB in Norway
253 (calculated from the raw data of Cequier et al., 2014), Iraq (Al-Omran and Harrad, 2016a),
254 USA (calculated from the raw data of Allgood et al., 2017) and UK (Al-Omran and Harrad,
255 2018) with respective correlation coefficient values of 0.673, 0.803, 0.875, and 0.952 for BEH-
256 TEBP, and 0.686, 0.656, 0.827, and 0.639 for EH-TBB respectively. Concentrations of BDE-
257 47 (in three studies), BDE-99, BDE-209 and DBDPE (in two studies) in ESD were
258 significantly correlated with those in FD. In addition, chlorinated OPEs showed significant
259 positive linear correlation between ESD and FD for TCEP in USA (calculated from the raw
260 data of Allgood et al., 2017), UK (calculated from the raw data of Ortiz Carrizales, 2018), and
261 China (Wu et al., 2016), as well as for TCIPP in Japan (Tajima et al., 2014) and China (Wu et
262 al., 2016). This suggests that the sources of these contaminants in ESD and FD are similar.
263 Tables S4 and S5 show the results of Pearson correlation analyses of concentrations of BFRs
264 and OPEs in paired samples of ESD and FD reported in various studies.

265 **3.1.1.3 Causes of variability in FR concentrations between ESD and FD**

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

281 Building materials are an important source of FRs, especially floor materials (Kanazawa et al.,
282 2010). Thus, the wide use of TBOEP as a plasticiser in floor wax and PVC floor coverings
283 (Kajiwara et al., 2011; van der Veen and de Boer, 2012) likely explains the higher
284 concentrations observed in floor as opposed to elevated surface dust. In contrast, wooden-
285 structure houses displayed lower levels of TCIPP, TCEP, and TPHP in FD than other types of
286 houses, such as those with reinforced concrete floors (Araki et al., 2014; Tajima et al., 2014).
287 TPHP is commonly used in combination with halogenated OPEs added to polyurethane foam
288 as well as sealing and heat-insulating materials (van der Veen and de Boer, 2012; Tajima et
289 al., 2014). As shown in Figure 1, Table 1 and Table S3, the distribution profile of TPHP and
290 chlorinated OPEs between ESD and FD are similar. Specifically, except for TCEP in Norway
291 (for which the median ESD:FD ratio = 1); median ESD:FD ratios of TPHP (1.7, 2.5 and 1.2)
292 were consistent (>1) with those of TCEP (1.0, 1.4 and 2.4), TCIPP (2.6, 3.0 and 1.5) and
293 TDCIPP (2.8, 3.9 and 1.3) from the same dust sample in Norway, Japan and UK respectively.

294 Contaminated fine particles originating from the floor materials may be suspended into indoor
295 air before settling on elevated surfaces (Wu et al., 2016). This may explain the presence of
296 lower vapour pressure compounds in ESD even where floor materials are the source. Also, as
297 finer particles are more atmospherically mobile, ESD likely contains a greater proportion of
298 fine particles containing chemicals that have accumulated via volatilisation and subsequent
299 deposition (Rauert and Harrad, 2015). Moreover, where ESD samples are collected from
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
302 surfaces may be less frequently cleaned than floors, this provides another explanation for FR
303 concentrations in ESD that exceed those in FD (Tajima et al, 2014; Rauert et al., 2014b).
304 Furthermore, outdoor particles such as sand and soil tracked indoors with footwear are more
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,
308 which may preferentially dilute ESD. Al-Omran and Harrad (2016a) examined the hypotheses
309 that differences in concentrations of organic carbon (OC) and particle size distribution between
310 ESD and FD contribute to observations of significant differences in concentrations of FRs
311 between such dust types. The authors reported significantly ($p<0.05$) higher proportions of fine
312 particles ($<125 \mu\text{m}$) in ESD samples and no substantial influence of OC on the observed

313 differences in concentrations between ESD and FD. To our knowledge, this is the only study
314 addressing 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**

