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1 **Distribution pattern of legacy and “novel” brominated flame**
2 **retardants in different particle size fractions of indoor dust in**
3 **Birmingham, United Kingdom**

4
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9
10 **ABSTRACT**

11 This study investigates the particle size distribution of eight polybrominated diphenyl ethers
12 (PBDEs) and five “novel” brominated flame retardants (NBFRs) in settled house dust.
13 Elevated surface dust (ESD) and floor dust (FD) were collected from 5 homes in
14 Birmingham, UK, yielding a total of 10 samples. Each sample was fractionated into three
15 different particle sizes: 125-250 μm (P1), 63-125 μm (P2) and 25-63 μm (P3). Non-
16 fractionated bulk dust samples (BD) were also analysed. BDE-209 predominated, comprising
17 an average 74.3%, 77.3%, 69.2%, and 62.7% ΣBFRs of BD, P1, P2 and P3 respectively.
18 $\Sigma_5\text{NBFRs}$ contributed 24.2%, 21.5%, 29.0% and 35.3% ΣBFRs , while $\Sigma_7\text{tri-hepta-BDEs}$
19 represented 1.5%, 1.2%, 1.7%, and 2.0% ΣBFRs . BEH-TEBP was the predominant NBFR
20 contributing 76.9%, 75.1%, 83.1%, and 83.9% ΣNBFRs in BD, P1, P2 and P3 respectively;
21 followed by DBDPE which contributed 20.1%, 21.9%, 14.1% and 13.9% ΣNBFRs . EH-TBB,
22 BTBPE and PBEB were the least abundant NBFRs. Concentrations of $\Sigma_7\text{tri-hepta-BDEs}$ and
23 BEH-TEBP in P3 exceeded significantly ($P < 0.05$) those in P2, with those in P2 exceeding
24 significantly those in P1. In contrast, no significant differences were found between
25 concentrations of BDE-209, EH-TBB, BTBPE, and DBDPE in different particle size
26 fractions. Concentrations of $\Sigma_7\text{tri-hepta-BDEs}$, BDE-209, and BEH-TEBP in ESD exceeded
27 significantly those in FD ($P < 0.05$). Normalising BFR concentrations to organic carbon
28 content, did not alter these findings. This suggests that differences in BFR concentrations
29 between different particle size fractions are caused by variations in particle surface area to
30 volume ratio, rather than by variations in organic carbon content.

31 **1. Introduction**

32 Polybrominated diphenyl ethers (PBDEs) are a group of brominated flame retardants (BFRs)
33 added to several polymers used in various consumer products such as electronics, textiles,
34 and polyurethane foams. They are blended physically with – rather than chemically bound to
35 - polymeric materials, which likely lead them to migrate into the environment (WHO, 1997;
36 USEPA, 2010 Alaei et al., 2003). PBDEs are persistent and bioaccumulative toxicants, with
37 available information suggesting that they affect negatively the nervous system, the liver and
38 the thyroid system (NICNAS, 2007; USEPA, 2008; Shaw et al., 2009; Noyes et al., 2010; Yu
39 et al., 2011; Johnson et al., 2013; Yang et al., 2014; Zhang et al., 2014; Fang & Stapleton,
40 2014). Hence, their manufacture and new use is now banned or severely restricted in many
41 jurisdictions, and they are listed – or in the case of the Deca-BDE product nominated for
42 listing - under the Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP,
43 2008; 2013a; b). Such bans and restriction on the use of established BFRs like PBDEs has
44 resulted in increased production of several other BFRs collectively referred to here as novel
45 brominated flame retardants (NBFRs). These NBFRs display physicochemical properties
46 similar to those of established BFRs (Covaci 2011; EFSA, 2012), and there
47 is growing concern about increasing environmental contamination with NBFRs (Shi et al.,
48 2009; Shoeib et al., 2012; Ali et al., 2013; Fromme et al., 2014; Poma et al., 2014; Hassan et
49 al., 2015; Al-Omran and Harrad in press; Newton et al., 2015) and their potential negative
50 health effects (Johnson et al., 2013).

51

52 Among the various human exposure pathways, ingestion of indoor settled dust appears to
53 represent a major pathway for human exposure to BFRs in North America and Europe,
54 particularly for young children as a result of normal hand-to-mouth activity (Besis and
55 Samara 2012). Indoor settled dust refers to “particles in building interiors that have settled
56 onto objects, surfaces, floors, and carpeting. These particles may include soil particles that
57 have been tracked or blown into the indoor environment from outdoors as well as organic
58 matter” (USEPA, 2011).

59

60 In general, indoor dust falls into a wide range of particle size fractions ranging from < 2.5 µm
61 to over 2 mm and possessing organic contents between 5% and 95% (Morawska and
62 Salthammer, 2003). However, for human exposure assessment, dust particles < 250 µm are
63 considered of particular concern (USEPA, 2000; 2003), as these have been proposed as those

64 most likely to stick to hands and be ingested (Duggan and Inskip, 1985; Hee et al., 1985;
65 Edwards et al., 1999; Yamamoto et al., 2006).

66
67 A large and growing body of literature has investigated human exposure to BFRs via indoor
68 dust using a disparate range of particle sizes, for example, < 75 μm (Xu et al., 2015), < 100
69 (Kang et al., 2011), < 125 μm (Wu et al., 2007), < 150 μm (Wilford et al., 2005; Shoeib et
70 al., 2012; Whitehead et al., 2012), 25-250 μm (Al-Omran and Harrad, in press), < 500 μm
71 (Allen et al., 2008; Wang et al., 2010), 25-500 μm (Harrad et al. 2008a; 2008b; Muenhor et
72 al., 2010; Brommer et al., 2012), < 1000 μm (Suzuki et al., 2006), < 2000 μm (Gevao et al.,
73 2006) and all fractions (Takigami et al., 2008). Early studies on polycyclic aromatic
74 hydrocarbons (PAHs) and pesticides in indoor dust and airborne particles, indicate that, due
75 to the inverse relationship between particle size and specific surface area, levels of these
76 pollutants increase gradually with decreasing particle size (Lewis et al., 1999; Sygiyama et
77 al., 2000).

