

# Within-room and within-home spatial and temporal variability in concentrations of legacy and “novel” brominated flame retardants in indoor dust

Al-Omran, Layla Salih; Harrad, Stuart

DOI:

[10.1016/j.chemosphere.2017.11.147](https://doi.org/10.1016/j.chemosphere.2017.11.147)

License:

Creative Commons: Attribution-NonCommercial-NoDerivs (CC BY-NC-ND)

*Document Version*

Peer reviewed version

*Citation for published version (Harvard):*

Al-Omran, LS & Harrad, S 2018, 'Within-room and within-home spatial and temporal variability in concentrations of legacy and “novel” brominated flame retardants in indoor dust', *Chemosphere*, vol. 193, pp. 1105-1112. <https://doi.org/10.1016/j.chemosphere.2017.11.147>

[Link to publication on Research at Birmingham portal](#)

**Publisher Rights Statement:**

Checked for eligibility: 22/01/2018

**General rights**

Unless a licence is specified above, all rights (including copyright and moral rights) in this document are retained by the authors and/or the copyright holders. The express permission of the copyright holder must be obtained for any use of this material other than for purposes permitted by law.

- Users may freely distribute the URL that is used to identify this publication.
- Users may download and/or print one copy of the publication from the University of Birmingham research portal for the purpose of private study or non-commercial research.
- User may use extracts from the document in line with the concept of 'fair dealing' under the Copyright, Designs and Patents Act 1988 (?)
- Users may not further distribute the material nor use it for the purposes of commercial gain.

Where a licence is displayed above, please note the terms and conditions of the licence govern your use of this document.

When citing, please reference the published version.

**Take down policy**

While the University of Birmingham exercises care and attention in making items available there are rare occasions when an item has been uploaded in error or has been deemed to be commercially or otherwise sensitive.

If you believe that this is the case for this document, please contact [UBIRA@lists.bham.ac.uk](mailto:UBIRA@lists.bham.ac.uk) providing details and we will remove access to the work immediately and investigate.

1  
2  
3  
4 1 **WITHIN-ROOM AND WITHIN-HOME SPATIAL AND TEMPORAL**  
5 2 **VARIABILITY IN CONCENTRATIONS OF LEGACY AND “NOVEL”**  
6 3 **BROMINATED FLAME RETARDANTS IN INDOOR DUST**  
7  
8 4

9  
10 5 Layla Salih Al-Omran <sup>a,b</sup> and Stuart Harrad<sup>a</sup>

11 6 <sup>a</sup>School of Geography, Earth, and Environmental Sciences, University of Birmingham,  
12 7 Birmingham, B15 2TT, UK

13 8 <sup>b</sup>Division of Ecology, College of Science, University of Basrah, Basrah, Iraq  
14 9

15 10 **ABSTRACT**

16 11 To test the hypothesis that assessments of human exposure to PBDEs and NBRs (PBEB,  
17 12 EH-TBB, BEH-TEBP, BTBPE and DBDPE) via dust ingestion should take into account  
18 13 spatial and temporal variability in dust contamination; 238 dust samples were collected from  
19 14 nine different rooms within three homes in Birmingham UK. In each room, three different  
20 15 dust samples were taken at monthly intervals for nine months, one sample from elevated  
21 16 surfaces and two samples from two different floor areas. Substantial within-room and within-  
22 17 home spatial variability in BFR concentrations was apparent between two floor areas and  
23 18 between different rooms due to the varying distances of sampled surfaces from potential BFR  
24 19 sources. With the exception of DBDPE, BFR concentrations in elevated surface dust  
25 20 exceeded significantly ( $p < 0.05$ ) those in floor dust from the same rooms. Considerable  
26 21 within-room and within-home temporal variability in BFR concentrations was also apparent  
27 22 over a nine month sampling period. This is likely attributable to changes in room contents.  
28 23 The relative standard deviation of BFR concentrations observed in such temporal variation  
29 24 sample series exceeded those obtained from replicate analyses of SRM2585. Based on  
30 25 observed spatial and temporal variability, exposure estimates based on analysis of a single  
31 26 dust sample taken from one specific floor area at one specific point in time may not be entirely  
32 27 representative of human exposure in that room. Noticeable variability in BFR concentrations  
33 28 was also observed between colder and warmer seasons. In 13 out of 17 floor areas,  
34 29 concentrations of  $\Sigma_8$ tri-deca-BDEs were higher in colder seasons, while those of  $\Sigma_5$ NBRs  
35 30 were higher in warmer seasons. Significant negative correlation was observed in three rooms  
36 31 between concentrations of BDE-99,  $\Sigma_6$ tri-hepta-BDEs and BEH-TEBP and dust loading  
37 32 ( $\text{g}/\text{m}^2$ ), suggesting “dilution” occurs at higher dust loadings.

60  
61  
62 **KEYWORDS:** PBDEs; NBFRs; Indoor dust; Spatial and temporal variability; Human  
63 exposure.  
64  
65  
66

## 67 **1. INTRODUCTION**

68  
69 The toxicity of some brominated flame retardants (BFRs), such as polybrominated diphenyl  
70 ethers (PBDEs) and “novel” brominated flame retardants (NBFRs) has led to concern about  
71 human exposure (USEPA, 2006; 2008a; 2008b; 2008c; NICNAS, 2007; Noyes et al., 2010;  
72 Chevrier et al., 2010; EFSA, 2012; European Commission, 2012; Johnson et al., 2013; Li et  
73 al., 2014; Mankidy et al., 2014; Mariani, et al., 2015). Moreover, several studies show  
74 significant positive correlation between concentrations of BFRs in indoor dust and human  
75 tissues such as human milk (Wu et al., 2007, Toms et al., 2009; Coakley et al., 2013), human  
76 hair (Kang et al., 2011; Tang et al., 2013) and serum samples (Johnson et al., 2010; Stapleton  
77 et al., 2012); suggesting that indoor dust ingestion is a major pathway of exposure to such  
78 chemicals, particularly for young children due to their hand-to-mouth behaviour (Stapleton et  
79 al., 2005; Wang et al., 2010; Hoffman et al., 2015)  
80  
81  
82  
83  
84  
85  
86  
87

88 Assessments of human exposure to chemical pollutants via indoor dust ingestion require  
89 knowledge about locations where people spend their time and thus come into contact with  
90 such pollutants. However, few studies have investigated within-room (dust samples taken at  
91 the same time from different locations within the same room) and within-home (dust samples  
92 taken at the same time from different rooms within the same home) spatial variability. From  
93 five separate floor areas within the same room in five dwellings, Harrad et al., (2008a) found  
94 substantial within-room spatial variability in BFR concentrations. The relative standard  
95 deviations (RSD) for  $\Sigma$ tri-hexa-BDE ranged between 28% and 80%. Another study of HBCDs  
96 revealed little spatial variability in some rooms (RSD = 7% - 8%), while others displayed  
97 large variability (RSD = 19% - 100%) (Harrad et al., 2009). More recently, Muenhor and  
98 Harrad, (2012) also examined within-room spatial variability in PBDE contamination of dust  
99 from separate rooms, finding that the PBDE concentrations in an area close to putative PBDEs  
100 sources (TV, laptop, and sofa) exceeded significantly those in an area 2 m away from the  
101 same sources. This is considered to reflect the relationship between contamination and  
102 potential emission sources. In terms of within-room vertical variability in dust contamination,  
103 our previous studies (Al-Omran and Harrad 2016a; 2016b) revealed concentrations of several  
104 BFRs to be significantly higher ( $p < 0.05$ ) in dust collected from elevated surfaces (ESD) like  
105 chairs and tables than in floor dust (FD) from the same rooms. This is likely due to differences  
106  
107  
108  
109  
110  
111  
112  
113  
114  
115  
116  
117  
118

119  
120  
121 67 in the particle size distribution in ESD and FD. Higher proportions of finer particle sizes were  
122  
123 68 found in ESD with higher concentrations of BFRs detected in the finer particle sizes. Another  
124  
125 69 study (Cequier et al., 2014) reported that median concentrations of BFRs in elevated surface  
126  
127 70 dust exceeded those in floor dust. Within-home spatial variability has also been reported in a  
128  
129 71 small number of studies. Concentrations of Penta- and Deca-BDE congeners were, on  
130  
131 72 average, significantly higher in the living room than those in the bedroom (Allen et al., 2008),  
132  
133 73 while average concentrations of  $\Sigma$ PBDE in the bedroom ( $430\pm 180$  ng/g) exceeded  
134  
135 74 substantially those in another bedroom ( $170\pm 340$  ng/g) from the same home (Muenhor and  
136  
137 75 Harrad, 2012). However, recent studies (Venier et al., 2016; Kuang et al., 2016) found no  
138  
139 76 statistically significant differences in BFR concentrations in dust from the living room and  
140  
141 77 bedroom.

142  
143 78  
144 79 To date, only four studies (Allen et al., 2008; Harrad et al., 2008a; 2009; Muenhor and Harrad,  
145  
146 80 2012) have investigated within-room and within-home temporal variations in concentrations  
147  
148 81 of BFRs; collectively suggesting that most temporal variability is attributable to changes in  
149  
150 82 room contents of putative BFR sources. Over a 9-10 month monitoring period, a substantial  
151  
152 83 month-to-month rise in BDE-209 contamination of dust was found following fitting of a new  
153  
154 84 fabric padded bed and polyester fabric blinds (Harrad et al., 2008a). In a similar vein,  
155  
156 85 Muenhor and Harrad (2012) reported substantial within room temporal variability in  $\Sigma$ PBDE  
157  
158 86 concentrations in monthly samples collected over an 8 month sampling period as a  
159  
160 87 consequence of the introduction and removal of putative sources such as a TV and a bed. The  
161  
162 88 RSD values for  $\Sigma$ PBDEs were between 15% and 200% (Muenhor and Harrad, 2012). Another  
163  
164 89 study (Allen et al., 2008) reported no significant difference between Penta- and Deca-BDE  
165  
166 90 concentrations in house dust in 20 homes collected 8 months apart, attributing this to minimal  
167  
168 91 changes in room furnishings between the sampling periods.