316 As discussed above, concentrations of several FRs in ESD samples collected from tables,
317 shelves, chairs, etc. exceeded significantly those in FD. However, the majority of assessments
318 of human exposure to FRs via indoor dust ingestion have to date focused on collection and
319 analysis of floor dust samples. This is likely appropriate for young children and toddlers as
320 they are far more likely to have contact with FD when playing close to the floor; however,
321 adults, adolescents, and older children are more likely to come into contact with ESD. Coupled
322 with observations of higher FR concentrations in ESD, this indicates that human exposure
323 assessments based on FD alone will likely underestimate exposure via dust ingestion for adults.
324 As shown in Tables 1, S2 and S3, and Figure 1, the extent of underestimation is greatest for
325 BDE-99, BEH-TEBP, BDE-209, chlorinated OPEs, and TPHP. For example, such
326 underestimates for the adult population can be substantial e.g., reaching as high as factors of
327 5.0 and 19 for “typical” exposure rate of BDE-183 in USA (Allgood et al., 2017) and Egypt
328 (Khairy and Lohmann, 2018) respectively.

329

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

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

346 Other studies in the UK tested within-room spatial variability in concentrations of BFRs
347 (Muenhor and Harrad, 2012; Al-Omran and Harrad 2018) and OPEs (Ortiz Carrizales, 2018)
348 in floor dust samples (2-5) taken from different 1 m² areas within the same rooms (n=5-9).
349 Significant differences (p <0.05) were observed in concentrations of Σ_7 tri-hepta-BDEs and
350 Σ NBFRs in different areas of the same room for three out of nine rooms studied, but not for
351 BDE-209 (calculated from the raw data of Al-Omran and Harrad 2018). For OPEs, significant
352 differences (p <0.05) were also found in two out of four rooms for TPHP and EHDPP and in
353 one room for both TNBP and TDCIPP. No significant differences were observed for TCIPP
354 and TCEP (Ortiz Carrizales, 2018). Concentrations of TCIPP and TCEP were strongly
355 associated with floor materials such as tatami, tiles, and wall-to-wall carpet (Tajima et al.,
356 2014). However, TCIPP displayed strong spatial variability in dust samples from the Czech
357 Republic. Concentrations of TCIPP decreased with increasing distance from a sofa, implying
358 the use of TCIPP in the foam filling of the sofa concerned (Jilkova et al., 2018).

359 Overall, the evidence suggests higher concentrations of FRs accumulate in dust samples located
360 closest to potential emission sources, such as sofas, chairs, TVs, and laptops etc. Harrad et al.,
361 (2009) reported that concentrations of HBCDDs declined with increasing distance from a TV.
362 In other words, within-room spatial variability is likely driven by varying distances from
363 potential emission sources. Greater spatial variability was observed in large rooms and between
364 bare floor and carpeted floor areas within the same room (Al-Omran and Harrad, 2018). In
365 addition, in one office (1450 m²) from China, higher concentrations of PBDEs were detected
366 in some areas, compared to those in areas close to the entrance, probably due to the poor
367 ventilation away from the entrance (Li et al., 2015). Table S7 reports the RSD values for BFRs
368 in different microenvironments, while Figure S2 compares median concentrations of selected
369 BFRs and OPEs in floor dust from two different areas of the same room located in the UK.

370 These results indicate that appreciable variation in FR contamination can be found, depending
371 on where in a given room, floor dust samples are taken. This suggests that for accurate
372 estimation of exposure, such “spot” samples may not be fully representative of the
373 contamination of the room as a whole. For example, if in a given room (assuming the two floor
374 areas are F1 and F2), the concentration of Σ_7 tri-hepta-BDEs in F1 exceeds substantially that in
375 dust samples from F2 with ratio of F1:F2=61:4 for one sampling event; in this room, the
376 exposure estimate will vary by a factor of just over 15 depending on the area samples (Al-
377 Omran and Harrad, 2018). Overall, while the substantial and sometimes significant variation
378 in FR concentrations in floor dust from different areas of the same room, may imply that

379 sampling of the entire floor area is the best approach to estimating human exposure; the fact
380 that doing so includes dust from rarely-frequented parts of the room, led Al-Omran and Harrad
381 (2018) to conclude that floor dust samples should be taken from the most-frequented parts of
382 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
386 spatial variability between the living room and bedroom (Allen et al., 2008; Stapleton et al.,
387 2008; Muenhor and Harrad 2012; Venier et al., 2016; Al-Omran and Harrad, 2017; 2018; Ortiz
388 Carrizales, 2018). To our knowledge, only two studies have investigated within-home spatial
389 variability between the kitchen and living area/bedroom (Kuang et al., 2016; Ortiz Carrizales,
390 2018), with two other studies reporting spatial variability between different rooms in academic
391 microenvironments (Dodson et al., 2017; Jilkova et al. 2018).