78
79 The first study to investigate PBDE concentrations as a function of dust particle size was ten
80 years later (Wei et al., 2009). By analysing four different dust fractions (250- 420 μm , 150-
81 250 μm , 75-150 μm , and < 75 μm) in indoor dust (1 car and 2 homes), Wei et al., (2009)
82 found that PBDE levels in car dust were inversely related to particle size, while they were
83 comparable in homes. Limited studies since, suggest BFR concentrations are influenced
84 significantly by dust particle size. Kefeni et al., (2014) reported that of the PBDEs detected in
85 dust particles < 250 μm from 2 homes and two offices; 93.4% were associated with particles
86 < 150 μm . In a similar study, Chao et al., (2014) found no significant difference in
87 concentrations of Σ_{28} PBDE in different particle sizes of house dust and electronic dust. Based
88 on analysis of office dust particle size fractions, Cao et al., (2013) reported some variation in
89 concentrations of PBDEs with particle size. Concentrations of tri-hexa PBDEs were highest
90 in the 74-100 μm and 100-200 μm particle size fractions, those of hepta-PBDEs were greatest
91 in 200-300 μm and 300-400 μm fractions, octa- and deca-PBDE concentrations peaked in
92 particles < 50 μm , while 2-bis (2,4,6-tribromophenoxy) ethane (BTBPE) was highest in the
93 50-74 μm and 75-100 μm size range. In a subsequent study by the same authors, Cao et al.,
94 (2014) found that in several non-domestic microenvironments, BDE-209 showed higher
95 levels in coarser particles in kindergartens (500-900 μm) and dormitories (900-2000 μm).
96 Moreover, BFR concentrations did not increase constantly with decreasing particle size.
97 Instead, the variation of concentrations with particle size was multi-modal, with the highest

98 levels associated with particle sizes around 900, 100, and 10 μm (Cao et al., 2014). A later
99 study revealed no significant variation in concentrations of HBCDs between different particle
100 size fractions (Cao et al., (2015). Other studies have used forensic microscopy to show that
101 the highest levels of BFRs are present in particles with different morphology, containing
102 more fibre-like material (Wei et al., 2009; Cao et al., 2013).

103

104 From the above, it is apparent that while concentrations of many BFRs vary according to the
105 dust particle size fraction, the exact nature of this variation remains unclear. Moreover, the
106 reasons for such variation have yet to be conclusively elucidated. A related issue is whether
107 BFR concentrations differ significantly between floor and elevated surface dust. We
108 hypothesised previously in a study of house dust from Basrah, Iraq, that the existence of such
109 differences would suggest a need to sample both dust sample types, in order to fully reflect
110 exposure of the room occupants (Al-Omran and Harrad, in press). In that same study, we
111 found that differences in the organic carbon content of dust samples, could not explain the
112 higher concentrations of PBDEs and NBFRs detected in elevated surface dust compared to
113 floor dust in the same rooms. It instead seems more plausible that BFR concentrations will be
114 greater in dust particles with a greater surface area to mass ratio, a hypothesis consistent with
115 the higher proportion of finer particles found in elevated surface compared to floor dust in
116 Basrah (Al-Omran and Harrad, in press).

117

118 This study therefore reports concentrations of eight PBDEs (BDE-28, BDE-47, BDE-99,
119 BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) and five NBFRs:
120 pentabromoethylbenzene (PBEB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis
121 (2-ethylhexyl) 3,4,5,6-tetrabromophthalate (BEH-TEBP), 2-bis (2,4,6-tribromophenoxy)
122 ethane (BTBPE), and decabromodiphenylethane (DBDPE) in both elevated surface dust and
123 floor dust from Birmingham, UK. Each dust sample is separated into three different particle
124 size fractions (125-250 μm , 63-125 μm , and 25-63 μm). BFR concentrations are reported
125 both as dry weight and normalised to dust organic carbon content. These data are used to test
126 our hypotheses that: (a) BFR concentrations in elevated surface dust will exceed those in
127 floor dust; (b) BFR concentrations will increase with decreasing particle size; and (c)
128 variations in dust organic carbon content cannot account for any variations in BFR
129 concentrations between different particle size fractions.

130

131 **2. Materials and methods**

132 **2.1. Chemicals and standards**

133 Native and labelled PBDEs (BDE-28, BDE-47, BDE-77, BDE-99, BDE-100, BDE128, BDE-
134 153, BDE-154, BDE-183, BDE-209 and ¹³C-BDE-209) and NBRs (PBEB, EH-TBB,
135 BTBPE, ¹³C-BTBPE, BEH-TEBP, ¹³C-BEH-TEBP and DBDPE) standards were purchased
136 from Wellington Laboratories Inc. Guelph, Canada as stock solutions in iso-octane, except
137 BDE-209 and ¹³C-BDE-209 which were purchased as stock solutions in nonane. The
138 recovery determination standard PCB-129 in hexane was purchased from Qmx Laboratories,
139 UK. The purity of all standards is > 98%. Ethyl acetate (EA), Acetone (Ac), *n*-Hexane,
140 dichloromethane (DCM), iso-octane, phosphoric acid and sulfuric acid were obtained from
141 Fisher Scientific UK Ltd. All solvents used during analysis were of HPLC analytical grade.
142 Silica gel (pore size 60 Å, 70-230 mesh) was purchased from Sigma Aldrich, Switzerland,
143 anhydrous sodium sulphate was obtained from Sigma Aldrich, USA, and Florisil® (particle
144 size 60-100) acquired from Fluka, USA. Sodium bicarbonate was purchased from Nacalai
145 Tesque, Japan, and sucrose (analytical reagent grade) was purchased from Fisher Scientific,
146 UK. The standard reference material (SRM 2585, “Organic Contaminants in House Dust”)
147 was purchased from the National Institute of Standards and Technology (NIST)
148 Gaithersburg, MD, USA. Finally, ISOLUTE amino propyl columns, SPE cartridges and frits
149 were purchased from Biotage (Uppsala), Sweden.

