

# Emerging and legacy flame retardants in UK indoor air and dust

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**Emerging and legacy flame retardants in UK indoor air and dust: evidence for replacement of PBDEs by emerging flame retardants?**

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1 **Emerging and legacy flame retardants in UK indoor air and dust:**  
2 **evidence for replacement of PBDEs by emerging flame retardants?**

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## 16 Abstract

17 Concentrations of 27 emerging (EFRs) and legacy flame retardants (LFRs) were measured in  
18 samples of indoor air (n=35) and indoor dust (n=77) from UK homes and offices. All target  
19 compounds were detected in indoor air and dust samples. Relatively volatile EFRs (e.g.  
20 tetrabromoethylcyclohexane – DBE-DBCH) were more frequently detected in indoor air  
21 (detection frequencies > 60%), while less volatile EFRs (e.g. tetrabromobisphenolA-bis(2,3-  
22 dibromopropyl ether (TBBPA-BDBPE) and decabromodiphenyl ethane (DBDPE)) were  
23 predominant in dust. Concentrations of some EFRs (e.g. DBDPE) exceeded significantly those  
24 reported previously in UK dust ( $p < 0.05$ ), while concentrations of BDE-209 in office dust were  
25 significantly lower ( $p < 0.05$ ) than those reported previously in UK offices, consistent with the  
26 application of DBDPE as an alternative to the Deca-BDE formulation, of which BDE-209 is the  
27 principal constituent. Moreover, concentrations of BDEs 47 and 99 (both major constituents of  
28 the Penta-BDE product) in office air were significantly lower ( $p < 0.05$ ) than those in previous  
29 UK studies. Our results constitute important early evidence that restrictions on PBDEs have  
30 increased demand for EFRs in the UK, with the result that human exposure to PBDEs in UK  
31 homes and offices has decreased while exposure to EFRs has risen.

## 32 Introduction

33 Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) were among  
34 the most extensively used brominated flame retardants (FRs) up to 2004.<sup>1,2</sup> Since then, the Penta-  
35 BDE and Octa-BDE commercial mixtures have been listed under the Stockholm Convention on  
36 Persistent Organic Pollutants (POPs)<sup>3</sup> and production and use of both commercial mixtures  
37 phased out in Europe and North America.<sup>4</sup> Additionally, use of Deca-BDE in the EU has been  
38 banned in electrical and electronic applications since 1 July 2008<sup>5</sup>, and it is currently slated for  
39 listing under the Stockholm Convention. Moreover, HBCDD has also been listed as a POP under  
40 the same convention.<sup>6</sup> Following these bans and restrictions on PBDEs and HBCDD  
41 (collectively referred to as “legacy” flame retardants - LFRs), without concomitant relaxation of  
42 flame retardancy regulations, there is potential for these products to be replaced by so-called  
43 “emerging” flame retardants – EFRs – whose use is not restricted but that possess chemical  
44 structures similar to those of LFRs.<sup>1,7</sup> While this similarity is advantageous in the context of their  
45 ability to impart flame retardancy; there are concerns it extends to their toxicity and adverse  
46 environmental behavior – e.g. persistence and capacity for bioaccumulation.

47 While knowledge of environmental contamination with EFRs remains scant compared to our  
48 understanding of LFRs like PBDEs; recent reports show a broad suite of EFRs including DBDPE,  
49 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-  
50 TBB), bis(2-ethylhexyl)-tetrabromophthalate (BEH-TEBP), dechlorane plus (DDC-CO) and  
51 TBBPA-BDBPE to be present in indoor dust from various countries including the USA<sup>8</sup>, China<sup>9</sup>,  
52 Sweden<sup>10</sup>, Germany<sup>11</sup>, Belgium<sup>12</sup> and the UK<sup>13</sup>. In a previous UK study, BTBPE and DBDPE  
53 were detected in both office and home dust.<sup>13</sup> However, very little is known about concentrations  
54 of EFRs in indoor air. DBE-DBCH, pentabromotoluene (PBT), and hexabromobenzene (HBB)

55 were detected frequently in Swedish indoor air<sup>14</sup>, while Cequier et al.<sup>15</sup> also found DBE-DBCH,  
56 PBT, HBB, DBDPE and pentabromobenzene (PBBz) in most indoor air samples from  
57 Norwegian households and primary schools.