392 In two studies from the USA (Allen et al., 2008; Stapleton et al., 2008), concentrations of Penta-
393 BDE, Deca-BDE, HBCDD, EH-TBB, and BEH-TEBP were significantly ($p < 0.05$) higher in
394 the main living area compared to the bedroom. No significant differences were apparent for
395 BTBPE and DBDPE (Stapleton et al., 2008). In UK house dust samples, median concentrations
396 of individual PBDEs and NBFrs in the living room were noticeably higher than those in the
397 corresponding bedrooms (statistically significant for BDE-183) (Al-Omran and Harrad, 2017).
398 Kuang et al., (2016) also reported no significant differences ($p > 0.05$) between BFR
399 concentrations in the living room and bedroom in UK dust samples. In contrast, another study
400 investigating PBDE and NBFr concentrations in living rooms and bedrooms in the same
401 homes in three countries (USA, Canada, and Czech Republic) reported different patterns
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
404 rooms and were significantly ($p < 0.05$) higher for BDE-153, BTBPE, and BDE-209 in USA,
405 Canada, and the Czech Republic respectively (calculated from the raw data of Venier et al.,
406 2016). In Australia, except for BDE-154 (for which concentrations were significantly higher
407 in the living room), no significant differences were found between concentrations of PBDEs
408 and NBFrs in living rooms and bedrooms from the same homes (calculated from the raw data
409 of McGrath et al., 2018). Figure 2 illustrates the distribution profiles of the most common
410 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 (Jílková 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 Jílková et al.,
442 2018). From another academic microenvironment (2 college campuses in the USA), Dodson et

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
447 microenvironments in the same building. This is considered a reflection of variability between
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
450 living rooms compared to bedrooms may be attributed to the presence in the former, of specific
451 putative source items such as televisions and couches (Allen et al., 2008). Moreover, higher
452 concentrations of TCEP and TPHP in student dormitory rooms, compared to common areas,
453 may be related to the electronics and furnishings introduced to dormitories by students (Dodson
454 et al., 2017). Similar explanations were proposed by other studies (Stapleton et al., 2008;
455 Muenhor and Harrad, 2012; Al-Omran and Harrad, 2017; 2018; Jilkova et al., 2018; McGrath
456 et al., 2018), which is supported by the lack of strong correlations between FR concentrations
457 in rooms within the same building, as indicated by Table S8 that shows Pearson's correlation
458 coefficient (r) values between different rooms within the same building in various studies.
459 Alternatively, the observed lack of significant differences in concentrations of FRs between
460 bedrooms and living rooms in the same home suggests that emission sources are similar in both
461 rooms (Kuang et al., (2016)). However, a similar lack of significant differences between rooms
462 in the same house was attributed by Venier et al., (2016) to air exchange and concomitant
463 transfer of FRs between rooms. This latter explanation likely applies when the main mechanism
464 via which FRs enter dust in the building concerned, is volatilisation from sources with
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.

467
468 Between-room differences in source types and numbers that lead to substantial within-building
469 spatial variability means that exposure estimates based on measuring BFRs in dust from one
470 room may not be entirely representative of exposure via dust ingestion to *all* individuals
471 frequenting a given building. For example, in one home, concentrations of BEH-TEBP in the
472 bedroom (average \pm SD = 3992 ± 1906 ng/g) exceeded those in the living room (average \pm SD
473 = 1811 ± 1498 ng/g) (Al-Omran and Harrad 2018). This implies that exposure assessment in
474 that home based on living room dust only may result in a substantial underestimate.