150

151 **2.2. Sampling and sample preparation**

152 Between September 2013 and February 2014, elevated surface dust (ESD) and floor dust
153 (FD) was collected from the living room and two bedrooms in each of 5 homes in
154 Birmingham, UK. ESD and FD was collected every month for 4 months in two homes (*n* =
155 16), and for 5 months in three (*n* = 30). In order to provide sufficient dust mass for
156 fractionation (especially necessary for ESD for which dust mass loadings in these UK homes
157 were very low), the ESD and FD samples from each home were then combined to yield two
158 bulk dust samples (one ESD and one FD) from each of the 5 homes. Samples were collected
159 using a handheld vacuum cleaner (DIRT DEVIL-DDMHH1-1100W), according to a clearly
160 defined standard protocol (Harrad et al., 2008a). 1 m² of carpeted floor was vacuumed for 2
161 min and, in the case of bare floor, 4 m² for 4 min using 25 µm pore size nylon sample socks
162 (Allied Filter Fabric Pty Ltd, Australia) that were mounted in the furniture attachment tube of
163 the vacuum cleaner. Elevated surfaces (typically between 50-150 cm height) were vacuumed

164 for 2-4 min depending on the surface area. After sampling, socks were closed with a twist tie,
165 sealed in plastic bags and stored at $-20\text{ }^{\circ}\text{C}$. Before sampling, the furniture attachment and the
166 vacuum tubing were cleaned thoroughly using an isopropanol-impregnated disposable wipe
167 and dried between collections. Information on the potential influences on BFR contamination
168 such as: the number and type of putative sources like electronic devices, foam-filled furniture
169 and floor material, ventilation system, house cleaning method was recorded.

170 Initially, to obtain bulk dust samples (referred to here as BD), all samples were sieved using a
171 pre-cleaned, n-hexane rinsed $250\text{ }\mu\text{m}$ mesh stainless testing sieve, covered with the lid and
172 shaken for 3-5 min. After weighing, BD samples were fractionated into three different
173 particle size fractions, $125\text{-}250\text{ }\mu\text{m}$ (referred to as large particle size - P1), $63\text{-}125\text{ }\mu\text{m}$
174 (referred to as medium particle size - P2) and $25\text{-}63\text{ }\mu\text{m}$ (referred to as fine particle size –
175 P3). Fractionation was achieved by using $63\text{ }\mu\text{m}$, $125\text{ }\mu\text{m}$, and $250\text{ }\mu\text{m}$ stainless sieves which
176 were placed over each other and shaken by hand for 5-7 min. Following fractionation, the
177 mass of each fraction was recorded.

178

179 **2.3. Sample extraction and extract purification**

180 Analysis of PBDEs and NBRs in fractionated and BD samples was conducted following the
181 same extraction and clean-up as reported elsewhere (Al-Omran and Harrad, in press).
182 Briefly, samples were spiked with internal standards (BDE-77, BDE-128, ^{13}C -BDE-209, ^{13}C -
183 BTBPE and ^{13}C -BEH-TEBP), extracted by ultrasonication with $3 \times 2\text{ mL}$ n-hexane/acetone
184 (3:1, v/v), evaporated to incipient dryness under a gentle nitrogen stream and resolubilised in
185 1 mL n-hexane (Ali et al., 2011; van den Eede et al., 2012). For sample purification, the
186 extract was fractionated into two fractions (F1 and F2) using 2 g Florisil. F1 (containing
187 PBDEs and DBDPE) was eluted with 12 mL of hexane and F2 (containing the rest of the
188 targeted NBRs) was eluted with 15 mL ethyl acetate. After evaporation to 1 mL , a second
189 purification step for F1 was conducted on 2 g acid silica (44% w/w) and eluted with 15 mL n-
190 hexane/DCM (1:1, v/v). F2 was evaporated to dryness, resolubilised in $3\text{-}5\text{ mL}$ of hexane,
191 then evaporated to 1 mL , and eluted with 12 mL n-hexane/DCM (1:1, v/v) using an
192 aminopropyl functionalised silica column (0.5 g). F1 and F2 were combined and evaporated
193 to incipient dryness, before resolubilisation in $100\text{ }\mu\text{L}$ of iso-octane containing PCB-129 at
194 $250\text{ pg}/\mu\text{L}$ ready for GC/MS analysis (Al-Omran and Harrad, in press).

195

196 **2.4. Instrumental analysis**

197 Analysis of our target compounds was performed using a gas chromatograph (GC) (Trace
198 1310 Gas Chromatograph) coupled to a mass spectrometer (MS) (ISQ Quadrupole MS); both
199 (Thermo Fisher Scientific, USA). The GC was equipped with a programmable temperature
200 vaporiser (PTV) injector and fitted with a capillary fused silica column (RESTEK, USA, 15
201 m x 0.25 mm inner diameter, 0.25 µm film thickness). 2 µL of purified extract were injected
202 on the column. The inlet temperature was set at 92 °C. The GC oven temperature program
203 was set at 50 °C for 0.5 min, ramp 20 °C/min to 240 °C, hold 5 min, ramp 5 °C/min to 270 °C,
204 and ramp 20 °C/min to 305 °C, hold 19 min. Helium was used as a carrier gas with a flow
205 rate of 1.5 mL/min. The MS was operated in ECNI mode. The electron lens voltage was set
206 at 15 V and emission current set at 50 µA. The ion source and transfer line temperatures were
207 300 °C and 320 °C respectively. BDE-77 was used as internal standard for quantification of
208 BDE-28, PBEB, BDE-47, BDE-99, BDE-100; BDE-128 was used to quantify BDE-153,
209 BDE-154 and BDE-183; ¹³C-BTBPE for BTBPE and EH-TBB; ¹³C-BEH-TEBP for BEH-
210 TEBP; and ¹³C-BDE-209 used for BDE-209 and DBDPE.

211 **2.5. QA/QC**

212 All glassware were cleaned by soaking in an alkali solution overnight, before rinsing with tap
213 water, followed by deionised water, baking at 450 °C for 4.5 h, cooling, washing with
214 acetone, and covering with aluminium foil until use. To assess any possible contamination
215 during sample preparation and analysis method, one laboratory blank was processed in
216 parallel with every set of 6 dust samples. In total 8 blanks were conducted. In none of these
217 were any of the target compounds detected, and hence data are not corrected for blank
218 concentrations. Similar results were obtained for field blanks (n=5). These consisted of
219 sodium sulphate that spread on aluminium foil, collected using the vacuum cleaner and
220 treated as a sample. For on-going evaluation of accuracy, an aliquot of SRM2585 was
221 analysed with every 15 samples (or part thereof). The obtained data compared favourably
222 with certified concentrations where provided and with previously reported data for this SRM
223 where certified concentrations were not available, (Tables S1 and S2, Supplementary Data).
224 Recoveries of internal standards ranged from 76 to 91 % in all dust samples (Table S3,
225 Supplementary Data). Limits of detection (LOD) were estimated based on a signal to noise
226 ratio 3:1, with limits of quantification (LOQ) estimated based on signal to noise ratio of 10:1,
227 (Table S4, Supplementary Data)

228 **2.6. Determination of organic carbon content in dust**

229 The total organic carbon (TOC) content of the dust samples was obtained by using a Total
230 Organic Carbon analyzer TOC-V_{CSH/CSN} fitted with a Solid Sample Module SSM-5000, both
231 from SHIMADZU, Japan. The instrument provided measurements of Total Carbon (TC) and
232 Inorganic Carbon (IC), hence the TOC was deduced by subtracting the IC value from TC.
233 Due to the low dust mass available from H5 for fractionation and BFR analysis, TOC was not
234 determined in dust samples from this house.