169  
170 92  
171 93 Noticeable seasonal variability in BFR concentrations has also been observed between colder  
172  
173 94 and warmer months or between different seasons. Out of fourteen floor areas, while in seven  
174  
175 95 sampled areas, average concentrations of  $\Sigma$ PBDEs in the colder months was higher than in  
176  
177 96 warmer months, the reverse was observed in the other seven areas (Muenhor and Harrad  
178  
179 97 2012). According to Muenhor and Harrad (2012), the lack of clear seasonal variation is  
180  
181 98 attributable to the greater volatile emissions of BFRs in warmer months being offset by higher  
182  
183 99 ventilation during the same period. Elsewhere, Yu et al., (2012) noted that PBDE  
184  
185 100 concentrations were summer > winter > spring > autumn; while over a 10 months monitoring

178  
179  
180 101 period, Cao et al., (2014) reported maximum: minimum concentration ratios were between 2  
181  
182 102 and 10, underlining the importance of the time of dust collection for exposure assessments.  
183  
184 103

185 104 Despite a lack of data on how dust ingestion rates vary with dust loading, it is plausible that  
186  
187 105 higher dust loadings will lead to increased dust ingestion rates. While this would suggest  
188  
189 106 higher exposures in dustier rooms, it is also plausible that higher dust loadings will dilute BFR  
190  
191 107 concentrations in dust, and it is not clear how these two competing factors will impact on  
192  
193 108 exposure. To date, while three studies (Harrad et al., 2008a; 2009; Muenhor and Harrad 2012)  
194  
195 109 have examined the evidence for such "dilution" of BFR concentrations in the dust at higher  
196  
197 110 dust loadings; their findings are inconclusive. It has been hypothesised that, under certain  
198  
199 111 conditions, "dilution" of BFR concentrations will occur at greater dust loadings. These  
200  
201 112 conditions are: (a) BFR emissions remain constant through the monitoring period, and (b) the  
202  
203 113 source of the dust and BFRs are independent – i.e. the main source of the BFR to dust is not  
204  
205 114 direct abrasion of fibres or particles from a source material (Harrad et al., 2008a; 2009).  
206  
207 115

208 116 Against this background; the location of the sample, time of sampling, and surface loading  
209  
210 117 are potentially important factors affecting the levels of pollutants in indoor dust. This study  
211  
212 118 therefore aims to test the hypothesis that assessments of human exposure to PBDEs (BDE-  
213  
214 119 28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183 and BDE-209) and their  
215  
216 120 potential replacement NBFRs{(pentabromoethylbenzene (PBEB), 2-ethylhexyl-2,3,4,5-  
217  
218 121 tetrabromobenzoate (EH-TBB), bis (2-ethylhexyl) 3,4,5,6-tetrabromophthalate (BEH-  
219  
220 122 TEBP), 2-bis (2,4,6-tribromophenoxy) ethane (BTBPE), and decabromodiphenylethane  
221  
222 123 (DBDPE)}via dust ingestion, are affected by spatial, temporal and seasonal (warmer and  
223  
224 124 colder months) variability in dust contamination. We furthermore investigate the relationship  
225  
226 125 between BFR concentrations (ng/g) and BFR dust loading (g/m<sup>2</sup>). To the best of  
227  
228 126 our knowledge, this study is the first to examine spatial and temporal variability in  
229  
230 127 concentrations of PBEB, EH-TBB, BEH-TEBP and BTBPE in indoor dust.  
231  
232 128

## 233 129 **2. MATERIALS AND METHODS**

### 234 130 **2.1. Sampling and sample preparation**

235 131 From three homes (H1, H2, and H3) in Birmingham, UK, 238 indoor dust samples were  
236  
237 132 collected at monthly intervals from three different rooms (R1 = living room, R2 = adult  
238  
239 133 bedroom, and R3 = study or child's bedroom in H3). From each room, two dust samples were  
240  
241 134 obtained from two different floor areas F1 and F2, following the sampling protocol described

237  
238  
239  
240  
241  
242  
243  
244  
245  
246  
247  
248  
249  
250  
251  
252  
253  
254  
255  
256  
257  
258  
259  
260  
261  
262  
263  
264  
265  
266  
267  
268  
269  
270  
271  
272  
273  
274  
275  
276  
277  
278  
279  
280  
281  
282  
283  
284  
285  
286  
287  
288  
289  
290  
291  
292  
293  
294  
295

135 elsewhere (Harrad et al., 2008a), with an additional dust sample collected from the elevated  
136 surfaces (ES), such as sofas, tables, shelves, and large articles present on tables and shelves  
137 (Al-Omran and Harrad 2016a, 2016b). Dust was not collected from under furniture or from  
138 highly elevated surfaces with which human contact is rare, such as the tops of wardrobes. For  
139 calculation of dust loadings, the mass of floor dust collected per unit surface area sampled  
140 was recorded. Sampling was conducted for nine months between May 2013 and March 2014,  
141 with no samples collected in July and August 2013. Information on the potential influences  
142 on BFR contamination such as: the number and type of putative sources like electronic  
143 devices, foam-filled furniture and floor material, ventilation system, and house cleaning  
144 method was recorded. Because of the low dust loading on elevated surfaces, 2-3 dust samples  
145 from elevated surfaces were combined into one sample for analysis, yielding a total of 193  
146 samples. Figures S1, S2, and S3 illustrate the room contents of Home 1, Home 2 and Home 3  
147 respectively, showing both floor dust sample areas (F1 and F2) and elevated surface dust  
148 sample locations (ES).

## 149 150 **2.2. Analytical methods**

151 PBDEs and NBFRs in dust samples were analysed following the same extraction and clean-  
152 up methods as reported in our previous study (Al-Omran and Harrad, 2017). Briefly,  
153 accurately weighted aliquots of dust (~0.1 g) were spiked with a mixture of internal standards  
154 (20 ng of BDE-77, BDE-128, <sup>13</sup>CBTBPE, <sup>13</sup>CBEH-TEBP, and 40 ng of <sup>13</sup>CBDE-209) and  
155 extracted with *n*-hexane: acetone (3:1 v/v) using an ultrasonic extraction method.  
156 Concentrated crude sample extracts were purified involving two steps. In the first step, the  
157 extract was fractionated into two fractions (Fraction 1 and Fraction 2) using a 2 g Florisil SPE  
158 cartridge. Fraction 1 (containing PBDEs, DBDPE and PBEB) was eluted with *n*-hexane and  
159 fraction 2 (containing the rest of the targeted NBFRs) was eluted with ethyl acetate. A second  
160 purification steps were conducted on acid silica (44% w/w) for fraction 1 and aminopropyl  
161 functionalised silica for fraction 2. The both fractions were eluted with *n*-hexane/DCM (1:1,  
162 v/v) and combined then evaporated to incipient dryness, before resolubilisation in 100 µL of  
163 iso-octane containing PCB-129 at 250 pg/µL ready for GC/MS analysis. Target PBDEs and  
164 NBFRs were quantified using a gas chromatograph (GC) (Trace 1310 Gas Chromatograph)  
165 coupled to a mass spectrometer (MS) (ISQ Quadrupole MS); both (Thermo Fisher Scientific,  
166 USA). The GC was equipped with a programmable temperature vaporiser (PTV) injector and  
167 fitted with a capillary fused silica column (RESTEK, USA, 15 m x 0.25 mm inner diameter,

296  
297  
298 168 0.25  $\mu\text{m}$  film thickness). The MS was operated in the electron capture negative ion (ECNI)  
299  
300 169 mode.

### 302 170 **2.3 Quality assurance/Quality control**

304 171 To avoid any degradation that may occur via exposure to light, glassware and the turbo vap  
305  
306 172 instrument were covered with aluminium foil. To assess any possible contamination during  
307  
308 173 sample preparation and analysis method, one laboratory blank was processed in parallel with  
309  
310 174 every set of 6 dust samples and one quality control sample (NIST SRM 2585, organics in  
311  
312 175 indoor dust) was processed with every 20 real dust samples. Limits of detection (LOD) were  
313  
314 176 estimated based on a signal to noise ratio 3:1 and limits of quantification (LOQ) were  
315  
316 177 estimated based on signal to noise ratio 10:1. Field blanks ( $n = 9$ ) were also conducted to  
317  
318 178 assess any contamination contributed as a result of sampling, transport and storage of samples,  
319  
320 179 in addition to any introduced as a result of extraction and clean-up. The average of internal  
321  
322 180 standard recoveries in dust samples ranged from 78-90%.

### 321 181 **2.4. Statistical analysis**

322 182 Statistical analysis of our data was performed using Microsoft Excel 2013 and IBM SPSS  
323  
324 183 statistics software (V. 20). Within-room spatial variability in concentrations of PBDEs and  
325  
326 184 NBFRs was evaluated using a paired t-test applied to samples: a) taken from two different  
327  
328 185 floor areas; and b) taken from elevated surfaces and floors. Within- home spatial variability  
329  
330 186 was tested on samples taken from three different rooms in the same home via a repeated  
331  
332 187 measures ANOVA test. For the purposes of statistical evaluation, all concentrations below  
333  
334 188 LOQ were assigned a value of 0.5 LOQ. A  $p$  value  $< 0.05$  was taken to indicate statistical  
335  
336 189 significance. A Pearson correlation was used to test the relationship between concentrations  
337  
338 190 of BFRs ( $\text{ng/g}$ ) and dust loading ( $\text{g/m}^2$ ).

## 339 192 **3. RESULTS AND DISCUSSION**

### 341 193 **3.1. Concentrations of PBDEs and NBFRs in indoor dust samples**

343 194 In the three investigated homes, the detection frequencies of BDE-209 and BEH-TEBP were  
344  
345 195 100%, followed by DBDPE with 100%, 97% and 94% in Home 1, Home 2 and Home 3  
346  
347 196 respectively. Only those BFRs ( $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  
348  
349 197  $\Sigma_5$ NBFRs) displaying detection frequencies  $\geq 90\%$  were taken into account for statistical  
350  
351 198 summary.  $\Sigma_7$ tri-hepta-BDEs refers to the summation of seven congeners (BDE-28, BDE-47,

355  
356  
357 199 BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183),  $\Sigma_5$ NBFRs represent the sum of  
358  
359 200 PBEB, EH-TBB, BTBPE, BEH-TEBP, and DBDPE with  $\Sigma$ BFRs equalling the sum of  $\Sigma_7$ tri-  
360  
361 201 hepta-BDEs, BDE-209 and  $\Sigma_5$ NBFRs. Among all target BFRs, BDE-209 was predominant,  
362  
363 202 making average percentage contributions to  $\Sigma$ BFRs of 92.3%, 90.9%, and 62.8% in H1, H2  
364  
365 203 and H3 respectively. The high relative abundance of BDE-209 is not surprising, as Deca-BDE  
366  
367 204 was used extensively in the UK (Harrad et al., 2008a: 2008b). The next most abundant was  
368  
369 205  $\Sigma_5$ NBFRs making average percentage contributions of 6.6%, 7.8% and 36.7% in H1, H2 and  
370  
371 206 H3 respectively.  $\Sigma_7$ tri-hepta-BDEs made the lowest average percentage contributions of our  
372  
373 207 target BFRs; specifically 1.1%, 1.3% and 0.5% of  $\Sigma$ BFRs in H1, H2 and H3 respectively.  
374  
375 208 Table 1 lists average concentrations and relative standard deviation values (RSD) of  $\Sigma_7$ tri-  
376  
377 209 hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust samples from two floor  
378  
379 210 areas (F1 and F2) and elevated surface dust (ES) from the three rooms (R1, R2 and R3) of  
380  
381 211 three homes (H1, H2 and H3) during a nine-month sampling period. Figure S4 displays  
382  
383 212 distribution profiles of our target compounds.  
384  
385 213

### 381 214 **3.2 Within-room spatial variation of PBDEs and NBFRs in floor dust from two** 382 383 215 **different areas.**

384 216 In dust samples taken from different floor areas within the same room in nine rooms, no  
385  
386 217 significant difference in BDE-209 concentrations was observed, while  $\Sigma_7$ tri-hepta-BDEs (in  
387  
388 218 three rooms), BEH-TEBP (in one room) DBDPE and  $\Sigma_5$ NBFRs (in two rooms) were  
389  
390 219 significantly ( $p < 0.05$ ) different between different floor areas. Where observed, such spatial  
391  
392 220 variability in BFR concentrations is likely driven by varying distances from potential emission  
393  
394 221 sources, which are influenced by room dimensions, For instance, in the bedroom of Home 1,  
395  
396 222 concentrations of BEH-TEBP, DBDPE and consequently  $\Sigma_5$ NBFRs in samples from F1  
397  
398 223 exceeded significantly those from F2, with  $p$  values of 0.012, 0.053 and 0.006 respectively. As  
399  
400 224 shown in Figure S1 (H1R2), F1 is the rug area closest to the iron, foam chair, and the curtain,  
401  
402 225 while F2 is the bare floor area located closest to the door and further away ( $\approx 3$  m) from these  
403  
404 226 potential emission sources. Figure 1 illustrates average concentrations of  $\Sigma_7$ tri-hepta-BDEs,  
405  
406 227 BDE-209, BEH-TEBP and DBDPE in floor areas F1 and F2 in the three rooms (R1, R2 and  
407  
408 228 R3) of Home 1, Home 2 and Home 3, along with standard deviation (y error bar). Table S5  
409  
410 229 shows  $p$  values obtained from t-test comparison of concentrations of our target compounds in  
411  
412 230 floor dust samples within the same room. These data indicate that dust from a single area  
413



414  
415  
416 231 within a given room will likely not provide a representative measure of contamination in the  
417  
418 232 room overall.