58 EFRs like DBE-DBCH, BTBPE, DBDPE and DDC-CO display potential for persistence and  
59 bioaccumulation that exacerbates concern over their adverse human health effects.<sup>16–22</sup> Moreover,  
60 animal studies suggest some EFRs pose health risks including: endocrine disruption (DBDPE,  
61 BTBPE, EH-TBB, BEH-TEBP and DBE-DBCH), neurodevelopmental and behavioural effects  
62 (EH-TBB, BEH-TEBP), hepatotoxicity (DBDPE), impaired reproductive physiology (DBDPE  
63 and DBE-DBCH), and gene expression (BTBPE); along with morphological abnormalities and  
64 mortality (DBE-DBCH), DNA damage (EH-TBB, BEH-TEBP and DBE-DBCH) and possibly  
65 cancer (DBE-DBCH).<sup>22–35</sup>

66 This study measured 16 EFRs, 8 PBDEs, and 3 HBCDD diastereomers in indoor air and settled  
67 floor dust from UK homes and offices. Concentrations of EFRs and LFRs were compared with  
68 those in previous UK studies to evaluate whether restrictions on PBDEs and HBCDD have  
69 reduced concentrations in indoor environments. Moreover, given that unlike other EU member  
70 states, the UK has – since 1988 - flame retardancy regulations that apply to furniture and  
71 furnishings, we compared our concentrations of target FRs with those reported from other  
72 countries. In addition, we estimated exposure to our target FRs via inhalation and dust ingestion  
73 by UK adults and toddlers.

## 74 **Materials and Methods**

### 75 **Sample collection**

76 Air samples were collected from February to May 2015 in offices (n=20) and houses (n=15) in

77 Birmingham, UK using double-bowl passive air samplers containing a polyurethane foam disk  
78 (PUF) and a glass fiber filter (GFF) (further information in the Supporting Information (SI)  
79 section). Each sampler was deployed for ~45 days at a height of 1-2 m.<sup>36</sup> Sampling rates (m<sup>3</sup>/day)  
80 for PBDEs, HBCDDs and EFRs were derived from previously reported studies,<sup>36-38</sup> with detailed  
81 information provided as SI. In summary, the air sampling rates used were: a uniform value of  
82 1.677 m<sup>3</sup>/day for EFRs, and 1.74, 1.472, 0.992, 1.014, 0.685, 0.691, 0.55, 0.567, 0.807, 0.775,  
83 and 0.761 m<sup>3</sup>/day for BDEs-47, 99, 100, 153, 154, 183, and 209, plus  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD  
84 respectively.

85 Dust was collected in offices (n=42) and houses (n=30) between June 2013 and May 2015.  
86 Samples were collected using a standard protocol under normal office and house use conditions  
87 to reflect actual human exposure.<sup>13</sup> (detailed information provided as SI). All dust samples were  
88 sieved through a pre-cleaned 500  $\mu$ m mesh sieve, homogenized thoroughly, transferred to clean  
89 glass vials and stored at -20 °C until analysis. Details of the sampling methods are provided as SI.

## 90 **Analytical protocols**

91 Samples were spiked with internal standards (<sup>13</sup>C-BDE 209, <sup>13</sup>C-BTBPE, <sup>13</sup>C-BEH-TEBP, BDE  
92 77, BDE 128) before extraction. Air samples (combined PUF and GFF) and aliquots of dust  
93 samples (typically between 50 and 200 mg) were extracted with hexane/acetone (3:1, v/v) using  
94 pressurized liquid extraction (Dionex, ASE 350). Crude extracts were purified by washing with  
95 3-4 mL concentrated sulfuric acid before reconstitution in 200  $\mu$ L (dust) and 50  $\mu$ L (air) iso-  
96 octane containing 250 pg/ $\mu$ L PCB-129 as recovery determination standard for quality  
97 assurance/quality control (QA/QC) purposes.

98 FRs were analyzed on a Trace 1310 gas chromatograph coupled to an ISQ™ single quadrupole

99 mass spectrometer (Thermo Scientific, TX, USA) operated in electron capture negative  
100 ionization (ECNI) mode. Two  $\mu\text{L}$  of cleaned extract were injected onto a Thermo Scientific  
101 TraceGOLD (TG)- Standard Quality Control (SQC) column ( $15\text{ m} \times 0.25\text{ mm} \times 0.25\text{ }\mu\text{m}$ ) using a  
102 programmable-temperature vaporizer injector in solvent vent mode.<sup>39</sup> After gas  
103 chromatography/mass spectrometry (MS) analysis, samples were evaporated and reconstituted in  
104 200  $\mu\text{L}$  of methanol containing  $\text{d}_{18}\text{-}\gamma\text{-HBCDD}$  (25  $\text{pg}/\mu\text{L}$ ) as recovery determination standard for  
105 determination of HBCDDs by liquid chromatography (LC)-MS/MS.<sup>40</sup> Quality assurance  
106 included method and field blanks. Five-point calibration curves were constructed for each target  
107 compound with excellent linearity ( $R^2 > 0.99$ ) over a concentration range relevant to those in  
108 samples. Average internal standard recoveries were:  $113 \pm 17\%$  (BDE-77),  $115 \pm 15\%$  (BDE-128),  
109  $98 \pm 21\%$  ( $^{13}\text{C}$ -BEH-TEBP),  $108 \pm 27\%$  ( $^{13}\text{C}$ -BTBPE), and  $95 \pm 28\%$  ( $^{13}\text{C}$ -BDE-209). Method  
110 accuracy and precision were evaluated via replicate analyses ( $n=6$ ) of NIST SRM 2585 (organics  
111 in indoor dust). Detailed description of analytical methods and QA/QC measurements is  
112 provided as SI.