235 **2.7. Statistical analysis**

236 Statistical analysis of the data was performed using Microsoft Excel 2013 and IBM SPSS
237 statistics software (V. 20). The distribution of the concentration data for target pollutants was
238 tested using the Shapiro-Wilk test. This revealed concentrations of all our target BFRs to be
239 log normally distributed. Hence, data were log-transformed prior to comparison of means via
240 T-test and One-way Repeated Measures ANOVA for testing significant differences between
241 arithmetic means. For the purposes of statistical evaluation, all concentrations below LOQ
242 were assigned a value of 0.5 LOQ. A *p* value < 0.05 was taken to indicate statistical
243 significance.

244

245 **3. Results and discussion**

246 **3.1. Relationship between BFRs in different particle sizes**

247 In all dust samples analysed (n=40), BDE-47, BDE-99, BDE-153, BDE-209, BEH-TEBP,
248 and DBDPE were found with detection frequencies of 100 %, while the detection frequency
249 of other target compounds fell between 40-100%. Concentrations of BDE-28, BDE-100,
250 BDE-154, and PBEB were very low, and they are thus excluded from statistical analysis for
251 individual comparison. However, they were included in calculation of Σ_7 tri-hepta-BDEs and
252 Σ_5 NBFRs. Σ_7 tri-hepta-BDEs refers to the summation of seven congeners (BDE-28, BDE-47,
253 BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183), Σ_5 NBFRs represent the sum of
254 PBEB, EH-TBB, BTBPE, BEH-TEBP, and DBDPE, with Σ BFRs equalling the sum of Σ_7 tri-
255 hepta-BDEs, BDE-209 and Σ_5 NBFRs. Of all target compounds, BDE-209 was the
256 predominant congener making average percentage contributions to Σ BFRs of 74.3%, 77.3%,
257 69.2%, and 62.7% in BD, P1, P2 and P3 dust samples respectively. Σ_5 NBFRs contributed
258 24.2%, 21.5%, 29.1% and 35.3% Σ BFRs, while Σ_7 tri-hepta-BDEs represented 1.5 %, 1.2 %,
259 1.7 %, and 2.0 % of Σ BFRs. Of the target NBFRs, BEH-TEBP predominated, with
260 corresponding mean percentage contributions to Σ_5 NBFRs of 76.9%, 75.1%, 83.1%, and
261 83.9%, followed by DBDPE with percentages 20.1 %, 21.9 %, 14.1 % and 13.9 % in BD, P1,

262 P2 and P3 dust samples respectively. EH-TBB and BTBPE were the least abundant of the
263 target NBFRs, with EH-TBB comprising 0.95 %, 1.08 %, and 1.05 % and 0.79% Σ_5 NBFRs,
264 and BTBPE 1.94%, 1.74%, 1.64% and 1.24% Σ_5 NBFRs in BD, P1, P2 and P3 respectively.
265 Figure 1 shows average concentrations of Σ_7 tri-hepta-BDEs, BDE-209, and Σ_5 NBFRs in BD,
266 P1, P2 and P3, additionally, Figure 2 illustrates concentrations and profiles of tri-hepta-BDEs
267 and NBFRs.

268

269 As hypothesised and consistent with our previous study in Iraq (Al-Omran and Harrad, in
270 press), BFR concentrations in elevated surface dust exceeded those in floor dust. The t-test
271 analysis revealed that with the exception of DBDPE ($p = 0.978$), our target compounds in
272 elevated surface dust exceeded significantly those in floor dust with p values < 0.002 for each
273 of the three different particle size fractions and bulk dust. Average concentrations in ESD
274 exceeded those in FD by the following factors: for Σ_7 tri-hepta-BDEs, 2.4, 2.5, 3.2 and 2.9, for
275 BDE-209, 1.4, 1.6, 1.4, and 1.3, and for Σ_5 NBFRs, 2.9, 3.6, 4.7 and 4.6 in BD, P1, P2 and P3
276 respectively. Figure S1 (supplementary data) compares mean concentrations of Σ_7 tri-hepta-
277 BDEs, BDE-209 and Σ_5 NBFRs in three particle sizes and bulk dust for both floor dust (FD)
278 and elevated surface dust (ESD).

279 **3.2 Distribution patterns of PBDEs and NBFRs with particle size**

280 The average mass percentage of dust fractions P1, P2 and P3 were about 20.6%, 32.7% and
281 46.6%, respectively. In other words, on average ~79% of the bulk dust mass was associated
282 with particles $< 125 \mu\text{m}$, which is comparable to the 80% figure reported for indoor dust
283 particles $< 150 \mu\text{m}$ (Wei et al., 2009). Figure S2 (supplementary data) illustrates the mass
284 percentage contribution of dust fractions P1, P2 and P3 to BD.