419  
420 233

### 421 234 **3.3 Within-room spatial variation of PBDEs and NBFs between floor and elevated** 422 423 **surface dust**

424 236 Taking all 9 investigated rooms together, concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209,  
425 237 BEH-TEBP and  $\Sigma_5$ NBFs in elevated surface dust exceeded significantly ( $p < 0.001$ ) those  
426 238 in floor dust. The one exception to this is that concentrations of DBDPE in floor dust exceeded  
427 239 significantly ( $p = 0.015$ ) those in elevated surfaces. On an individual room basis,  
430 240 concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP and DBDPE (in 7, 4, 5 and 4 of  
431 241 9 rooms respectively) in dust samples from elevated surfaces exceeded significantly ( $p < 0.05$ )  
432 242 those from the floor in the same room. Figure 2 illustrates average concentrations of the target  
433 243 BFRs in floor dust and elevated surface dust in the three different rooms (R1, R2 and R3) of  
434 244 Home 1, Home 2 and Home 3. Table S6 shows  $p$  values obtained from t-test comparison of  
435 245 concentrations of our target compounds between elevated surface dust and floor dust samples.  
436 246 These results indicate that both floor and elevated surface dust should be considered for  
437 247 human exposure assessment, particularly for adults who likely are in contact with elevated  
438 248 surfaces more than the floor.

439  
440  
441  
442  
443  
444  
445 249

### 446 250 **3.4 Within-home spatial variation in concentrations of PBDEs and NBFs**

448 251 Among the nine rooms investigated, limited within-home variability in BFR concentrations  
449 252 between different rooms was observed, that is likely attributable to differences in the putative  
450 253 sources present in the rooms studied. In Home 1, only concentrations of BDE-209 in the  
451 254 bedroom (H1R2) exceeded significantly those in the living room (H1R1) with a  $p$  value of  
452 255 0.010, while for other BFRs, no significant differences were found between different rooms.  
453 256 In Home 2, concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, and DBDPE  
454 257 displayed significant differences between different rooms. Concentrations of  $\Sigma_7$ tri-hepta-  
455 258 BDEs in the study (H2R3) exceeded significantly ( $p = 0.050$ ) those in the living room (H2R1).  
456 259 In contrast, BDE-209 concentrations in the living room exceeded significantly ( $p = 0.033$ )  
457 260 those in the study, while BEH-TEBP and DBDPE concentrations in the study exceeded  
458 261 significantly those in the bedroom with  $p$  values of 0.041, 0.001 respectively. Meanwhile, in  
459 262 Home 3, significant differences were found between concentrations of BEH-TEBP in the two  
460 263 bedrooms and living room. BEH-TEBP concentrations in H3 fall in the order of: child's

473  
474  
475 264 bedroom > adult's bedroom > living room. The high levels in the bedrooms might be due to  
476  
477 265 the new mattresses that may have been treated with BEH-TEBP. However, there is no obvious  
478  
479 266 reason for the high concentrations of BEH-TEBP in the child's bedroom compared with  
480  
481 267 adult's bedroom. Figure 3 illustrates within-home spatial variability in concentrations of  $\Sigma_7$ tri-  
482  
483 268 hepta-BDEs, BDE-209 and BEH-TEBP and DBDPE in the three investigated homes.  
484  
485 269

### 485 270 **3.5 Temporal and seasonal variability in concentrations of BFRs in indoor dust.**

486  
487 271 The relative standard deviation of concentrations of individual BFRs in the 18 floor area  
488  
489 272 samples taken in each room ranged between 4% and 159%, and in the corresponding 9  
490  
491 273 elevated surface dust samples ranged between 9% and 117%. In both instances, these RSD  
492  
493 274 values exceeded those obtained from replicate analysis of SRM2585, which ranged from 9%  
494  
495 275 to 14%. This observed temporal variation in BFR concentrations is likely attributable to  
496  
497 276 concomitant changes in room contents with respect to putative sources of target BFRs.  $\Sigma_7$ tri-  
498  
499 277 hepta-BDEs concentrations were associated with the presence/absence of electronic devices  
500  
501 278 and old foam furniture, while those in BDE-209 were associated with carpets and fabric  
502  
503 279 materials. BEH-TEBP variability was associated with new bedroom furnishings, while  
504  
505 280 DBDPE temporal variability was not associated with any specific source. However, changes  
506  
507 281 in room contents did not explain the gradual decline in concentrations of BEH-TEBP in the  
508  
509 282 bedrooms of H3 over the first seven months of sampling. This might instead reflect gradual  
510  
511 283 attainment of equilibrium between the gas phase and particulate phase of this BFR in indoor  
512  
513 284 air. Figures S7, S8 and S9 illustrate the intra-room temporal variation in concentrations of  
514  
515 285  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE, and  $\Sigma_5$ NBFRs in dust from different floor  
516  
517 286 areas (F1 and F2) from different rooms (R1, R2 and R3) during the nine monitored months in  
518  
519 287 Home 1, Home 2 and Home 3 respectively. In addition, noticeable variation in maximum:  
520  
521 288 minimum BFR levels were found depending on a given area, particularly for  $\Sigma_7$ tri-hepta-  
522  
523 289 BDEs and DBDPE. The ratio of maximum: minimum concentrations of  $\Sigma_7$ tri-hepta-BDEs  
524  
525 290 were 30, 24 and 21 in areas H1R2F1, H1R2F2 and H1R3F1 respectively, and for DBDPE  
526  
527 291 were 28, 71, 61, 43 and 42 in areas H2R1F2, H3R1F1, H3R2F1, H3R3F1, and H3R3F2,  
528  
529 292 respectively. Table S10 lists maximum: minimum concentration ratios of these compounds in  
530  
531 293 floor areas.  
532  
533 294

525 295 Noticeable seasonal variability in BFR concentrations was also observed between colder and  
526  
527 296 warmer seasons. In 13 out of 17 floor areas, average concentrations of  $\Sigma_8$ tri-deca-BDEs were

532  
533  
534  
535  
536  
537  
538  
539  
540  
541  
542  
543  
544  
545  
546  
547  
548  
549  
550  
551  
552  
553  
554  
555  
556  
557  
558  
559  
560  
561  
562  
563  
564  
565  
566  
567  
568  
569  
570  
571  
572  
573  
574  
575  
576  
577  
578  
579  
580  
581  
582  
583  
584  
585  
586  
587  
588  
589  
590

297 higher in colder seasons than warmer, while in the same number of floor locations,  $\Sigma_5$ NBFRs  
298 were higher in warmer seasons, with the exception of DBDPE. In general, average  
299 concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209 and BEH-TEBP in elevated surface dust  
300 samples were higher in warmer seasons than in colder, while in floor dust, average  
301 concentrations of BDE-209 were comparable in both colder and warmer seasons. With the  
302 exception of  $\Sigma_8$ tri-deca-BDEs in two floor areas and  $\Sigma_5$ NBFRs in four floor areas, no  
303 significant differences in concentrations of these two groups were apparent between warmer  
304 and colder seasons. Higher concentrations in colder seasons were only observed for BDE-209  
305 and DBDPE, which might be due to the low vapour pressure of these compounds which  
306 facilitate partitioning to indoor dust, which will be more favoured at lower temperatures.

### 3.6 The relationship between the BFR dust concentration and dust loading

309 To test the relationship between BFR dust concentration (ng/g) and dust loading (g/m<sup>2</sup>), we  
310 used our data addressing temporal variability in BFR concentrations in dust from Home 1,  
311 Home 2, and Home 3. The Pearson correlation showed a significant negative correlation  
312 between the logarithms of BFR concentrations and dust loadings for Home 2 and Home 3 for  
313 BDE-99 ( $R = 0.675, p = 0.046$ ) and  $\Sigma_7$ tri-hepta-BDEs ( $R = 0.760, p = 0.018$ ) in H2R2F2 and  
314 for BEH-TEBP ( $R = 0.749, p = 0.020$ ) in H3R2F2. In other words, in three out of seventeen  
315 individual floor areas, concentrations of lower brominated compounds (i.e. BDE-99 and  $\Sigma_6$ tri-  
316 hepta-BDEs) and BEH-TEBP decreased as dust loading increased. This implies that  
317 “dilution” has occurred in these rooms due to the high dust loading and indicates that the  
318 source of these compounds and of indoor dust are independent. However, in one sampled  
319 area, a positive correlation between DBDPE concentration and dust loading suggested the  
320 source(s) of both dust and DBDPE in that area to be the same, implying that DBDPE enters  
321 indoor dust via abrasion of fibres or particles from a putative source.

### 3.7 The impact of spatial and temporal variability on human exposure assessments

324 To evaluate the extent to which human exposure to our target contaminants via dust ingestion  
325 are affected by spatial variability, we compared the mean  $\pm$  SD concentration in dust samples  
326 collected from: 1) different floor areas in the same room, 2) elevated surfaces and floor in the  
327 same room and 3) different rooms in the same home. As observed in Figure 1, substantial  
328 differences are apparent in concentrations of BFRs between the two floor areas (F1 and F2),  
329 particularly for  $\Sigma_7$ tri-hepta-BDEs and DBDPE. For example, in H2R2, concentrations of

591  
592  
593  
594  
595  
596  
597  
598  
599  
600  
601  
602  
603  
604  
605  
606  
607  
608  
609  
610  
611  
612  
613  
614  
615  
616  
617  
618  
619  
620  
621  
622  
623  
624  
625  
626  
627  
628  
629  
630  
631  
632  
633  
634  
635  
636  
637  
638  
639  
640  
641  
642  
643  
644  
645  
646  
647  
648  
649

330  $\Sigma_7$ tri-hepta-BDEs in floor area F2 (average  $\pm$  SD =  $62 \pm 17$  ng/g) exceed substantially those  
331 in floor area F1 (average  $\pm$  SD =  $27 \pm 17$  ng/g). In this room, F1:F2 = 61:4 for one sampling  
332 event, implying that exposure assessment in that room could vary by a factor of 15 depending  
333 on the sampling area. In addition, substantial within-room spatial variability in BFR  
334 concentrations was observed between elevated surface dust and floor dust in the nine rooms  
335 studied (Figure 2). To illustrate, in H3R1, BEH-TEBP concentrations in elevated surface dust  
336 (average  $\pm$  SD =  $4187 \pm 2004$  ng/g) exceeded substantially those in floor dust (average  $\pm$  SD  
337 =  $1196 \pm 301$  ng/g), with ESD:FD  $\sim$ 5 during 1 sampling event. Moreover, BFR  
338 concentrations in separate rooms in the same house can differ quite markedly (Figure 3). For  
339 example, concentrations of BEH-TEBP in H3R3 (average  $\pm$  SD =  $3992 \pm 1906$  ng/g)  
340 exceeded those in H3R1 (average  $\pm$  SD =  $1811 \pm 1498$  ng/g). Due to this substantial within-  
341 room and within-home spatial variability, exposure estimates based on dust taken from one  
342 specific floor area, floor surface only or one room alone are subject to uncertainty.