### 113 **Statistical Analysis**

114 Statistical analysis was conducted using Excel (Microsoft Office 2010) and IBM SPSS Statistics  
115 21.0 (Chicago, IL, U.S.A.). Independent t-tests and ANOVA analyses were only conducted for  
116 target compounds with detection frequencies  $\geq 60\%$ . Where analyte peaks  $< \text{LOQ}$ , concentrations  
117 of target compounds were assumed to equal  $\text{LOQ}/2$ . Statistical analysis was performed on log-  
118 transformed concentrations, as concentrations in all data sets were revealed to be log-normally  
119 distributed using Kolmogorov-Smirnov test and visual inspection of quantile-by-quantile graphic  
120 plots. A  $p$ -value  $< 0.05$  was considered significant.



## 121 Results and discussion

### 122 Levels of FRs in indoor air

123 All target FRs were detected in indoor air from UK homes and offices (Table 1).

#### 124 EFRs

125 In both homes and offices, more volatile EFRs (DBE-DBCH, PBBz, PBT, HBB, tetrabromo-*o*-  
126 chlorotoluene (TBCT), 1,2,4,5-tetrabromo-3,6-dimethylbenzene (TBX),  
127 pentabromoethylbenzene (PBEB), and 2,3-dibromopropyl 2,4,6-tribromophenyl ether (TBP-  
128 DBPE)) showed higher detection frequencies in air (DF > 60%), while the less volatile DDC-CO  
129 and TBBPA-BDBPE were only detected in <20% of samples (Table 1). This is likely  
130 attributable to greater indoor use of these chemicals coupled with their relatively higher vapor  
131 pressures and lower  $K_{OA}$  values (Table S3). The most abundant compounds in air were  $\alpha$ - and  $\beta$ -  
132 DBE-DBCH, which when summed as  $\Sigma$ DBE-DBCH account for 63% and 80% of  $\Sigma$ EFRs in  
133 home and office air respectively. Median concentrations of  $\Sigma$ DBE-DBCH in home and office air  
134 were 110 and 290  $\text{pg}/\text{m}^3$  respectively, which exceeds slightly those reported in Norwegian  
135 households (77.9  $\text{pg}/\text{m}^3$ ), classrooms (46.6  $\text{pg}/\text{m}^3$ )<sup>15</sup> and in Swedish indoor microenvironments  
136 (55  $\text{pg}/\text{m}^3$ ).<sup>14</sup> We are aware of only one commercial DBE-DBCH formulation (Saytex BCL-462),  
137 which contains equal amounts of the two diastereomers,  $\alpha$ - and  $\beta$ -DBE-DBCH.<sup>41</sup> The ratio of  $\beta$ -  
138 to  $\alpha$ -DBE-DBCH ( $f_{\beta\text{-DBE-DBCH}}$ ) here ranged from 0.5 to 1.0, with a median value of 0.7, which is  
139 lower than in the commercial product. This is the first report of  $f_{\beta\text{-DBE-DBCH}}$  in indoor air, which  
140 precludes comparison to other studies. Possible explanations for the different isomer distribution  
141 in air compared to the commercial product include: differences in physicochemical properties  
142 (e.g. vapour pressure) or atmospheric degradation between the two diastereomers and possible

143 isomerisation post-emission. The latter explanation appears unlikely, as thermal conversion of  
144 the DBE-DBCH isomers occurs  $\geq 125$  °C.<sup>41</sup> However, such high temperatures could occur during  
145 incorporation of DBE-DBCH into polymers. Consequently,  $f_{\beta\text{-DBE-DBCH}}$  values in indoor air in this  
146 study may reflect the pattern present in DBE-DBCH-treated items rather than that in the  
147 commercial formulation.

148 Other frequently detected EFRs in air were: EH-TBB, BEH-TEBP and BTBPE. EH-TBB and  
149 BEH-TEBP were major constituents of Firemaster 550 and Firemaster BZ-54.<sup>42</sup> BEH-TEBP was  
150 also used in the flame retardant DP-45<sup>42</sup> and as a plasticizer in polyvinyl chloride and  
151 neoprene.<sup>43</sup> The fraction of EH-TBB  $f_{\text{EH-TBB}} = \text{EH-TBB}/(\text{EH-TBB} + \text{BEHTBP})$ . The  $f_{\text{EH-TBB}}$  value  
152 was  $0.77 \pm 0.03$  in Firemaster 550 and 0.70 in Firemaster BZ-54.<sup>43</sup> The values of  $f_{\text{EH-TBB}}$  in  
153 indoor air here are 0.03-0.99 (median=0.64) which is comparable to those reported in indoor air  
154 from Canada (0.3-0.99; median=0.83)<sup>44</sup>, the USA (0.16-0.95; median=0.63)<sup>44</sup> and the Czech  
155 Republic (0.27-0.89; median=0.72)<sup>44</sup> and outdoor air from Toronto, Canada (0.69)<sup>45</sup> and Chicago,  
156 the USA (0.70)<sup>46</sup> but higher than outdoor air from the Great Lakes area (0.26-0.54)<sup>42</sup> and  
157 Canada's Western Sub-Arctic (0.48)<sup>47</sup>. Such variation in  $f_{\text{EH-TBB}}$  ratios between studies may be  
158 attributable to different applications of BEH-TEBP across jurisdictions.