285

286 Table 1 provides a statistical summary of concentrations of PBDEs in BD, P1, P2 and P3
287 respectively, with the equivalent data for NBFRs provided in Table 2. Following log
288 transformation of BFR concentrations (ng/g dw), one-way repeated measures ANOVA was
289 applied to test the hypothesis that concentrations of BFRs in P3 would exceed significantly
290 those in P2 and P1. This analysis revealed no significant differences ($p > 0.05$) between
291 concentrations in different dust particle size fractions of: BDE-209, BTBPE, DBDPE, and
292 EH-TBB. In contrast, in the finest particle size fraction (P3), concentrations of BDE-47 ($p =$
293 0.03), BDE-99 ($p = 0.004$), BDE-183 ($p = 0.046$), BEH-TEBP ($p = 0.001$), Σ_5 NBFRs ($p =$
294 0.008), and Σ_7 tri-hepta-BDEs ($p < 0.001$) exceeded significantly those detected in the

295 coarsest fraction (P1). Concentrations of BDE-99 ($p = 0.009$), BEH-TEBP ($p = 0.017$),
296 Σ_5 NBFRs ($p = 0.007$), and Σ_7 tri-hepta-BDEs ($p < 0.001$) in the finest particles (P3)
297 significantly exceeded those in the medium particle size fraction P2. In addition,
298 concentrations in P3 of BDE-183 ($p = 0.053$) were near-significantly elevated over those in
299 P2. Moreover, BDE-99 ($p = 0.008$), BDE-153 ($p = 0.002$) BHH-TEPB ($p = 0.003$), and Σ tri-
300 hepta-BDEs ($p = 0.003$) in the medium particle size fraction (P2) exceeded significantly
301 those in the coarsest size fraction (P1). These findings show that for some BFRs,
302 concentrations will increase with decreasing particle size; while for other BFRs, such a
303 relationship does not exist. Figure 3 illustrates the distribution of Σ_7 tri-hepta-BDEs, BDE-
304 209, BEH-TEBP and DBDPE in particle size fractions from elevated surface dust and floor
305 dust samples from five homes.

306

307 Interestingly, concentrations of our target BFRs in bulk dust (25-250 μm) exceeded
308 significantly those in one or more of the 3 particle size sub-fractions for BDE-153, BEH-
309 TEBP, and Σ_7 tri-hepta-BDEs only. This suggests that the use in many past studies of a
310 relatively broad particle size range has likely not unduly influenced exposure assessment.

311

312 As semi-volatile organic compounds (SVOCs) and additive flame retardants, our target BFRs
313 can be released from the products via volatilisation into surrounding air, depending on their
314 vapour pressure (V_p). Such volatilised BFRs may then undergo deposition to both suspended
315 and settled indoor dust, with the relative partitioning between these two phases governed by
316 the octanol-air partition coefficient (K_{OA}) of the BFRs (Li et al., 2006; Weschler and
317 Nazaroff, 2010). This volatilisation with subsequent deposition process will be more
318 important for BFRs with higher vapour pressures/lower K_{OA} values. By comparison, the
319 presence of less volatile BFRs may be governed more by other processes such as direct
320 contact between BFR source and dust and abrasion of BFR source materials (Rauert and
321 Harrad, 2015; Suzuki et al., 2009; Webster et al., 2009). These factors, combined with the
322 fact that atmospheric deposition of BFRs to dust particles will be greater for finer particle
323 sizes due to their greater surface area to volume ratio (Lewis et. al., 1999; Wei et al., 2009;
324 Mercier et al., 2011); means that significantly higher concentrations on finer dust particles
325 would be anticipated for more volatile BFRs, with this likely less influential for their less
326 volatile counterparts. With the exception of BEH-TEBP – for which the available data on its
327 vapour pressure suggests it would behave in the same way as BDE-209, BTBPE, and

328 DBDPE (see table S5 listing available data for key physicochemical properties of our target
329 BFRs) - this is consistent with our data, that show higher concentrations of BDEs 47, 99, and
330 BEH-TEBP in finer dust particles, but no particle size preference for BDE-153, BDE-209,
331 BTBPE, DBDPE, and EHTBB.

332

333 **3.3 Influence of organic carbon content of dust on BFR particle size distribution**

334 In addition to the particle surface area to volume/mass ratio, variations in organic carbon
335 content between ESD and FD and between different particle size fractions could potentially
336 influence BFR concentrations in both ESD versus FD and in different particle size fractions.
337 Figure S3 illustrates TOC contents of different particle size fractions in both FD and ESD.
338 That this is feasible, is underlined by the fact that significant positive linear correlations were
339 observed between BFR concentrations in all samples and their corresponding total organic
340 carbon (TOC) content, with R values ranging between 0.883 and 0.979 ($p < 0.001$). We
341 therefore examined our data to check whether normalising BFR concentrations to the organic
342 carbon content of the dust fraction analysed, exerted any influence on our observations. We
343 found that doing so, made no difference to our findings based on concentrations normalised
344 to dry weight of dust alone. This is consistent with our previous findings comparing BFRs in
345 ESD and FD in Iraq (Al-Omran and Harrad, in press), that differences in organic carbon
346 content of dust cannot explain either the differences in BFR concentrations in ESD and FD,
347 or the variation in BFR concentrations with particle size fraction. Figure S4 (Supplementary
348 Data) shows BFR concentrations and their corresponding total organic carbon (TOC) content
349 in different particle sizes in elevated surface dust and floor dust from the four homes for
350 which this was possible.

351

352 **4. Conclusions**

353 While based on a small number of homes, our findings suggest that BFR concentrations
354 recorded in indoor dust samples are influenced by the particle size fraction analysed.
355 However, there is substantial variation between different BFRs. Specifically, while
356 concentrations of more volatile BFRs like Σ tri-hepta-BDEs are significantly higher in dust
357 particles of 25-63 μm diameter than in those of 125-250 μm and 63-125 μm diameter;
358 concentrations of less volatile BFRs such as BDE-209, BTBPE, EH-TBB, and DBDPE do
359 not differ significantly between different dust particle size fractions. This is consistent with
360 hypotheses that less volatile BFRs enter dust predominantly via mechanisms that do not

361 display clear preference for any one particle size fraction, such as abrasion of and direct
362 contact with flame retarded materials, Consistent with our previous research in Iraq,
363 concentrations of most of our target BFRs in elevated surface dust exceed significantly those
364 in floor dust from the same UK microenvironments. Variations in the organic carbon content
365 of dust explain neither the higher concentrations of BFRs in elevated surface compared to
366 floor dust, nor the higher concentrations of some BFRs in finer dust particles. Instead, our
367 data suggest that it is the greater surface area to volume ratio of finer dust particles that
368 accounts for their higher BFR concentrations. Despite our small sample size precluded
369 meaningful human exposure assessment, the results of this research support the idea that
370 human exposure assessment could be affected by the dust particle size fractions analysed, and
371 more detailed study of this issue is recommended.