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

476 Dust from indoor microenvironments includes a wide range of particle sizes ranging from <
477 2.5 μm to over 2000 μm in diameter. Particles > 30 μm may be classified as settled particles,
478 which include “skin flakes, fragments of hair, microorganisms, such as fungal spores and
479 pollen, food crumbs, abrasion of textiles and fittings, sand, loam, clay, and soot” (Morawska,
480 2004). Studies investigating human exposure to FRs via indoor dust ingestion have analysed a
481 wide range of dust particle sizes, such as <2000 μm (Gevao et al., 2006), <500 μm (Abdallah
482 and Covaci, 2014), <250 μm (Al-Omran and Harrad 2017), <150 μm (Shoeib et al., 2012),
483 <100 μm (Kang et al., 2011), and < 63 μm (Kopp et al., 2012). However, for assessment of
484 exposure via dust ingestion, it is reasonable to assume that only those particles adhering to
485 human hands and that can subsequently be orally ingested via hand-to-mouth contact and/or
486 release FRs via dermal uptake are pertinent. The most recent recommendation by the USEPA
487 is that adherence to hands is greatest for dust particles <150 μm diameter (USEPA, 2016),
488 which is smaller than their previous recommendation to focus on particles <250 μm diameter
489 (USEPA, 2008). Moreover, increased FR bioaccessibility via dust ingestion was observed with
490 decreasing particle size (Yu et al., 2013; Fang and Stapleton, 2014). This section highlights the
491 extent to which analysing different dust fractions influences FR concentrations and consequent
492 exposure assessments.

493
494 While in some studies, FR concentrations increased with decreasing dust particle size, other
495 studies revealed that specific pollutants accumulated in specific particle size fractions and
496 varied between different microenvironments. The first investigation of FRs in different particle
497 size fractions of indoor dust was reported by Wei et al., (2009) in the USA, concluding that
498 while in car dust, PBDE concentrations were inversely related to particle size, they were largely
499 comparable in four dust fractions (250- 420 μm , 150-250 μm , 75-150 μm , and < 75 μm) for
500 home dust. A comparison of dust from different microenvironments (offices, hotels,
501 kindergartens, and dormitories), showed that concentrations of DBDPE and HBCDD in hotel
502 dust were highest in fine particles (50-74 μm and >50 μm respectively), while in office dust
503 concentrations were highest in medium particles (100–200 μm) (Cao et al., 2014a; 2015).
504 These findings are consistent with other reported data from three different microenvironments
505 in China. Among four particle size fractions, BDE-209 and BEH-TEBP accumulated most
506 strongly in both fine (43-63 μm) and finest (<43 μm) particles of office dust, while they were
507 present at higher concentrations in coarse particles (150-200 μm) in public microenvironments
508 (PMEs) (He et al., 2018). In addition, differences in distribution pattern of FRs between
509 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 \sum 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

544 μm) of dust samples from three different microenvironments, the highest concentrations of
545 TCEP ($\text{VP}=5.2 \times 10^{-2}$ Pa) were found in the finest ($<43 \mu\text{m}$) particles with maximum:minimum
546 ratios of 2.2, 1.4 and 1.3 in office, public microenvironment and car dust samples respectively.
547 However, concentrations of TCIPP ($\text{VP}=7.4 \times 10^{-3}$ Pa), close to that of TCEP) were higher in
548 the coarse fraction (100-200 μm). In contrast, the concentrations of DBDPE and BDE-209
549 ($\text{VP}=2.6 \times 10^{-11}$ and 9.3×10^{-9} Pa respectively), were higher in the finest particles of the same
550 samples (He et al., 2018; Cao et al., 2013). These findings are consistent with other studies,
551 e.g. while concentrations of the more volatile tri-hepta BDEs were higher in medium sized
552 (125–212 μm) particles in a pooled sample of dust from air conditioner filters in dining halls,
553 the concentration of BDE-209 was higher in the fine particles ($<43\text{-}63 \mu\text{m}$) (Yu et al., 2013).
554 The above examples suggest that while in many scenarios, the predominant source-to-dust
555 transfer mechanism is volatilisation followed by deposition to finer particles with greater
556 specific surface area; in some situations, FRs in indoor dust particles might arise predominantly
557 from a source weathering or abrasion process rather than volatilisation/deposition. Thus, in
558 such situations, irrespective of the vapour pressure, the size of the abraded, FR-rich particles is
559 likely to determine the fraction with the highest FR concentration. Overall, regardless of the
560 predominant source-to-dust transfer mechanism, our meta-analysis suggests that significant
561 differences do exist between concentrations of FRs in different particle size fractions. This
562 supports focusing on analysis of those particle sizes shown to adhere to hands as the most
563 relevant for human exposure via unintentional dust ingestion. Current USEPA
564 recommendations are that these are particles $<150 \mu\text{m}$ diameter (USEPA, 2016).