343  
344 To assess the extent to which temporal and seasonal variability may affect human exposure  
345 assessment, we compared the RSD values for selected BFRs and examined the extremes of  
346 exposure assessment using maximum: minimum concentration ratios for a given room. Our  
347 findings highlighted uncertainties in exposure assessments for BFRs based on a single dust  
348 sample taken from a given area at a given point in time. In Home 1, the highest RSD values  
349 of  $\Sigma_7$ tri-hepta-BDEs were 92%, 86% and 123%, observed in H1R2F1, H1R2F2 and H1R3F1  
350 respectively. This implies that human exposure to  $\Sigma_7$ tri-hepta-BDEs via contact with dust in  
351 these areas would vary to the same extent. In addition, in these same floor areas,  $\Sigma_7$ tri-hepta-  
352 BDE maximum: minimum ratios were 30, 24, and 21 respectively, implying that exposure  
353 assessments could be underestimated or overestimated by factors of 30, 24, and 21 if by  
354 chance one sample was taken from these areas in the month recording the lowest concentration  
355 as opposed to the month when the highest concentration was recorded. The highest RSD value  
356 for BEH-TEBP (73%) was found in H1R1ES with the highest maximum: minimum ratio of  
357 9.2 in H2R1F2. Moreover, considerable temporal variation in concentrations of DBDPE were  
358 found in the three homes studied, particularly in Home 3. The RSD values of DBDPE in  
359 H3R2F2 and H3R3F2 were the highest among all BFRs, with values of 138% and 159%  
360 respectively.

361

650  
651  
652 362 The considerable temporal and seasonal variability observed in this study of just 9 rooms  
653  
654 363 indicates the uncertainty associated with basing exposure assessments via dust ingestion for  
655  
656 364 BFRs based on a single grab sample taken from a given area at a given point in time.  
657  
658 365

#### 659 366 **4. CONCLUSIONS AND RECOMMENDATIONS**

660 367 Substantial vertical spatial variations in BFR contamination indicate that both floor dust and  
661  
662 368 elevated surface dust should be considered for human exposure assessments, particularly for  
663  
664 369 adults who likely are in contact with elevated surfaces more than the floor. In addition, the  
665  
666 370 appreciable horizontal variations in BFR concentrations in floor dust, indicate that floor dust  
667  
668 371 samples should be taken from the most-frequented parts of the room in order to best reflect  
669  
670 372 human exposure. Our findings reveal substantial variability in the concentrations of some  
671  
672 373 BFRs during the sampling period. Temporal variations in BFR concentrations appear affected  
673  
674 374 by the addition or removal of a potential emission source. Our findings highlight the  
675  
676 375 uncertainty associated with assessments of exposure to BFRs based on a single dust sample  
677  
678 376 taken from a given area at a given point in time.

#### 679 377 680 378 **ACKNOWLEDGEMENTS**

681 379 The authors express their thanks to all the dust donors from Birmingham, UK. Layla Salih  
682  
683 380 Al-Omran acknowledges gratefully the Iraqi government for a PhD Scholarship, the Iraqi  
684  
685 381 Establishment of Martyrs for financial support and the Ministry of Higher Education and  
686  
687 382 Scientific Research for administrative support.  
688 383

#### 689 384 **REFERENCES**

690 385 Allen, J. G., M. D. McClean, H. M. Stapleton and T. F. Webster (2008). "Critical factors in  
691  
692 386 assessing exposure to PBDEs via house dust." Environment International **34**(8): 1085-1091.  
693  
694 387  
695 388 Al-Omran, L. S. and S. Harrad (2016a). "Polybrominated diphenyl ethers and "novel"  
696  
697 389 brominated flame retardants in floor and elevated surface house dust from Iraq: Implications  
698  
699 390 for human exposure assessment." Emerging Contaminants **2**(1): 7-13.  
700  
701 391  
702 392 Al-Omran, L. S. and S. Harrad (2016b). "Distribution pattern of legacy and "novel"  
703  
704 393 brominated flame retardants in different particle size fractions of indoor dust in Birmingham,  
705  
706 394 United Kingdom." Chemosphere **157**: 124-131.  
707  
708 395  
709 396 Al-Omran, L. S. and S. Harrad (2017). "Influence of sampling approach on concentrations of  
710  
711 397 legacy and "novel" brominated flame retardants in indoor dust." Chemosphere **178**: 51-58.  
712  
713 398

709  
710  
711  
712  
713  
714  
715  
716  
717  
718  
719  
720  
721  
722  
723  
724  
725  
726  
727  
728  
729  
730  
731  
732  
733  
734  
735  
736  
737  
738  
739  
740  
741  
742  
743  
744  
745  
746  
747  
748  
749  
750  
751  
752  
753  
754  
755  
756  
757  
758  
759  
760  
761  
762  
763  
764  
765  
766  
767

399 Cao, Z., F. Xu, A. Covaci, M. Wu, G. Yu, B. Wang, S. Deng and J. Huang (2014).  
400 "Differences in the seasonal variation of brominated and phosphorus flame retardants in office  
401 dust." Environment International **65**: 100-106.

402  
403 Cequier, E., A. C. Ionas, A. Covaci, R. M. Marcé, G. Becher and C. Thomsen (2014).  
404 "Occurrence of a Broad Range of Legacy and Emerging Flame Retardants in Indoor  
405 Environments in Norway." Environmental Science & Technology **48**(12): 6827-6835.

406  
407 Chevrier, J., K. G. Harley, A. Bradman, M. Gharbi, A. Sjödin and B. Eskenazi (2010).  
408 "Polybrominated Diphenyl Ether (PBDE) Flame Retardants and Thyroid Hormone during  
409 Pregnancy." Environmental Health Perspectives **118**(10): 1444-1449.

410  
411 Coakley, J. D., S. J. Harrad, E. Goosey, N. Ali, A.-C. Dirtu, N. Van den Eede, A. Covaci, J.  
412 Douwes and A. t. Marnettje (2013). "Concentrations of polybrominated diphenyl ethers in  
413 matched samples of indoor dust and breast milk in New Zealand." Environment International  
414 **59**: 255-261.

415  
416 EFSA, European Food Safety Authority (2012). "Scientific Opinion on Emerging and Novel  
417 Brominated Flame Retardants (BFRs) in Food." European Food Safety Authority Journal  
418 **10**(10):2908.

419  
420 European Commission (2012). "Recommendation from the Scientific Committee on  
421 Occupational Exposure Limits for Diphenyl ether, Octabromoderivative (commercial  
422 mixture)" (online). Available from:  
423 <http://ec.europa.eu/social/BlobServlet?docId=7721&langId=en>. (Accessed May 2016).

424  
425 Harrad, S., C. Ibarra, M. A.-E. Abdallah, R. Boon, H. Neels and A. Covaci (2008 a).  
426 "Concentrations of brominated flame retardants in dust from United Kingdom cars, homes,  
427 and offices: Causes of variability and implications for human exposure." Environment  
428 International **34**(8): 1170-1175.

429  
430 Harrad, S., C. Ibarra, M. Diamond, L. Melymuk, M. Robson, J. Douwes, L. Roosens, A. C.  
431 Dirtu and A. Covaci (2008b). "Polybrominated diphenyl ethers in domestic indoor dust from  
432 Canada, New Zealand, United Kingdom and United States." Environment international **34**(2):  
433 232-238.

434  
435 Harrad, S., M. A.-E. Abdallah and A. Covaci (2009). "Causes of variability in concentrations  
436 and diastereomer patterns of hexabromocyclododecanes in indoor dust." Environment  
437 international **35**(3): 573-579.

438  
439 Hoffman, K., S. Garantziotis, L. S. Birnbaum and H. M. Stapleton (2015). "Monitoring Indoor  
440 Exposure to Organophosphate Flame Retardants: Hand Wipes and House Dust." Environmental Health Perspectives **123**(2): 160-165.

441  
442  
443 Johnson, P. I., H. M. Stapleton, A. Sjödin and J. D. Meeker (2010). "Relationships between  
444 Polybrominated Diphenyl Ether Concentrations in House Dust and Serum." Environmental  
445 Science & Technology **44**(14): 5627-5632.

768  
769  
770  
771  
772  
773  
774  
775  
776  
777  
778  
779  
780  
781  
782  
783  
784  
785  
786  
787  
788  
789  
790  
791  
792  
793  
794  
795  
796  
797  
798  
799  
800  
801  
802  
803  
804  
805  
806  
807  
808  
809  
810  
811  
812  
813  
814  
815  
816  
817  
818  
819  
820  
821  
822  
823  
824  
825  
826

447 Johnson, P. I., H. M. Stapleton, B. Mukherjee, R. Hauser and J. D. Meeker (2013).  
448 "Associations between brominated flame retardants in house dust and hormone levels in  
449 men." Science of the total environment **445**: 177-184.

451 Kang, Y., H. S. Wang, K. C. Cheung and M. H. Wong (2011). "Polybrominated diphenyl  
452 ethers (PBDEs) in indoor dust and human hair." Atmospheric Environment **45**(14): 2386-  
453 2393.

455 Kuang, J., Y. Ma and S. Harrad (2016). "Concentrations of "legacy" and novel brominated  
456 flame retardants in matched samples of UK kitchen and living room/bedroom dust."  
457 Chemosphere **149**: 224-230.

459 Li, M., Z. Liu, L. Gu, R. Yin, H. Li, X. Zhang, T. Cao and C. Jiang (2014). "Toxic effects of  
460 decabromodiphenyl ether (BDE-209) on human embryonic kidney cells." Frontiers in  
461 Genetics **5**: 118.

463 Mankidy, R., Ranjan, B., Honaramooz, A., & Giesy, J. P. (2014). Effects of novel brominated  
464 flame retardants on steroidogenesis in primary porcine testicular cells. Toxicology  
465 letters **224**(1), 141-146.

467 Mariani, A., Fanelli, R., Depaolini, A., & Paola, M. (2015). Decabrominated diphenyl ether  
468 and methylmercury impair fetal nervous system development in mice at documented human  
469 exposure levels. Developmental neurobiology **75**(1), 23-38.

471 Muenhor, D. and S. Harrad (2012). "Within-room and within-building temporal and spatial  
472 variations in concentrations of polybrominated diphenyl ethers (PBDEs) in indoor dust."  
473 Environment international **47**: 23-27.

475 NICNAS, National Industrial Chemicals Notification and Assessment Scheme (2007).  
476 (online). Available from: [https://www.nicnas.gov.au/\\_data/assets/pdf\\_file/0003/4944/Final-Interim-Report-PBDE-March.pdf](https://www.nicnas.gov.au/_data/assets/pdf_file/0003/4944/Final-Interim-Report-PBDE-March.pdf). (Accessed January 2016).