372

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378

379 **References**

- 380 Alaei, M. (2003). "An overview of commercially used brominated flame retardants, their
381 applications, their use patterns in different countries/regions and possible modes of
382 release." Environment International **29**(6): 683-689.
- 383 Ali, N., S. Harrad, D. Muenhor, H. Neels and A. Covaci (2011). "Analytical characteristics
384 and determination of major novel brominated flame retardants (NBFRs) in indoor
385 dust." Analytical and Bioanalytical Chemistry **400**(9): 3073-3083.
- 386 Ali, N., L. Ali, T. Mehdi, A. C. Dirtu, F. Al-Shammari, H. Neels and A. Covaci (2013).
387 "Levels and profiles of organochlorines and flame retardants in car and house dust
388 from Kuwait and Pakistan: Implication for human exposure via dust ingestion."
389 Environment International **55**: 62-70.
- 390 Allen, J. G., M. D. McClean, H. M. Stapleton and T. F. Webster (2008). "Critical factors in
391 assessing exposure to PBDEs via house dust." Environment International **34**(8): 1085-
392 1091.
- 393 Al-Omran, L. S. and S. Harrad (2015). "Polybrominated diphenyl ethers and "novel"
394 brominated flame retardants in floor and elevated surface house dust from Iraq:
395 Implications for human exposure assessment." Emerging Contaminants. [http://dx.doi.
396 org/10.1016/j.emcon.2015.10.001](http://dx.doi.org/10.1016/j.emcon.2015.10.001).
- 397 Besis, A. and C. Samara (2012). "Polybrominated diphenyl ethers (PBDEs) in the indoor
398 and outdoor environments--a review on occurrence and human exposure."
399 Environmental Pollution **169**: 217-229.

400 Brommer, S., S. Harrad, N. Van den Eede and A. Covaci (2012). "Concentrations of
401 organophosphate esters and brominated flame retardants in German indoor dust
402 samples." Journal of Environmental Monitoring **14**(9): 2482-2487.

403 Cao, Z., F. Xu, A. Covaci, M. Wu, H. Wang, G. Yu, B. Wang, S. Deng, J. Huang and X.
404 Wang (2014). "Distribution patterns of brominated, chlorinated, and phosphorus flame
405 retardants with particle size in indoor and outdoor dust and implications for human
406 exposure." Environmental Science & Technology **48**(15): 8839-8846.

407 Cao, Z., G. Yu, Y. Chen, C. Liu, K. Liu, T. Zhang, B. Wang, S. Deng and J. Huang (2013).
408 "Mechanisms influencing the BFR distribution patterns in office dust and implications
409 for estimating human exposure." Journal of Hazardous Materials **252–253**: 11-18.

410 Cao, Z. G., G. Yu, Y. S. Chen, Q. M. Cao, H. Fiedler, S. B. Deng, J. Huang and B. Wang
411 (2012). "Particle size: a missing factor in risk assessment of human exposure to toxic
412 chemicals in settled indoor dust." Environment International **49**: 24-30.

413 Cao, Z., F. Xu, W. Li, J. Sun, M. Shen, X. Su, J. Feng, G. Yu, and A. Covaci (2015)."
414 Seasonal and particle size-dependent variations of hexabromocyclododecanes in
415 settled dust: Implications for sampling." Environmental Science &
416 Technology **49** (18): 11151-11157.

417 Chao, H.-R., C.-G. Shy, H.-L. Huang, T.-W. Koh, T.-S. Tok, S. C.-C. Chen, B.-A. Chiang,
418 Y.-M. Kuo, K.-C. Chen and G.-P. Chang-Chien (2014) "Particle-Size Dust
419 Concentrations of Polybrominated Diphenyl Ethers (PBDEs) in Southern Taiwanese
420 Houses and Assessment of the PBDE Daily Intakes in Toddlers and Adults." Aerosol
421 and Air Quality Research **14**: 1299-1309.

422 Covaci, A., S. Harrad, M. A. Abdallah, N. Ali, R. J. Law, D. Herzke and C. A. de Wit
423 (2011). "Novel brominated flame retardants: a review of their analysis, environmental
424 fate and behaviour." Environment International **37**(2): 532-556.

425 Duggan, M.J. and Inskip, M.J. (1985). "Childhood exposure to lead in surface dust and soil:
426 A community health problem" Public Health Review **13**: 1-54.

427 Edwards R. D. and Lioy P. J. (1999). "The EL sampler: a press sampler for the quantitative
428 estimation of dermal exposure to pesticides in housedust." Exposure Analysis and
429 Environmental Epidemiology **9**:521–9.

430 Fang, M. and H. M. Stapleton (2014). "Evaluating the bioaccessibility of flame retardants
431 in house dust using an in vitro Tenax bead-assisted sorptive physiologically based
432 method." Environmental Science & Technology **48**(22): 13323-13330.

433 EFSA. European Food Safety Authority (2012). "Scientific Opinion on Emerging and
434 Novel Brominated Flame Retardants (BFRs) in Food." EFSA Journal; **10**(10):2908.

435 Fromme, H., B. Hilger, E. Kopp, M. Miserok and W. Volkel (2014). "Polybrominated
436 diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and "novel" brominated
437 flame retardants in house dust in Germany." Environment International **64**: 61-68.

438 Gevao, B., M. Al-Bahloul, A. N. Al-Ghadban, A. Al-Omair, L. Ali, J. Zafar and M. Helaleh
439 (2006). "House dust as a source of human exposure to polybrominated diphenyl ethers
440 in Kuwait." Chemosphere **64**(4): 603-608.

441 Harrad, S., C. Ibarra, M. A.-E. Abdallah, R. Boon, H. Neels and A. Covaci (2008a).
442 "Concentrations of brominated flame retardants in dust from United Kingdom cars,
443 homes, and offices: Causes of variability and implications for human exposure."
444 Environment International **34**(8): 1170-1175.

445 Harrad, S., C. Ibarra, M. Diamond, L. Melymuk, M. Robson, J. Douwes, L. Roosens, A. C.
446 Dirtu and A. Covaci (2008b). "Polybrominated diphenyl ethers in domestic indoor dust
447 from Canada, New Zealand, United Kingdom and United States." Environment
448 International **34**(2): 232-238.