565

566 **3.3 Evaluation of dust sampling approaches**

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

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

580

581 The most widely used sampling technique is vacuuming, most commonly with conventional
582 household vacuum cleaners as inexpensive alternatives to the standard HVS3 (High Volume
583 Small Surface Sampler) developed by USEPA (Colt et al., 2008). Vacuum cleaner methods
584 may be split into two categories; (a) researcher collected dust (RCD), where dust is collected
585 by researchers using specific accessories and standardised methods; and (b) householder
586 vacuum dust (HHVD), where householders provide researchers with their vacuum cleaner
587 contents (Shen et al., 2015; Ali et al., 2016). The most common accessories that have been used
588 for RCD are nylon “socks” (Harrad et al., 2016), filters (Newton et al., 2015), and Soxhlet
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
591 ingestion, all of the above approaches have associated uncertainties. This section discusses to
592 what extent the sampling method used influences FR concentrations in the dust collected and
593 the consequent exposure assessment.

594

595 To date, a small number of studies have compared concentrations of FRs in RCD and HHVD.
596 Two early studies in the USA found that concentrations of Penta-BDE and Deca-BDE
597 congeners in RCD (obtained using cellulose extraction thimbles inserted in the vacuum cleaner
598 sampling hose) collected from living rooms and those of Penta-BDE congeners in bedrooms
599 exceeded significantly ($p < 0.05$) those in HHVD. In contrast, geometric mean concentrations
600 in HHVD were 2 times higher for HBCDD and comparable for BDE-209 compared to RCD
601 collected in bedrooms. In general, concentrations of PBDEs were weakly to moderately
602 correlated between dust collected via the two sampling methods (Allen et al., 2008; Stapleton
603 et al., 2008). Another study in Sweden examined BFRs in RCD (collected using cellulose filters
604 inserted in the vacuum cleaner hose) and HHVD samples, observing that the median
605 concentrations of PBDEs in RCD were 2-3 times higher than those in HHVD and significantly
606 ($P < 0.05$) higher for \sum PentaBDE, \sum OctaBDE, and \sum DecaBDE. In contrast, median
607 concentrations of HBCDD were 10 times higher in HHVD than RCD. Statistically significant
608 correlations were found for Octa- and Deca-BDE between the two sampling methods, while no
609 correlation was found for \sum PentaBDE (Björklund et al., 2012). In this study, RCD samples
610 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 Σ 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 Σ 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 $<150 \mu\text{m}$, 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 Σ 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

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

646

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

655

656 **3.4 Temporal and seasonal variability**

657 Several studies have investigated temporal variability in concentrations of FRs in indoor dust,
658 including PBDEs (Allen et al., 2008; Harrad et al., 2008; Batterman et al. 2009; Yu et al., 2012;
659 Muenhor and Harrad 2012; Al-Omran and Harrad, 2018; Niu et al., 2018), NBFRs (Cao et al.,
660 2014b, Al-Omran and Harrad 2018, Niu et al., 2019), HBCDD (Harrad et al., 2009; Cao et al.,
661 2015), and OPEs (Cao et al., 2014b; Ortiz Carrizales, 2018). While some of these studies
662 presented their data on a month-to-month basis, others presented it by season (spring, summer,
663 autumn, and winter) or classified into colder and warmer periods. However, results are
664 insufficient to elucidate seasonal trends of FRs when based on discontinuous (i.e. incomplete
665 calendar year) campaigns, although these results can be used for attribution of emission
666 source(s). This section highlights the extent to which assessments of human exposure to BFRs
667 and OPEs via indoor dust ingestion may be influenced by temporal and seasonal variability.
668 The relative standard deviation of concentrations (RSD) of individual FRs and
669 maximum:minimum concentration ratios were applied to measure the extent of variability over
670 the studied time period.