159 DFs of BTBPE, EH-TBB and BEH-TEBP exceed 70% in both microenvironment categories in  
160 this study (Table 1). This exceeded DFs ( $>33\%$ , with BTBPE not detected at all) for the same  
161 compounds in Norwegian and Swedish air).<sup>14,15</sup> This apparent greater detection of these EFRs in  
162 UK indoor air is at least partly attributable to the lower limits of quantitation (LOQs) in our  
163 study (BTBPE, EH-TBB and BEH-TEBP were 1.0, 0.1 and 0.1  $\text{pg}/\text{m}^3$ ) compared to the  
164 Scandinavian studies for which LOQs for BTBPE, EH-TBB and BEH-TEBP were 31, 330 and  
165 35  $\text{pg}/\text{m}^3$  in the Swedish study and 15.7, 7.4 and 2.9  $\text{pg}/\text{m}^3$  in the Norwegian study.<sup>14,15</sup> While no

166 production information is available for the UK or the EU, both EH-TBB and BEH-TEBP are  
167 reported replacements for Penta-BDE, and BTBPE a replacement for Octa-BDE.<sup>7</sup> It is therefore  
168 possible that following phase out of LFRs, these EFRs were more widely used in the UK than  
169 Norway and Sweden as a result of the UK's 1988 Furniture and Furnishings (Fire) (Safety)  
170 Regulations that differ substantially from those elsewhere in Europe.<sup>48</sup>

#### 171 *PBDEs*

172 BDEs-28, 47, 100, 99, and 209 were more frequently detected (DFs >85%) than other PBDEs  
173 (Table 1). Concentrations of  $\Sigma$ PBDEs (defined as the sum of PBDEs-28, 47, 100, 99, 154, 153,  
174 183 and 209) were 36-6400 pg/m<sup>3</sup> (median=280 pg/m<sup>3</sup>) in UK homes. Notwithstanding slight  
175 differences in the congeners constituting  $\Sigma$ PBDEs in different studies, the range of  $\Sigma$ PBDE  
176 concentrations reported here, is comparable to studies conducted in the USA (210-3980 pg/m<sup>3</sup>)  
177 and Sweden (72-1400 pg/m<sup>3</sup>), but higher than Germany (8.24-47 pg/m<sup>3</sup>), Australia (0.5-179  
178 pg/m<sup>3</sup>) and Japan (17-55 pg/m<sup>3</sup>)<sup>49-53</sup>. Concentrations of  $\Sigma$ PBDEs were 22-600 pg/m<sup>3</sup> (median=54  
179 pg/m<sup>3</sup>) in UK offices, similar to Australia (15-487 pg/m<sup>3</sup>) but much lower than USA (21-17200  
180 pg/m<sup>3</sup>) and Sweden (140-7300 pg/m<sup>3</sup>)<sup>49-54</sup>. BDE-209 predominated in homes and offices,  
181 contributing 73% and 50% of the total atmospheric PBDEs, respectively. This dominance of  
182 BDE-209 matches broadly that in Sweden<sup>50</sup>, Germany<sup>51</sup>, Australia<sup>52</sup>, and Japan<sup>53</sup>. In contrast,  
183 BDE-47 and BDE-99 predominated in USA homes<sup>49</sup> and offices<sup>54</sup>. This different pattern in the  
184 USA is likely due the far more extensive use of Penta-BDE in USA (94.7% of global Penta-BDE  
185 demand) than other countries<sup>55</sup>.

#### 186 *HBCDDs*

187 In homes, concentrations of  $\Sigma$ HBCDDs were 19-1500 pg/m<sup>3</sup> (median=110 pg/m<sup>3</sup>), comparable

188 to previously reported concentrations in UK houses (67-1300 pg/m<sup>3</sup>; median=180 pg/m<sup>3</sup>).<sup>56</sup> By  
189 comparison,  $\Sigma$ HBCDDs in our offices (median=41 pg/m<sup>3</sup>) were lower than those reported  
190 previously in UK offices (median=170 pg/m<sup>3</sup>).<sup>56</sup> Elsewhere, concentrations of  $\Sigma$ HBCDDs in  
191 offices, apartments, stores and two schools in Sweden (<1.3-19 pg/m<sup>3</sup>)<sup>14</sup> were exceeded  
192 substantially by those reported here. On average, the HBCDD isomer distribution was 87%  $\gamma$ -  
193 HBCDD, 8%  $\alpha$ -HBCDD, and 5%  $\beta$ -HBCDD in homes, and 71%  $\gamma$ -HBCDD, 18%  $\alpha$ -HBCDD,  
194 and 11%  $\beta$ -HBCD in offices. This pattern is similar to previous reports for UK and Swedish  
195 indoor air<sup>14,56</sup>.