- 449 Hassan, Y. and T. Shoeib (2015). "Levels of polybrominated diphenyl ethers and novel
450 flame retardants in microenvironment dust from Egypt: An assessment of human
451 exposure." Science of The Total Environment **505**: 47-55.
- 452 Hee, S. S. Q., B. Peace, C. S. Clark, J. R. Boyle, R. L. Bornschein and P. B. Hammond
453 (1985). "Evolution of efficient methods to sample lead sources, such as house dust and
454 hand dust, in the homes of children." Environmental Research **38**(1): 77-95.
- 455 Johnson, P. I., H. M. Stapleton, B. Mukherjee, R. Hauser and J. D. Meeker (2013).
456 "Associations between brominated flame retardants in house dust and hormone levels
457 in men." Science of The Total Environment **445-446**: 177-184.
- 458 Kang, Y., H. S. Wang, K. C. Cheung and M. H. Wong (2011). "Polybrominated diphenyl
459 ethers (PBDEs) in indoor dust and human hair." Atmospheric Environment **45**(14):
460 2386-2393.
- 461 Kefeni, K. K. and J. O. Okonkwo (2014). "Distribution of polybrominated diphenyl ethers
462 and dust particle size fractions adherent to skin in indoor dust, Pretoria, South Africa."
463 Environmental Science And Pollution Research **21**(6): 4376-4386.
- 464 Lewis, R. G., C. R. Fortune, R. D. Willis, D. E. Camann and J. T. Antley (1999).
465 "Distribution of pesticides and polycyclic aromatic hydrocarbons in house dust as a
466 function of particle size." Environmental Health Perspectives **107**(9): 721-726.
- 467 Li, X., J. Chen, L. Zhang, X. Qiao and L. Huang (2006). "The Fragment Constant Method
468 for Predicting Octanol–Air Partition Coefficients of Persistent Organic Pollutants at
469 Different Temperatures." Journal of Physical and Chemical Reference Data **35**(3):
470 1365-1384.
- 471 Mercier, F., P. Glorennec, O. Thomas and B. Le Bot (2011). "Organic contamination of
472 settled house dust, a review for exposure assessment purposes." Environmental
473 Science & Technology **45**(16): 6716-6727.
- 474 Morawska, L. and T. Salthammer (2003). "Indoor environment; airborne particles and
475 settled dust" Indoor Environment, WILEY-VCH Verlag GmbH & Co. KGaA; ISBN:
476 978-3-527-30525-4.
- 477 Muenhor, D., S. Harrad, N. Ali and A. Covaci (2010). "Brominated flame retardants
478 (BFRs) in air and dust from electronic waste storage facilities in Thailand."
479 Environment International **36**(7): 690-698.
- 480 Newton, S., U. Sellström and C. A. de Wit (2015). "Emerging Flame Retardants, PBDEs,
481 and HBCDDs in Indoor and Outdoor Media in Stockholm, Sweden." Environmental
482 Science & Technology **49**(5): 2912-2920.
- 483 NICNAS, 2007, Interim Public Health Risk Assessment of Certain PBDE congeners
484 (online). Available from: [http://www.nicnas.gov.au/_data/assets/pdf_file/0003/4944/
485 Final-Interim-Report-PBDE-March.pdf](http://www.nicnas.gov.au/_data/assets/pdf_file/0003/4944/Final-Interim-Report-PBDE-March.pdf), accessed September 2012.
- 486 Noyes P. D., S. M. Kelly, C. L. Mitchelmore, H. M. Stapleton. (2010) "Characterizing the
487 in vitro hepatic biotransformation of the flame retardant BDE 99 by common carp".
488 Aquatic Toxicology **97**(2): 142-150.
- 489 Poma, G., C. Roscioli and L. Guzzella (2014). "PBDE, HBCD, and novel brominated flame
490 retardant contamination in sediments from Lake Maggiore (Northern Italy)."
491 Environmental Monitoring and Assessment **186**(11): 7683-7692.
- 492 Rauert, C. and S. Harrad, (2015) "Mass transfer of PBDEs from plastic TV casing to indoor
493 dust via three migration pathways — A test chamber investigation", Science of the
494 Total Environment, 536, 568-574.

- 495 Shaw, S. D., M. L. Berger, D. Brenner, K. Kannan, N. Lohmann and O. Pöpke (2009).
496 "Bioaccumulation of polybrominated diphenyl ethers and hexabromocyclododecane in
497 the northwest Atlantic marine food web." Science of The Total Environment **407**(10):
498 3323-3329.
- 499 Shi, T., S. J. Chen, X. J. Luo, X. L. Zhang, C. M. Tang, Y. Luo, Y. J. Ma, J. P. Wu, X. Z.
500 Peng and B. X. Mai (2009). "Occurrence of brominated flame retardants other than
501 polybrominated diphenyl ethers in environmental and biota samples from southern
502 China." Chemosphere **74**(7): 910-916.
- 503 Shoeib, M., T. Harner, G. M. Webster, E. Sverko and Y. Cheng (2012). "Legacy and
504 current-use flame retardants in house dust from Vancouver, Canada." Environmental
505 Pollution **169**: 175-182.
- 506 Sugiyama, T., A. Takashi, M. Soma and H. Matsushita (2000). "Size distribution of
507 polycyclic aromatic hydrocarbons in indoor airborne particulates." Indoor and Built
508 Environment **9**:265-276.
- 509 Suzuki, G., K. Nose, H. Takigami, S. Takahashi and S. I. Sakai (2006). "PBDEs and
510 PBDD/Fs in house and office dust from Japan". Organohalogen Compounds **141**: 30-
511 41.
- 512 Suzuki, G., A. Kida, S.-i. Sakai and H. Takigami (2009). "Existence State of Bromine as an
513 Indicator of the Source of Brominated Flame Retardants in Indoor Dust." Environmental Science & Technology **43**(5): 1437-1442.
- 514 Takigami, H., G. Suzuki, Y. Hirai and S. Sakai (2008). "Transfer of brominated flame
515 retardants from components into dust inside television cabinets." Chemosphere **73**(2):
516 161-169.
- 517
518 USEPA. US Environmental Protection Agency. (2003) "Superfund Lead Contaminated
519 Residential Sites Handbook; Final; OSWER 9285.7-50; Office of Solid Waste and
520 Emergency Response, Lead Sites Workgroup: Washington, DC.
- 521 USEPA. US Environmental Protection Agency (2000) SHORT SHEET: TRW
522 RECOMMENDATIONS FOR SAMPLING AND ANALYSIS OF SOIL (online).
523 Available from: <http://www.itrcweb.org/ism-1/references/sssiev.pdf> (Accessed May
524 2015).
- 525 USEPA. US Environmental Protection Agency. (2008). "Toxicological Review of
526 Decabromodiphenyl Ether (BDE-209) (online). Available from:
527 <http://www.epa.gov/iris/toxreviews/0035tr.pdf> (Accessed May 2015).
- 528 USEPA. US Environmental Protection Agency. (2010). "An Exposure Assessment of
529 Polybrominated Diphenyl Ethers" (online) available from:
530 [file:///C:/Users/layla/Downloads/PBDE_FINAL_MAY2010%20\(12\).PDF](file:///C:/Users/layla/Downloads/PBDE_FINAL_MAY2010%20(12).PDF) (Accessed
531 January, 2016).
- 532 USEPA. US Environmental Protection Agency. (2011). "Exposure Factors Handbook;
533 Chapter 5—Soil and Dust Ingestion" (online) available from:
534 <https://www.epa.gov/sites/production/files/2015-09/documents/efh-chapter05.pdf>
535 (Accessed January, 2016).
- 536 UNEP. Stockholm Convention on POPs. (2008). "Risk management evaluation for
537 commercial octabromodiphenyl ether" (online) available from:
538 <http://chm.pops.int/portals/0/repository/poprc4/unep-pops-poprc.4-6.english.pdf>.
539 (Accessed December, 2015).
- 540 UNEP. Stockholm Convention on POPs. (2013a) "Listing of hexabromocyclododecane"
541 (online). Available from:
542 <https://treaties.un.org/doc/Publication/CN/2013/CN.934.2013-Eng.pdf> (Accessed
543 December 2015).