671

672 The first investigation of temporal variability in concentrations of PBDEs in dust samples from
673 US homes, revealed no significant differences in samples taken from the same rooms 8 months
674 apart, likely because home furnishings changed little over that time (Allen et al., 2008).
675 However, for no obvious reason, only those congeners associated with Octa-BDE in this study,
676 were significantly higher in dust samples collected in autumn compared with samples collected
677 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 Σ 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_7 NBFRs 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 Σ PBDE and Σ NBFR 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

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

713

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

721 **4. Conclusions**

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

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

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

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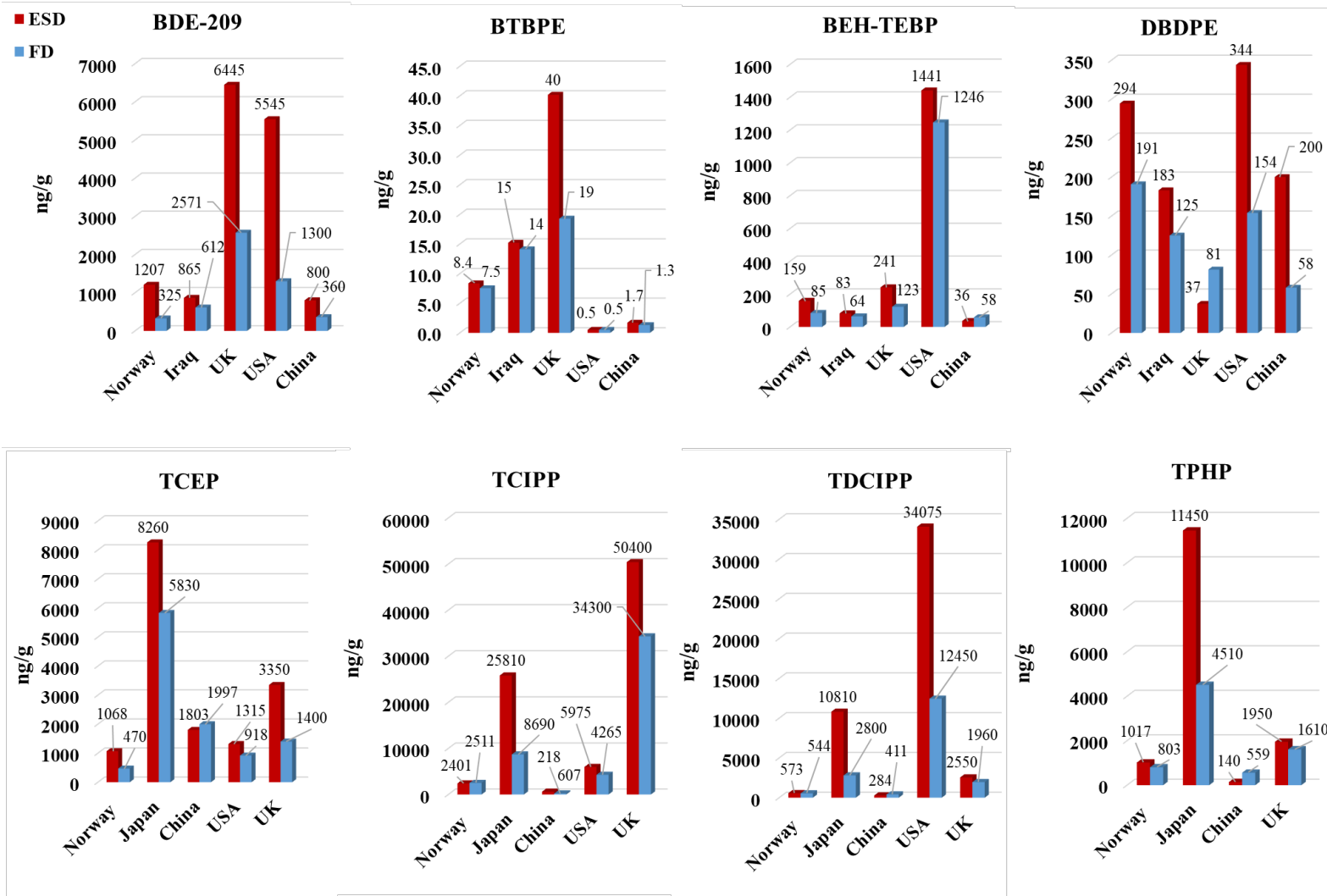
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777 **Table 1. Median ESD:FD ratios of BFRs and OPEs in indoor dust from Norway (Cequier**
778 **et al., 2014; Xu et al., 2016*), Iraq (Al-Omran and Harrad, 2016a), USA (Allgood et al.,**
779 **2017), UK (Al-Omran and Harrad 2018; Ortiz Carrizales, 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	-
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