#### 196 *Relative abundance of FRs in indoor air*

197 PBDEs predominated in homes, contributing 44%  $\Sigma$ FRs, while EFRs predominated in offices,  
198 accounting for 83%  $\Sigma$ FRs.  $\Sigma$ HBCDDs contributed 18% and 8%  $\Sigma$ FRs in homes and offices,  
199 respectively (Figure S2, SI). The comparatively high abundance of EFRs in both offices (38%)  
200 and homes, suggests their widespread use in the UK. The far greater abundance of EFRs in  
201 offices could be attributable to different and greater numbers of sources in offices compared to  
202 homes, and also reflect a more rapid turnover of such source items in offices – i.e. putative FR  
203 sources such as foam filled office furniture and electronic and electrical equipment are more  
204 frequently replaced in offices. Such rapid “turnover” of old source items containing LFRs with  
205 new replacements containing EFRs, may explain the predominance of the latter in offices.

#### 206 **Levels of FRs in indoor dust**

207 Table 2 summarises the concentrations of target FRs in UK homes and office dust.

208 *EFRs*

209 In contrast to air, less volatile compounds were more frequently detected in office and home dust.  
210 These include: DDC-CO, TBBPA-BDBPE and DBDPE (DF>60%) (Table 2). Of note is the very  
211 high concentration of 4,700,000 ng/g BTBPE detected in one office dust sample, which to the  
212 authors' knowledge is the highest BTBPE level reported in indoor dust to date. We are unable to  
213 explain this high concentration based on a survey of putative FR-treated items in the sampled  
214 office. TBBPA-BDBPE was the most abundant compound, comprising 87% and 71%  $\Sigma$ EFRs in  
215 homes and offices based on median concentrations, followed by BEH-TEBP, BTBPE, and  
216 DBDPE. TBBPA-BDBPE was also the predominant EFR detected previously in UK classroom  
217 dust, accounting for an average of 48%  $\Sigma$ EFRs,<sup>12</sup> indicating its extensive UK use. Concentrations  
218 of TBBPA-BDBPE in this study exceeded those in Belgian homes and offices<sup>12</sup> and USA homes<sup>8</sup>.  
219 In office dust, concentrations of BTBPE, BEH-TEBP, and DBDPE in this study were comparable  
220 to those in offices in Beijing, China<sup>57</sup> but exceeded those in Belgian offices<sup>12</sup>. For house dust, our  
221 concentrations were comparable to those in Belgium, New Zealand, Germany, Sweden, and  
222 Norway but lower than those found in USA and Canada.<sup>8,10-12,15,58-60</sup> In studies not targeting  
223 TBBPA-BDBPE; the most abundant EFRs in house dust from Norway, Germany, Sweden, USA  
224 and China were DBDPE and BEH-TEBP<sup>8-11,15</sup>. We observed a similar abundance of DBDPE and  
225 BEH-TEBP. Our data also represent the first report of DDC-CO in UK indoor dust at median  
226 concentrations of 5.7 and 73 ng/g in homes and offices, respectively. The contribution of *anti*-  
227 DDC-CO to  $\Sigma$ DDC-CO (expressed as  $f_{\text{anti}}$ ) in the technical mixture is 0.65–0.75.<sup>15</sup> By  
228 comparison,  $f_{\text{anti}}$  values in our indoor dust samples are 0.27-0.99. This wider range is similar to  
229 previous studies of indoor dust from Canada, China and Sweden.<sup>14,60,61</sup> Similar to DBE-DBCH in  
230 air, plausible explanations include: isomer-specific degradation, variable environmental

231 behaviour arising from different physicochemical properties, as well as possible thermal  
232 isomerization during incorporation of DDC-CO into consumer products.

233 DBE-DBCH was detected in all indoor dust samples underlining its wide UK application.  $f_{\beta\text{-DBE-DBCH}}$   
234  $f_{\beta\text{-DBE-DBCH}}$  in indoor dust was 0.32-2.88 (median=0.85). This median value is lower than in the  
235 commercial product. Interestingly,  $f_{\beta\text{-DBE-DBCH}}$  values in indoor dust significantly exceed those in  
236 indoor air ( $p < 0.01$ ). Few studies have addressed isomer-specific aspects of DBE-DBECH. Wong  
237 et al.<sup>63</sup> reported the fate of DBE-DBCH in urban soil, with  $\beta\text{-DBE-DBCH}$  showing a slightly  
238 higher soil-air partition coefficient ( $K_{SA}$ ) than  $\alpha\text{-DBE-DBCH}$ , indicating  $\beta\text{-DBE-DBCH}$  to be  
239 less volatile than  $\alpha\text{-DBE-DBCH}$ . This may provide a plausible explanation for the observed low  
240  $f_{\beta\text{-DBE-DBCH}}$  in indoor air and dust compared to the commercial formulation. Combined with the  
241 reported slightly faster degradation rate of  $\alpha\text{-}$  compared to  $\beta\text{-DBE-DBCH}$ <sup>63</sup>, this may contribute  
242 to the significantly lower  $f_{\beta\text{-DBE-DBCH}}$  observed in indoor dust compared to air. The fate of DBE-  
243 DBCH isomers in the environment, the reasons for different isomeric profiles in various matrices  
244 and possible implications of such variations on exposed organisms are a research priority.