479 Noyes P. D., S. M. Kelly, C. L. Mitchelmore, H. M. Stapleton. (2010) "Characterizing the in  
480 vitro hepatic biotransformation of the flame retardant BDE 99 by common carp". Aquatic  
481 Toxicology **97**(2): 142-150.

483 Stapleton, H. M., N. G. Dodder, J. H. Offenber, M. M. Schantz and S. A. Wise (2005).  
484 "Polybrominated diphenyl ethers in house dust and clothes dryer lint." Environmental science  
485 & technology **39**(4): 925-931.

487 Stapleton, H. M., Eagle, S., Sjödin, A., & Webster, T. F. (2012). Serum PBDEs in a North  
488 Carolina toddler cohort: associations with handwipes, house dust, and socioeconomic  
489 variables. Environmental health perspectives **120**(7): 1049.

491 Tang, L., B. Lei, G. Xu, J. Ma, J.-Q. Lei, S.-Q. Jin, G.-Y. Hu and M.-H. Wu (2013).  
492 "Polybrominated Diphenyl Ethers in Human Hair from the College Environment: Comparison  
493 with Indoor Dust." Bulletin of Environmental Contamination and Toxicology **91**(4): 377-381.

495 Toms, L.-M. L., A. Sjödin, F. Harden, P. Hobson, R. Jones, E. Edenfield and J. F. Mueller  
496 (2009a). "Serum Polybrominated Diphenyl Ether (PBDE) Levels Are Higher in Children (2–5

827  
828  
829  
830  
831  
832  
833  
834  
835  
836  
837  
838  
839  
840  
841  
842  
843  
844  
845  
846  
847  
848  
849  
850  
851  
852  
853  
854  
855  
856  
857  
858  
859  
860  
861  
862  
863  
864  
865  
866  
867  
868  
869  
870  
871  
872  
873  
874  
875  
876  
877  
878  
879  
880  
881  
882  
883  
884  
885

497 Years of Age) than in Infants and Adults." *Environmental Health Perspectives* **117**(9): 1461-  
498 1465.

500 USEPA. US Environmental Protection Agency (2006). "Toxicological review of 2,2', 4,4', 5-  
501 Pentabromodiphenyl ether (BDE-99)" (online). Available from:  
502 [https://ofmpub.epa.gov/eims/eimscomm.getfile?p\\_download\\_id=474496](https://ofmpub.epa.gov/eims/eimscomm.getfile?p_download_id=474496) (Accessed  
503 February 2016).

505 USEPA. US Environmental Protection Agency (2008a). "Toxicological Review of 2,2',4,4'-  
506 Tetrabromodiphenyl ether (BDE-47)" (online). Available from:  
507 <http://www.epa.gov/iris/toxreviews/1010tr.pdf> (Accessed May 2016).

509 USEPA. US Environmental Protection Agency (2008b). "Toxicological Review of  
510 2,2',4,4',5,5'-Hexabromodiphenyl ether (BDE-153)" (online). Available from:  
511 <http://www.epa.gov/iris/toxreviews/1009tr.pdf> (Accessed May 2016).

513 USEPA. US Environmental Protection Agency (2008c). "Toxicological Review of  
514 Decabromodiphenyl ether (BDE-209)" (online). Available from:  
515 <http://www.epa.gov/iris/toxreviews/0035tr.pdf> . (Accessed April 2016).

517 Venier, M., O. Audy, Š. Vojta, J. Bečanová, K. Romanak, L. Melymuk, M. Krátká, P.  
518 Kukučka, J. Okeme, A. Saini, M. L. Diamond and J. Klánová (2016). "Brominated flame  
519 retardants in the indoor environment — Comparative study of indoor contamination from  
520 three countries." *Environment International* **94**: 150-160.

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

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

531 Yu, Y.-X., Y.-P. Pang, C. Li, J.-L. Li, X.-Y. Zhang, Z.-Q. Yu, J.-L. Feng, M.-H. Wu, G.-Y.  
532 Sheng and J.-M. Fu (2012). "Concentrations and seasonal variations of polybrominated  
533 diphenyl ethers (PBDEs) in in- and out-house dust and human daily intake via dust ingestion  
534 corrected with bioaccessibility of PBDEs." *Environment International* **42**: 124-131.



886  
887  
888  
889  
890  
891  
892  
893  
894  
895  
896  
897  
898  
899  
900  
901  
902  
903  
904  
905  
906  
907  
908  
909  
910  
911  
912  
913  
914  
915  
916  
917  
918  
919  
920  
921  
922  
923  
924  
925  
926  
927  
928  
929  
930  
931  
932  
933  
934  
935  
936  
937  
938  
939  
940  
941  
942  
943  
944

543 **Table 1: Average concentrations (ng/g) and relative standard deviation (RSD) of  $\Sigma_7$ tri-**  
 544 **hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in indoor dust from two**  
 545 **floor areas (F1 and F2) and elevated surface (ES) dust samples in the three rooms (R1,**  
 546 **R2 and R3) over nine monitored months of three homes (H1, H2 and H3)**

Sampling area	$\Sigma_7$ tri-hepta-BDEs		BDE-209		BEH-TEBP		DBDPE		$\Sigma_5$ NBFRs	
	Average	RSD	Average	RSD	Average	RSD	Average	RSD	Average	RSD
H1R1F1	21	44	2061	29	93	23	40	64	151	22
H1R1F2	18	35	1901	45	85	16	42	60	142	27
H1R1ES	70	28	3679	22	323	73	131	36	540	50
H1R2F1	23	92	3342	51	127	13	71	80	216	35
H1R2F2	34	86	2786	24	106	19	41	50	159	27
H1R2ES	128	31	6506	25	168	27	90	117	268	49
H1R3F1	22	123	2334	30	110	35	70	80	207	40
H1R3F2	17	50	2777	12	66	29	29	62	113	13
H1R3ES	79	48	6572	42	225	13	78	61	365	17
H2R1F1	31	36	3414	27	120	21	130	72	263	42
H2R1F2	30	31	3123	28	105	40	102	78	217	53
H2R1ES	111	26	7269	40	445	65	56	62	523	60
H2R2F1	27	64	2687	17	113	27	92	56	231	32
H2R2F2	62	28	2947	24	111	55	117	57	247	48
H2R2ES	83	35	6675	41	135	32	27	96	186	36
H2R3F1	48	29	2672	19	120	15	134	50	265	27
H2R3F2	36	34	2924	21	122	12	274	38	411	25
H2R3ES	126	31	4309	16	428	46	49	103	502	42
H3R1F1	33	38	5639	94	1371	40	163	104	1553	35
H3R1F2	30	61	4403	39	926	49	37	80	976	46
H3R1ES	64	55	3568	11	4187	48	11	91	4274	47
H3R2F1	18	71	4252	6	2486	35	95	138	2622	32
H3R2F2	36	54	4129	9	2362	50	69	78	2462	47
H3R2ES	83	87	8451	25	5397	34	45	40	5635	32
H3R3F1	37	43	4498	4	3046	33	109	112	3199	29
H3R3F2	25	36	4401	5	3044	32	116	159	3201	28
H3R3ES	57	23	7138	30	7049	9	48	82	7559	5

945  
946  
947  
948  
949  
950  
951  
952  
953  
954  
955  
956  
957  
958  
959  
960  
961  
962  
963  
964  
965  
966  
967  
968  
969  
970  
971  
972  
973  
974  
975  
976  
977  
978  
979  
980  
981  
982  
983  
984  
985

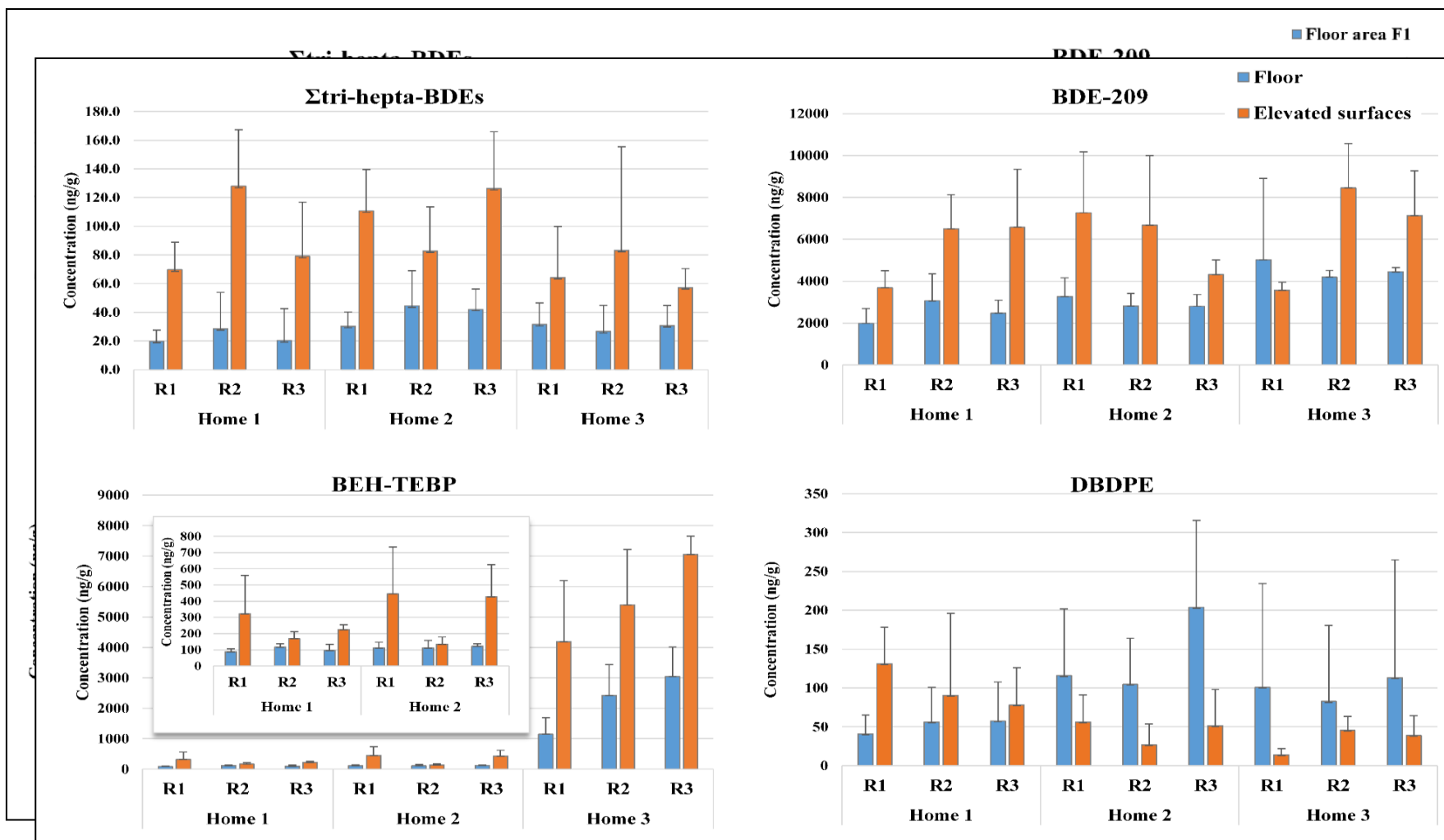


Figure 2: Average concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP and DBDPE in floor dust and elevated surface dust from different rooms (R1 = Living room, R2= Bedroom, and R3 = Study, except in Home 3= Bedroom) in Home 1, Home 2 and Home 3