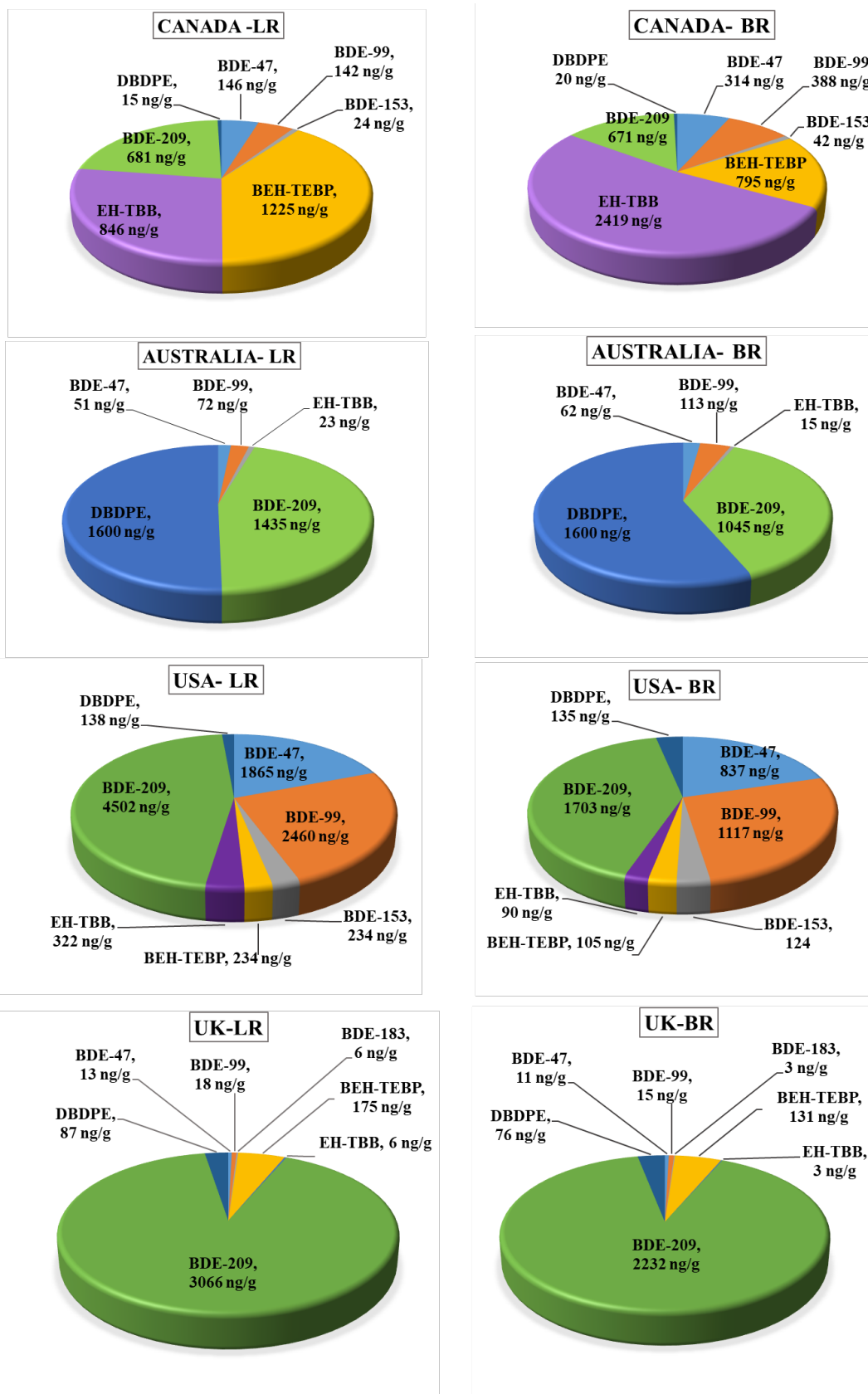
782 na = not applied, - = not detected or detection frequencies <50%, bold values= median ESD:FD
783 ratios >1, (*) = Xu et al., 2016, (**) =Tajima et al., 2014