#### 245 *PBDEs*

246 Each target PBDE was detected in >90% of indoor dust samples except for BDE-28 and BDE-47  
247 (Table 2). In house dust, concentrations of  $\sum\text{PBDEs}$  were 180-370000 ng/g (median=4600 ng/g).  
248 This exceeds those reported in other countries like: Germany (36.6-1580 ng/g), Denmark (65-  
249 61524), Sweden (53-4000 ng/g), USA (920-17000 ng/g), Belgium (4-6509 ng/g), and China  
250 (132-3887 ng/g) but is comparable to Canada (170-170000 ng/g).<sup>51,52,64-68</sup> In our office dust  
251 samples, concentrations of  $\sum\text{PBDEs}$  were 270-110000 ng/g (median=3000 ng/g). This is  
252 comparable to the USA, Sweden and China but higher than Belgium.<sup>51,55,67,69</sup> Similar to previous

253 UK studies, BDE-209 predominated in dust, contributing 99% and 94%  $\Sigma$ PBDEs in homes and  
254 offices, respectively. This predominance of BDE-209 concurs with other countries in Europe and  
255 Asia, but is more marked than in North America, where greater use of Penta-BDE leads to a  
256 greater contribution from BDEs-47 and 99.<sup>51,52,55,64-67,69</sup> The substantially higher concentrations  
257 of BDE-209 in UK indoor dust compared to other European countries, may be attributed to the  
258 extensive UK use of Deca-BDE in upholstery fabrics and textiles (1,000-1,200 tonnes/year out of  
259 an estimated 1,500 tonnes/year in the EU for this application) to comply with the UK Furniture  
260 and Furnishing Fire Safety Regulations 1988.<sup>70</sup>

#### 261 *HBCDDs*

262 HBCDDs were detected in all samples.  $\Sigma$ HBCDDs concentrations in homes were 50-110000  
263 ng/g (median=610 ng/g) in homes, which compares closely with those previously detected in UK  
264 house dust (140-140000 ng/g)<sup>57</sup>. In office dust, concentrations of  $\Sigma$ HBCDDs were 150-6400  
265 ng/g (median=1700 ng/g). This is also in line with previous UK data (90-6600 ng/g).<sup>57</sup> Our  
266  $\Sigma$ HBCDDs concentrations in office and house dust exceeded those in dust from offices,  
267 apartments, stores and two schools in Sweden (17-2900 ng/g).<sup>14</sup> In our study,  $\alpha$ -HBCDD was the  
268 dominant contributor to  $\Sigma$ HBCDD in home and office dust. The average isomer profile was 53%  
269  $\alpha$ -HBCDD, 29%  $\gamma$ -HBCDD, and 18%  $\beta$ -HBCDD in homes, while for offices; it was 56%  $\alpha$ -  
270 HBCDD, 27%  $\gamma$ -HBCDD, and 17%  $\beta$ -HBCDD. This pattern differs from that in indoor air. To  
271 investigate whether the difference is statistically significant, fractional contributions of  $\alpha$ -  
272 HBCDD and  $\gamma$ -HBCDD to  $\Sigma$ HBCDD detected in matched pairs (n=32) of indoor air and dust  
273 samples, i.e. collected from the same room at the same time were compared. Similar to a  
274 previous UK study, we found greater relative abundance of  $\alpha$ -HBCDD in dust relative to air,

275 with the opposite observed for  $\gamma$ -HBCDD ( $p < 0.001$ ). As previously, we attribute this to a post-  
276 depositional shift from  $\gamma$ -HBCDD to  $\alpha$ -HBCDD, and/or preferential degradation of  $\gamma$ -HBCDD in  
277 indoor dust.<sup>57</sup>

#### 278 *Relative abundance of FRs in indoor dust*

279 Similar to indoor air, PBDEs predominated in house dust, contributing 66 %  $\Sigma$ FRs, while EFRs  
280 dominated in office dust, accounting for 51%  $\Sigma$ FRs.  $\Sigma$ HBCDDs contributed 9% and 17%  $\Sigma$ FRs  
281 in home and office dust, respectively (Figure S3).

#### 282 **Temporal trends in UK indoor contamination**

##### 283 *EFRs*

284 To our knowledge, these are the first data about concentrations of EFRs in UK indoor air.  
285 Consequently, comparison with previous studies is not possible. With respect to indoor dust  
286 however, concentrations of BTBPE and DBDPE in dust taken from homes ( $n=30$ ) and offices  
287 ( $n=18$ ) in the West Midlands collected between July 2006 and June 2007 exist<sup>13</sup>. Following log-  
288 transformation of concentrations, we conducted a t-test to compare concentrations in these two  
289 studies. For house dust, concentrations of BTBPE in our study were statistically  
290 indistinguishable to the previous study ( $p > 0.05$ ); in contrast, concentrations of DBDPE exceeded  
291 significantly ( $p < 0.05$ ) those in the earlier study. Moreover, t-test comparison revealed  
292 concentrations of BTBPE and DBDPE in our office dust samples were both significantly higher  
293 than those reported previously ( $p < 0.05$ ).<sup>13</sup> While based on a relatively limited number of samples,  
294 these findings suggest restrictions on LFRs have led to a concomitant increase in concentrations  
295 in UK indoor dust of DBDPE and – to a lesser extent – BTBPE.