986  
987  
988  
989  
990  
991  
992  
993  
994  
995  
996  
997  
998  
999  
1000  
1001  
1002  
1003  
1004  
1005  
1006  
1007  
1008  
1009  
1010  
1011  
1012  
1013  
1014  
1015  
1016  
1017  
1018  
1019  
1020  
1021  
1022  
1023  
1024  
1025  
1026

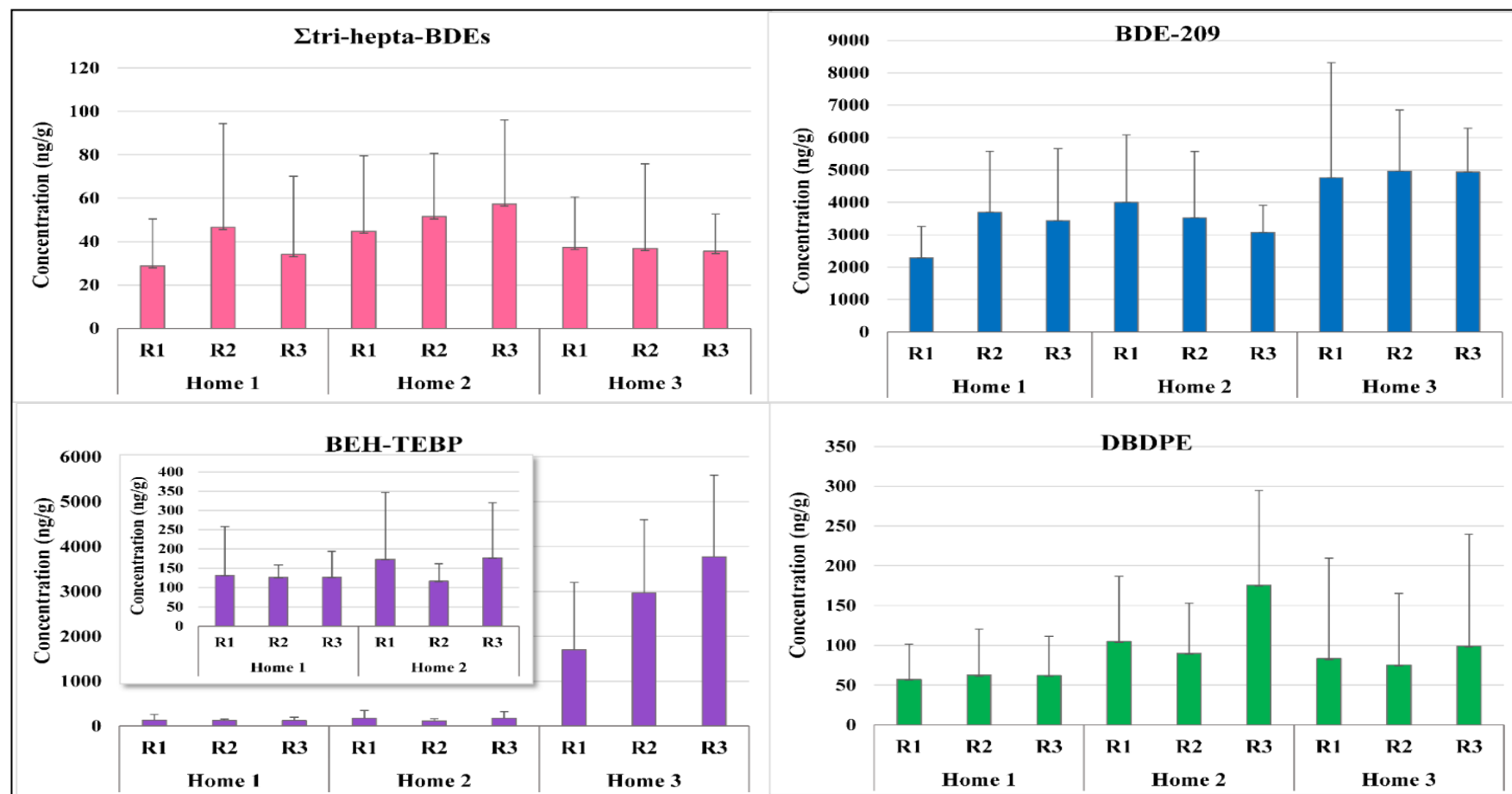


Figure 3: Average concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP and DBDPE in dust from different rooms (R1 = Living room, R2= Bedroom, and R3 = Study, Home 3 = Bedroom) within the same home in Home 1, Home 2 and Home 3

# WITHIN-ROOM AND WITHIN-HOME SPATIAL AND TEMPORAL VARIABILITY IN CONCENTRATIONS OF LEGACY AND “NOVEL” BROMINATED FLAME RETARDANTS IN INDOOR DUST

Layla Salih Al-Omran <sup>a,b</sup> and Stuart Harrad<sup>a</sup>

<sup>a</sup>School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, UK

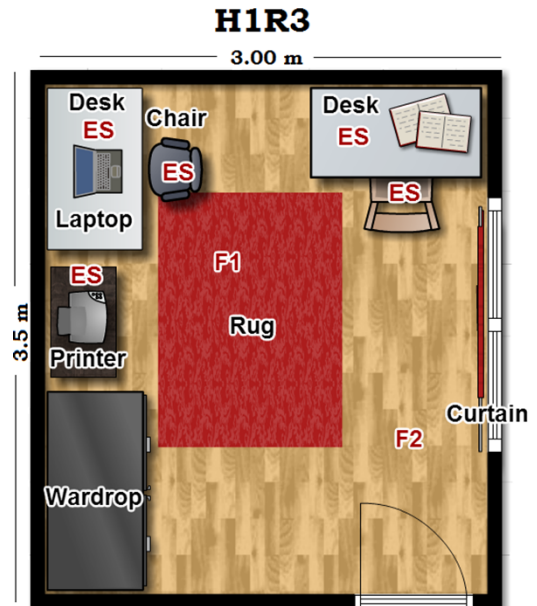
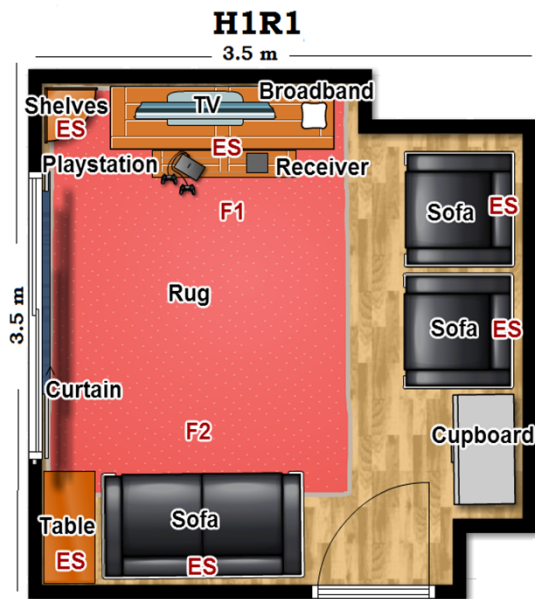
<sup>b</sup>Division of Ecology, College of Science, University of Basrah, Basrah, Iraq

## Electronic supporting information contains:

- Figure S1: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H1R1), bedroom (H1R2) and study room (H1R3) of Home 1
- Figure S2: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H2R1), bedroom (H2R2) and study room (H2R3) of Home 2
- Figure S3: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H3R1), adult bedroom (H3R2) and a child's bedroom (H3R3) of Home 3
- Figure S4: Average concentrations (ng/g) and distribution profiles of tri-hepta-BDEs and NBRs in Home 1, Home 2 and Home 3
- Table S5: *p* values obtained from the T-test comparison of concentrations of BFRs between the two floor areas (F1 and F2) within the same room.
- Table S6: *p* values obtained from the T-test comparison of concentrations of BFRs between the elevated surface dust and floor dust within the same room.
- Figure S7: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = bedroom and R3 = study) of Home 1

- Figure S8: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = bedroom and R3 = study) of Home 2
- Figure S9: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = adult bedroom and R3 = child's bedroom) of Home 3
- Table S10: Maximum: minimum ratio in concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in floor dust samples (F1 and F2) from three rooms (R1, R2 and R3) in Home1, Home2 and Home3 (H1, H2 and H3)