544 UNEP. Stockholm Convention on POPs. (2013b) "Proposal to list decabromodiphenyl ether
545 (commercial mixture, c-decaBDE) in Annexes A, B and/or C to the Stockholm
546 Convention on Persistent Organic Pollutants" (online). Available from:
547 <http://chm.pops.int/Portals/0/download.aspx?d¼UNEPPOPS- POPRC.9-2>.

548 Van den Eede, N., A. C. Dirtu, N. Ali, H. Neels and A. Covaci (2012). "Multi-residue
549 method for the determination of brominated and organophosphate flame retardants in
550 indoor dust." *Talanta* **89**: 292-300.

551 Wang, J., Y. J. Ma, S. J. Chen, M. Tian, X. J. Luo and B. X. Mai (2010). "Brominated
552 flame retardants in house dust from e-waste recycling and urban areas in South China:
553 implications on human exposure." *Environment International* **36**(6): 535-541.

554 Webster, T. F., S. Harrad, J. R. Millette, R. D. Holbrook, J. M. Davis, H. M. Stapleton, J.
555 G. Allen, M. D. McClean, C. Ibarra, M. A.-E. Abdallah and A. Covaci (2009).
556 "Identifying Transfer Mechanisms and Sources of Decabromodiphenyl Ether (BDE
557 209) in Indoor Environments Using Environmental Forensic Microscopy."
558 *Environmental Science & Technology* **43**(9): 3067-3072.

559 Wei, H., M. Turyk, S. Cali, S. Dorevitch, S. Erdal and A. Li (2009). "Particle size
560 fractionation and human exposure of polybrominated diphenyl ethers in indoor dust
561 from Chicago." *Journal of environmental science and health. Part A, Toxic/hazardous
562 substances & environmental* **44**(13): 1353-1361.

563 Weschler, C. J. and W. W. Nazaroff (2010). "SVOC partitioning between the gas phase and
564 settled dust indoors." *Atmospheric Environment* **44**(30): 3609-3620.

565 WHO, WORLD HEALTH ORGANIZATION, (1997). Environmental Health Criteria 192
566 (online). Available from: <http://www.inchem.org/documents/ehc/ehc/ehc192.htm>

567 Whitehead, T. P., J. R. Nuckols, M. H. Ward and S. M. Rappaport (2012). "Carpet-dust
568 chemicals as measures of exposure: Implications of variability." *Emerging Themes in
569 Epidemiology* **9**(1): 2.

570 Wilford, B. H., M. Shoeib, T. Harner, J. Zhu and K. C. Jones (2005). "Polybrominated
571 diphenyl ethers in indoor dust in Ottawa, Canada: implications for sources and
572 exposure." *Environmental Science & Technology* **39**(18): 7027-7035.

573 Wu, N., T. Herrmann, O. Paepke, J. Tickner, R. Hale, L. E. Harvey, M. La Guardia, M. D.
574 McClean and T. F. Webster (2007). "Human exposure to PBDEs: associations of
575 PBDE body burdens with food consumption and house dust concentrations."
576 *Environmental Science & Technology* **41**(5): 1584-1589.

577 Xu, F., Y. Liu, J. Wang, G. Zhang, W. Zhang, L. Liu, J. Wang, B. Pan and K. Lin (2015).
578 "Characterization of heavy metals and brominated flame retardants in the indoor and
579 outdoor dust of e-waste workshops: implication for on-site human exposure."
580 *Environmental Science and Pollution Research* **22**(7): 5469-5480.

581 Yamamoto, N., Y. Takahashi, J. Yoshinaga, A. Tanaka and Y. Shibata (2006). "Size
582 distributions of soil particles adhered to children's hands." *Archives of Environmental
583 Contamination and Toxicology* **51**(2): 157-163.

584 Yang, W., J. Fu, T. Wang, H. Liu, Y. Wang, Q. Zhou and G. Jiang (2014). "Alterations of
585 endogenous metabolites in urine of rats exposed to decabromodiphenyl ether using
586 metabonomic approaches." *Journal of Environmental Sciences* **26**(4): 900-908.

587 Yu, Y., Y. Pang, X. Zhang, C. Li, Z. Yu and J. Fu (2011). "Optimization of an in vitro
588 method to measure the bioaccessibility of polybrominated diphenyl ethers in dust using
589 response surface methodology." *Journal of Environmental Sciences* **23**(10): 1738-
590 1746.

591 Zhang, F., G. Lu, J. Liu, Z. Yan and Z. Zhang (2014). "Bioaccumulation, distribution and
592 metabolism of BDE-153 in the freshwater fish *Carassius auratus* after dietary
593 exposure." Ecotoxicology and Environmental Safety **108**: 16-22.

594

595

596