296 *PBDEs*

297 Two previous studies have reported concentrations of PBDEs in UK indoor air. The first reported  
298 concentrations of BDE-47, 99, 100, 153, and 154 in air from offices and homes using high-  
299 volume active air samplers from 2001-2002<sup>71</sup>; while the second - conducted between September  
300 2003 and November 2005 - measured BDE-28, 47, 49, 66, 85, 99, 100, 153, and 154 using  
301 double-bowl passive air samplers containing a PUF disk<sup>72</sup>. For home air, no significant  
302 difference was observed between our study and the two previous studies<sup>71,72</sup>. By comparison, for  
303 office air, concentrations of BDE 47 and 99 were significantly lower than in previous studies  
304 (ANOVA test of log transformed concentrations,  $p < 0.05$ )<sup>71,72</sup>. We are aware of the potential  
305 difficulties of comparing concentrations derived using different air sampler configurations. Two  
306 studies that examined such difficulties are: (a) Melymuk et al. (2012), who found no major  
307 differences between PBDE concentrations derived using passive air samplers and high volume  
308 active samples; and (b) Abdallah and Harrad (2010), who found no significant difference  
309 between indoor air concentrations of BFRs including PBDEs measured using both passive and  
310 low volume active samplers. On the basis of these previous studies, we conclude the influence of  
311 the different sampling methods used in the three studies compared here, should not overly  
312 obscure the temporal trend.

313 Concentrations of BDE-209 in UK office dust were significantly lower ( $p < 0.05$ ) than those  
314 reported in a previous study of UK offices<sup>13</sup>; while for UK house dust, BDE-209 concentrations  
315 were comparable ( $p > 0.05$ ) to those reported previously<sup>13</sup>, perhaps indicating more rapid removal  
316 of “legacy” PBDEs in offices than homes. No significant temporal differences were found for  
317 other PBDEs in both home and office dust. We attribute the lack of a decline in BDEs- 47 and 99  
318 in dust (in contrast to air) to the different sample populations for air and dust.

319 *HBCDDs*

320 We compared our data for HBCDDs in indoor air with those in the previous UK study<sup>57</sup> by  
321 independent t-test. In domestic air, no significant differences were detected; in contrast,  
322 concentrations of  $\gamma$ -HBCDD were significantly lower in office air in our study ( $p < 0.001$ )  
323 ( $\alpha$ -HBCDD and  $\beta$ -HBCDD were not compared as DFs were  $< 60\%$ ). This may possibly indicate  
324 replacement of HBCDDs by EFRs such as DBE-DBCH in expanded polystyrene for building  
325 insulation<sup>14</sup>. For indoor dust, our concentrations of  $\gamma$ -HBCDD in house dust were significantly  
326 lower ( $p < 0.001$ ) than in the previous study<sup>57</sup> but not for other diastereomers and  $\Sigma$ HBCDDs. No  
327 significant differences were observed for office dust.

328

### 329 **Comparisons between indoor microenvironments**

330 Previous studies of both air and dust revealed differences in concentrations of FRs between  
331 offices and homes<sup>13,67,71,72</sup>. Such differences are likely due to the different types and abundance  
332 of FR sources in these two microenvironment categories. We therefore examined our data for  
333 such differences.

334 *Indoor air*

335 We compared concentrations in home and office air using an independent t-test. For EFRs, DBE-  
336 DBCH, PBBz and EH-TBB were significantly higher in offices than homes ( $p < 0.001$  for DBE-  
337 DBCH,  $p < 0.05$  for PBBz and EH-TBB), with no significant differences between homes and  
338 offices detected for other EFRs. For PBDEs, concentrations of BDE-209 were significantly  
339 higher in homes than offices ( $p < 0.005$ ); but no differences were found for other congeners. In

340 contrast, previous studies in West Midlands indoor air focusing on tri- to hexa-BDEs found  
341 concentrations in offices to exceed those in homes ( $p < 0.05$ ).<sup>71,72</sup> Finally, for HBCDDs,  
342 concentrations of  $\gamma$ -HBCDD were significantly higher ( $p < 0.005$ ) in homes than offices.  
343 ( $\alpha$ -HBCDD and  $\beta$ -HBCDD were not compared as their DFs were  $< 60\%$ ).

#### 344 *Indoor dust*

345 For indoor dust, statistical analysis revealed concentrations of DBE-DBCH, EH-TBB, BEH-  
346 TEBP, BTBPE, DDC-CO and DBDPE in offices exceeded significantly those in homes. No  
347 significant differences between offices and homes were found for other EFRs.