**Figure S1: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H1R1), bedroom (H1R2) and study room (H1R3) of Home 1**



**Figure S2: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H2R1), bedroom (H2R2) and study room (H2R3) of Home 2**

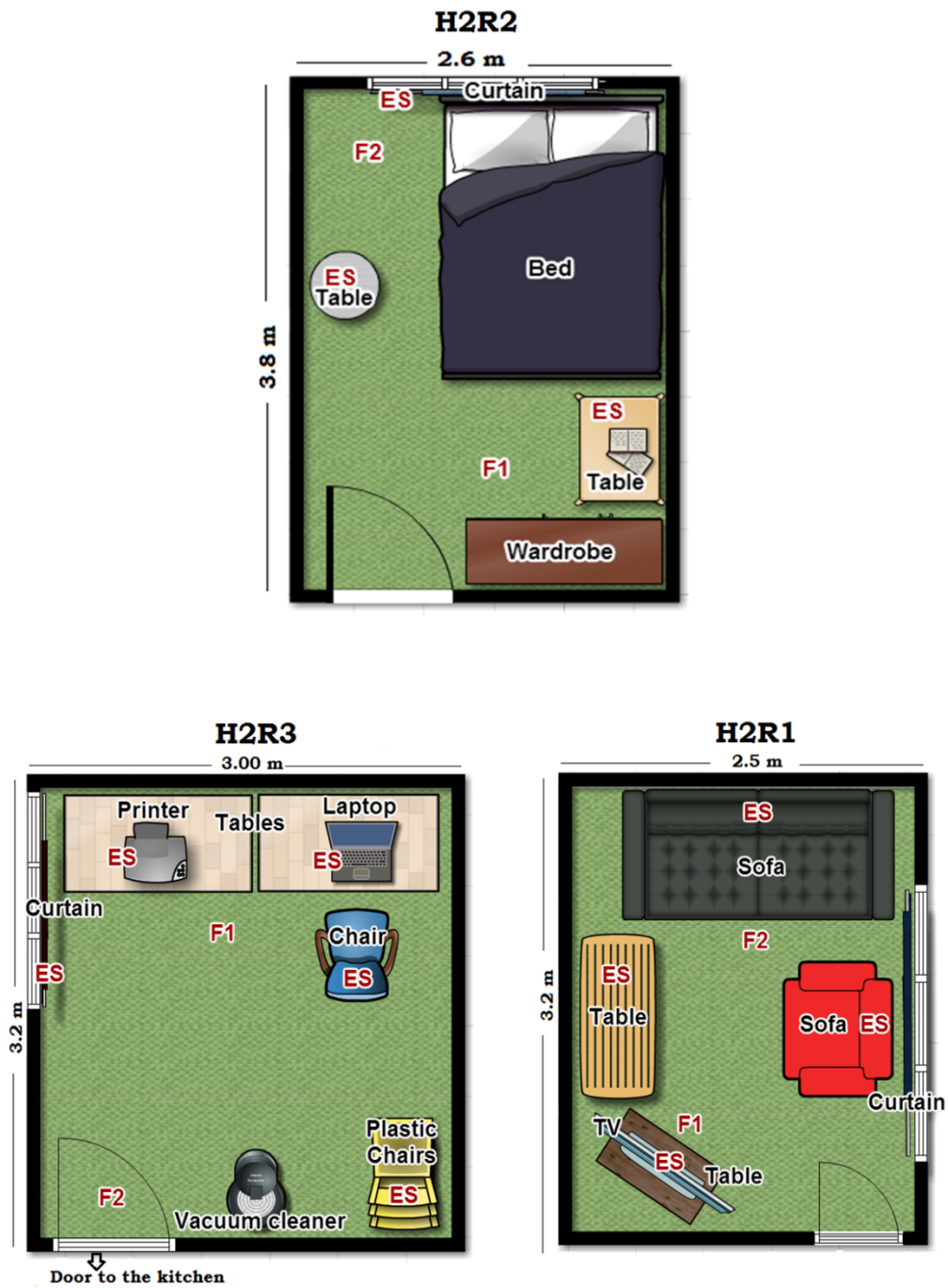
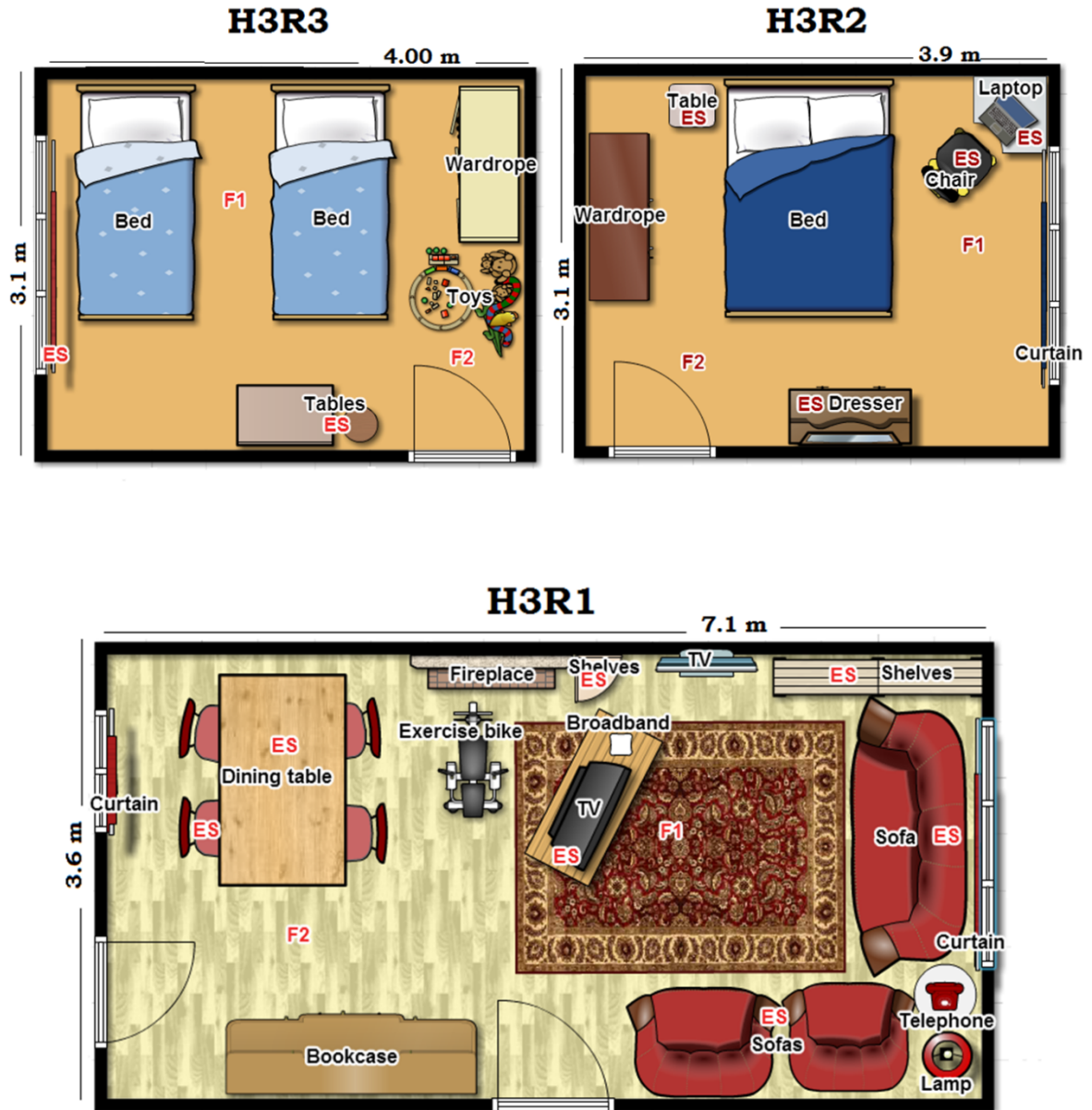
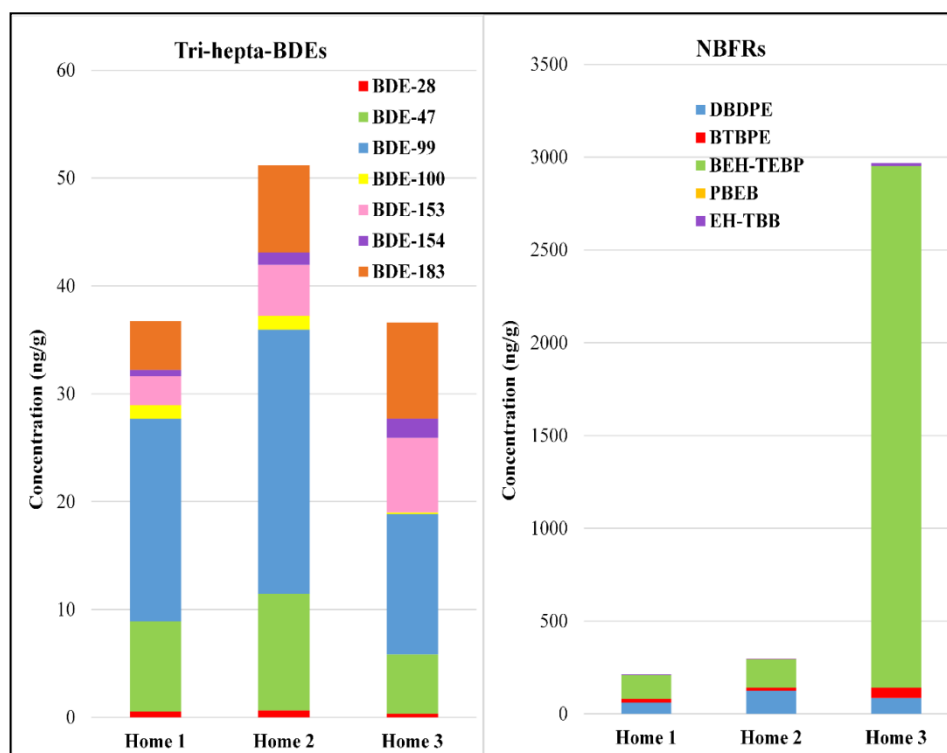


Figure S3: Room contents and sampling locations relative to floor (F1 and F2) and elevated surface (ES) dust sampling locations in the living room (H3R1), adult bedroom (H3R2) and a child's bedroom (H3R3) of Home 3





**Figure S4: Average concentrations (ng/g) and distribution profiles of tri-hepta-BDEs and NBRs in Home 1, Home 2 and Home 3**



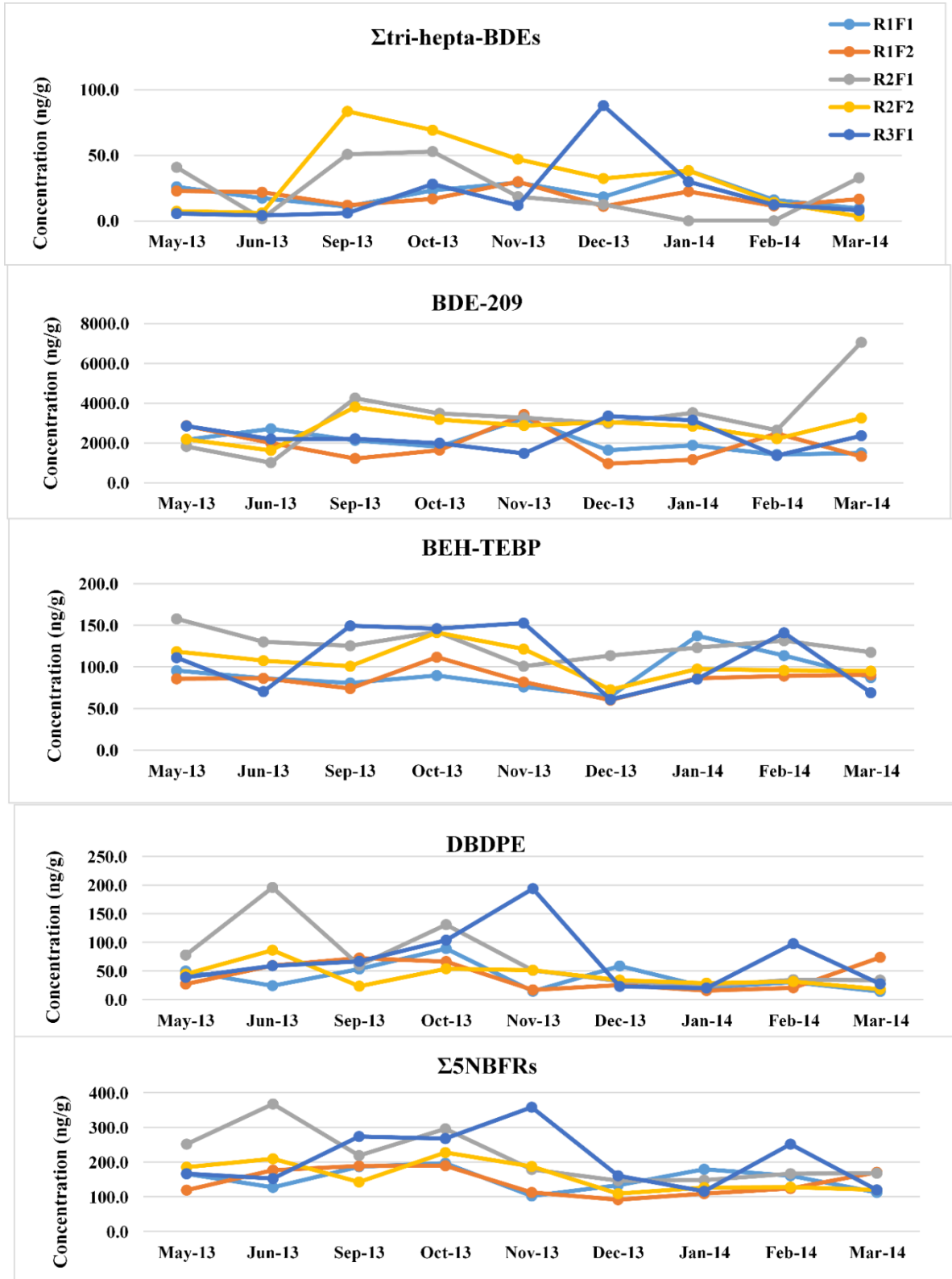
**Table S5: *p* values obtained from the T-test comparison of concentrations of BFRs between the two floor areas (F1 and F2) within the same room.**