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785

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

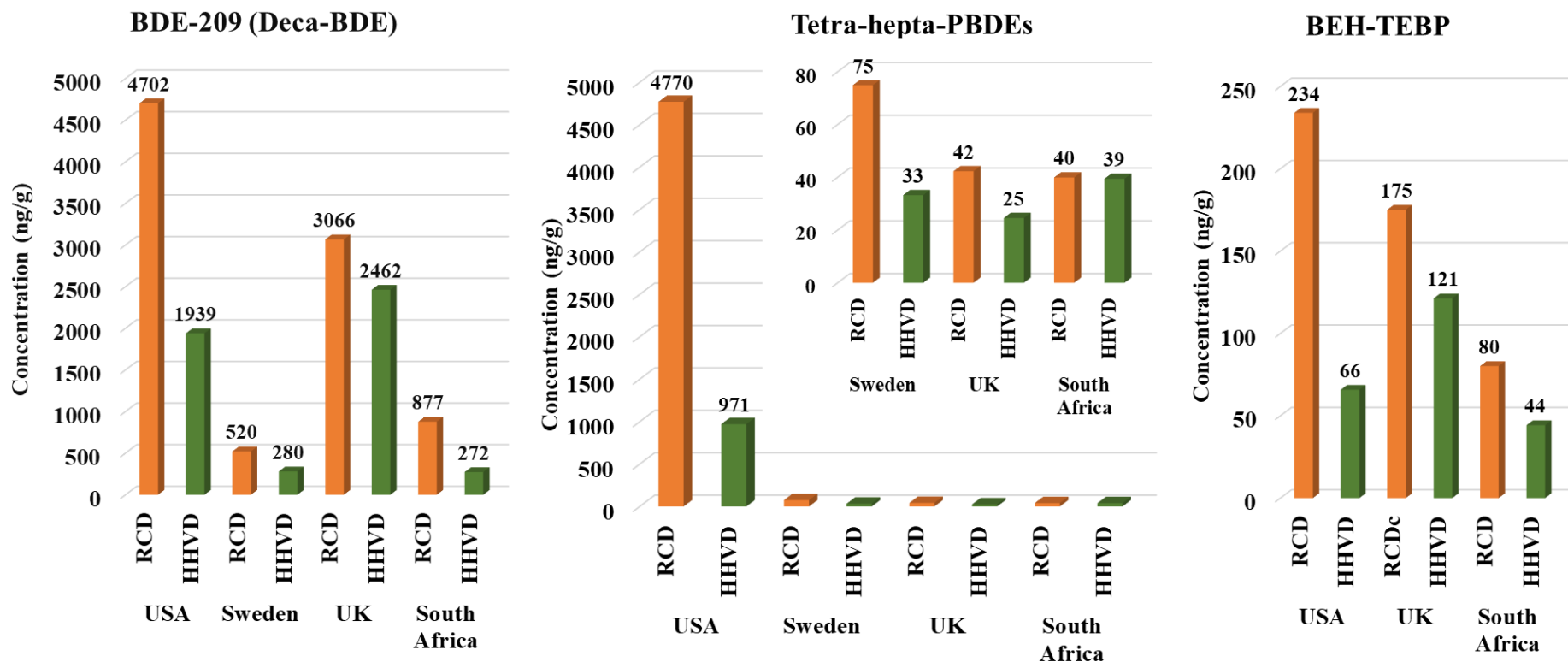


789

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

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)

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

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