Sampling room	$\Sigma_7$ tri-hepta	BDE-209	BEH-TEBP	DBDPE	NBRs
H1R1	0.277	0.508	0.323	0.81	0.55
H1R2	0.269	0.232	0.012	0.054	0.006
H1R3	0.359	0.576	0.411	0.613	0.285
H2R1	0.575	0.247	0.219	0.438	0.335
H2R2	0.0003	0.233	0.939	0.217	0.557
H2R3	0.006	0.109	0.561	0.001	> 0.001
H3R1	0.71	0.341	0.102	0.055	0.024
H3R2	0.052	0.347	0.66	0.405	0.572
H3R3	0.071	0.244	0.994	0.785	0.992

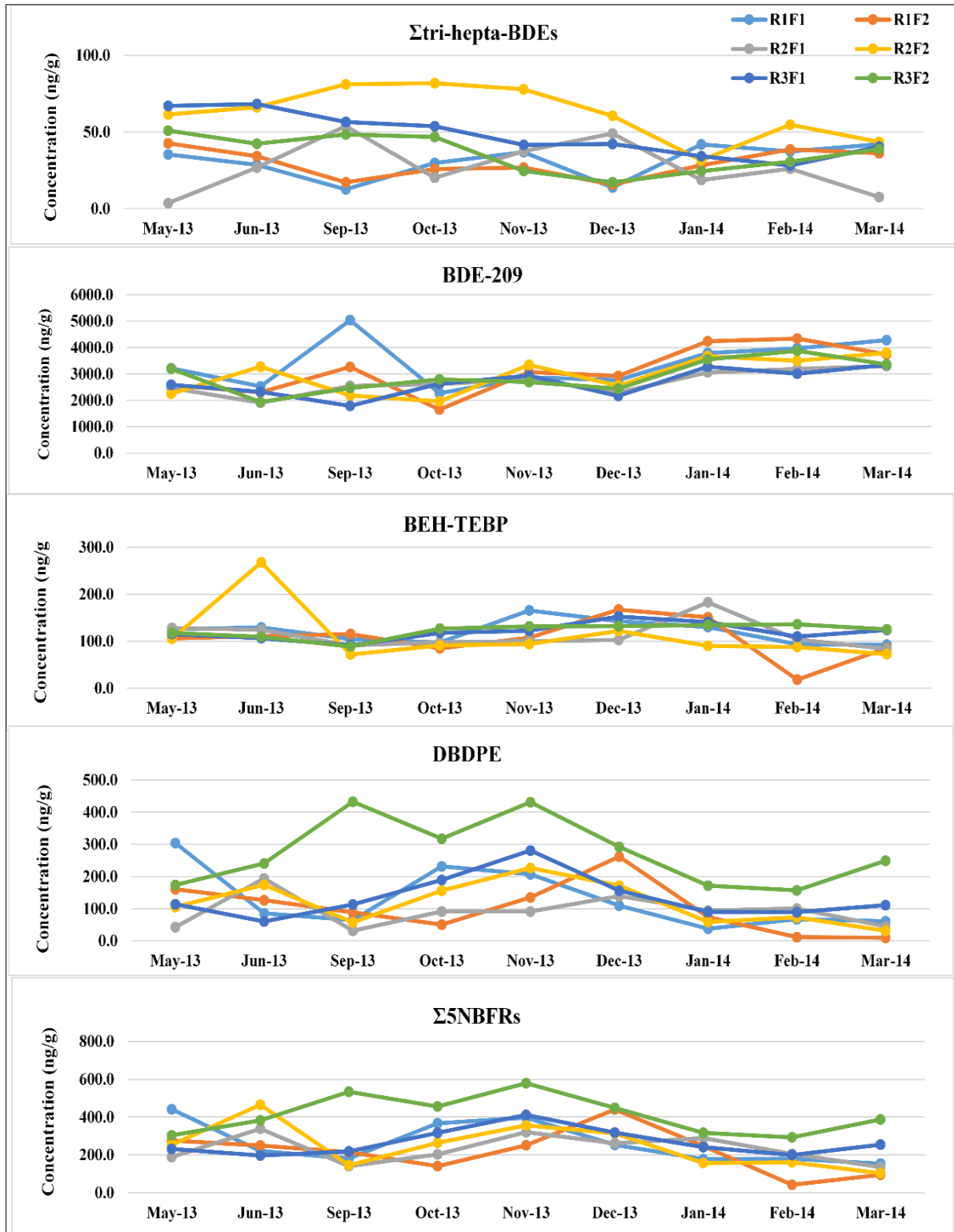
**Table S6: *p* values obtained from the T-test comparison of concentrations of BFRs between the elevated surface dust and floor dust within the same room.**

Sampling room	$\Sigma_7$ tri-hepta	BDE-209	BEH-TEBP	DBDPE	NBFRs
H1R1	0.022	0.045	0.138	0.026	0.062
H1R2	0.031	0.042	0.071	0.573	0.3
H1R3	0.046	0.058	0.029	0.398	0.003
H2R1	0.007	0.089	0.096	0.108	0.11
H2R2	0.056	0.107	0.68	0.012	0.199
H2R3	0.041	0.013	0.048	0.002	0.205
H3R1	0.15	0.354	0.047	0.016	0.056
H3R2	0.201	0.03	0.04	0.393	0.037
H3R3	0.042	0.092	0.008	0.34	0.003

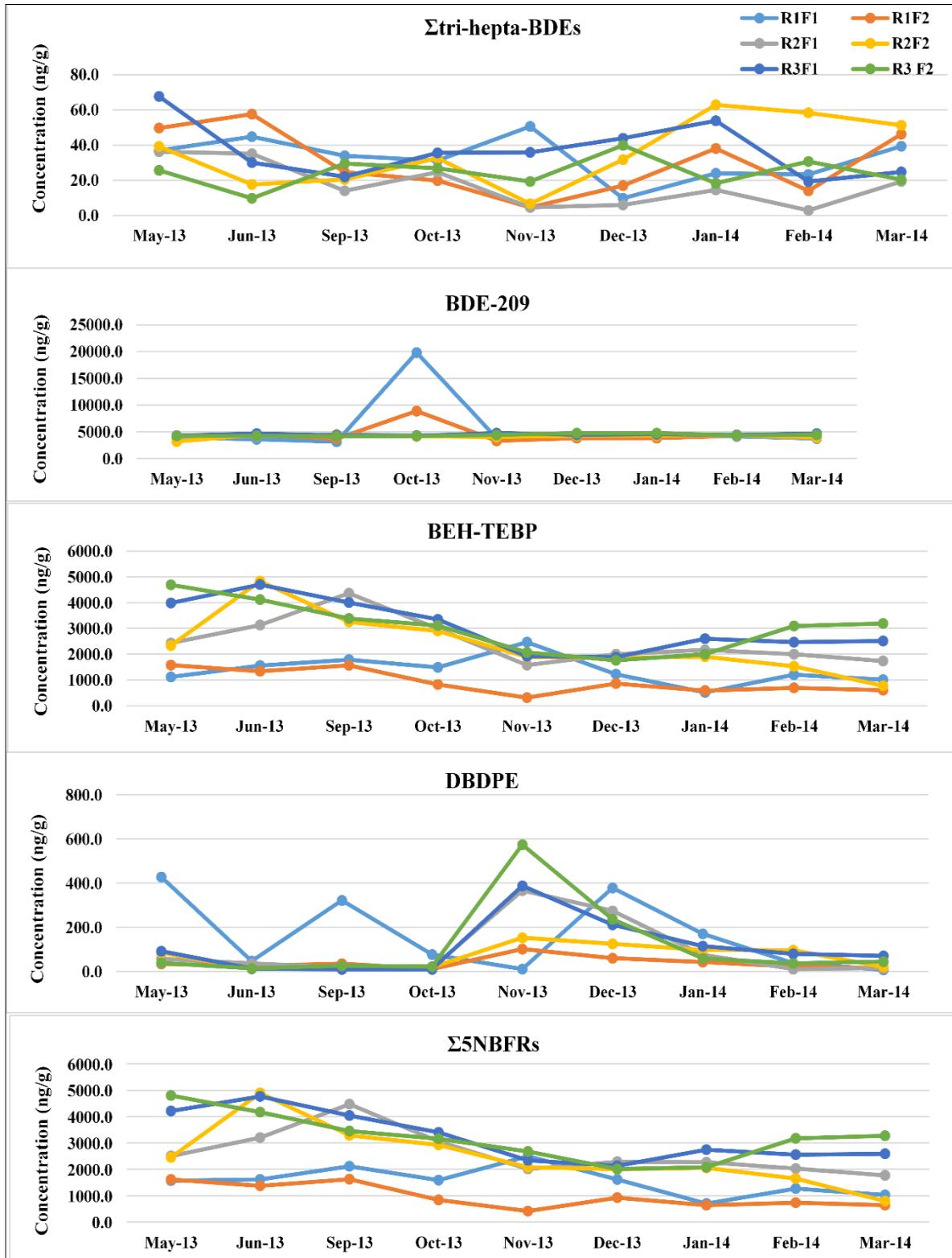
**Figure S7: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = bedroom and R3 = study) of Home 1**



**Figure S8: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = bedroom and R3 = study) of Home 2**



**Figure S9: Within-room temporal variation in concentrations (ng/g) of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in dust from two floor areas (F1 and F2) in different rooms (R1 = living room, R2 = adult bedroom and R3 = child's bedroom) of Home 3**



**Table S10: Maximum: minimum ratio in concentrations of  $\Sigma_7$ tri-hepta-BDEs, BDE-209, BEH-TEBP, DBDPE and  $\Sigma_5$ NBFRs in floor dust samples (F1 and F2) from three rooms (R1, R2 and R3) in Home1, Home2 and Home3 (H1, H2 and H3)**

Sampling area	$\Sigma_7$ tri-hepta-BDEs	BDE-209	BEH-TEBP	DBDPE	$\Sigma_5$ NBFRs
H1R1F1	4.0	2.3	2.1	6.4	1.9
H1R1F2	2.7	3.5	1.9	4.7	2.1
H1R2F1	29.5	7.0	1.6	8.1	2.5
H1R2F2	23.8	2.3	1.9	4.7	2.1
H1R3F1	21.1	2.4	2.5	9.7	3.1
H1R3F2	2.9	1.3	1.8	3.5	1.4
H2R1F1	3.4	2.2	1.8	8.0	2.9
H2R1F2	2.7	2.6	9.2	28.0	10.4
H2R2F1	15.1	1.7	2.2	6.2	2.5
H2R2F2	2.6	1.9	3.7	7.3	4.5
H2R3F1	2.4	1.9	1.7	4.7	2.1
H2R3F2	3.0	2.0	1.5	2.8	2.0
H3R1F1	5.2	6.3	4.8	71.4	3.6
H3R1F2	12.6	2.7	5.1	16.8	3.9
H3R2F1	12.0	1.2	2.8	60.9	2.5
H3R2F2	9.3	1.4	6.3	13.4	6.2
H3R3F1	3.5	1.1	2.5	43.2	2.2
H3R3F2	4.1	1.1	2.7	42.3	2.4