348 In contrast to our observations for air, BDE-209 concentrations in homes and offices were  
349 comparable ( $p > 0.05$ ), while for BDE-47 and BDE-99, concentrations in offices exceeded  
350 significantly ( $p < 0.001$ ) those in homes. This result concurs with previous studies of PBDEs in  
351 Belgian and UK home and office dust<sup>13,67</sup>. We attribute the differences between air and dust (e.g.  
352 BDE-209 higher in homes than offices for air but dust) to the larger number of dust samples  
353 collected.

354 For HBCDDs, no significant differences were detected between concentrations of  $\Sigma$ HBCDDs in  
355 dust from homes and offices ( $p > 0.05$ ); consistent with a previous study<sup>57</sup>.

#### 356 **Correlations between FRs in the studied microenvironments**

357 To investigate potential common sources of our FRs. Pearson correlation analysis was applied to  
358 assess relationships between the target compounds (Table S5 and 6).  $\alpha$ -DBE-DBCH  
359 concentrations correlated significantly with those of  $\beta$ -DBE-DBCH in both air and dust samples  
360 ( $p < 0.001$ ) with similar findings observed for HBCDD diastereomers. Significant correlations

361 between concentrations of tetra- to hexa-BDEs were observed in indoor air and dust ( $p < 0.05$ ),  
362 with BDE-183 correlated with BDE-47 ( $p < 0.005$ ), BDE-100 ( $p < 0.05$ ), BDE-153 ( $p < 0.01$ ) and  
363 BDE-154 ( $p < 0.001$ ) in indoor dust. This probably reflects the presence of these congeners in  
364 Penta-BDE and Octa-BDE.

365 For EFRs, PBBz was correlated significantly with PBEB, TBP-DPTE and HBB in indoor air and  
366 dust. HBB also correlated with PBEB and TBP-DPTE in indoor air ( $p < 0.001$ ), with HBB and  
367 PBBz correlated with BDE-28, 47 and 99 in air and dust ( $p < 0.05$ ). While some of these observed  
368 correlations are hard to rationalise and may have occurred by chance; those between PBEB,  
369 HBB and PBBz may originate from a common source resulting from thermal degradation of  
370 polymeric FRs. Conditions that may favour such formation may be encountered during the  
371 process of incorporating these FRs into goods – i.e. the elevated temperatures experienced by  
372 FRs when incorporated into molten polymers.<sup>74–76</sup>

### 373 **Correlations between FR concentrations in air and dust**

374 If semi-volatile organic compounds (SVOCs) partitioning between the gaseous and particulate  
375 phases have reached equilibrium under ideal conditions, their concentrations in air and dust  
376 should correlate.<sup>77</sup> To investigate this hypothesis, Pearson correlation analysis was conducted to  
377 investigate the relationship between FR concentrations in matched air and dust samples ( $n=30$ ,  
378 Table S7). Significant correlations between concentrations of EFRs in air and dust were observed  
379 ( $p < 0.005$ ), especially for the more volatile EFRs, e.g. DBE-DBCH and PBT. Moreover,  
380 concentrations of BDE-28 and 47 in air and dust were also significantly correlated ( $p < 0.05$ ). The  
381 partition coefficient between dust and air is expressed as  $K_{DA}$  ( $K_{DA} = C_{dust}/C_{air}$ , where  $C_{dust}$  and  
382  $C_{air}$  are the concentrations of FR in dust and air, respectively). Median values of Log  $K_{DA}$  in

383 homes and offices were linearly associated with  $K_{OA}$  values of the studied FRs (Figure S4),  
384 suggesting thermodynamic equilibrium to exist between air and dust for most target FRs. Similar  
385 relationships were reported by Cequier et al.<sup>41</sup> for PFRs and PBDEs in Norwegian homes and  
386 classrooms who concluded knowledge of concentrations of FRs in either air or dust permits  
387 prediction of their levels in the other phase given the  $K_{OA}$  of the compound in question.<sup>41</sup>

388 No correlation between  $\text{Log } K_{DA}$  and  $K_{OA}$  of HBCDD diastereomers was found. This is  
389 consistent with the previously observed post-depositional transformation in dust of  
390 HBCDDs.<sup>57,78</sup>

### 391 **Exposure to FRs via inhalation and dust ingestion**

392 Human exposure to FRs via inhalation and dust ingestion was estimated based on concentrations  
393 reported here. The algorithms and assumptions applied to estimate exposure via different routes  
394 are provided in SI. These data are important – especially for the EFRs – as they facilitate: (a)  
395 evaluation of the relative importance of inhalation as opposed to dust ingestion (and dietary  
396 exposure once such data are available) for different chemicals, and (b) future risk assessment by  
397 comparison of exposure with health-based limit values once set. Median estimated exposure for  
398 toddlers via air inhalation to  $\sum\text{EFRs}$ ,  $\sum\text{PBDEs}$  and  $\sum\text{HBCDDs}$  was 0.66, 0.43 and 0.14 ng/day,  
399 respectively. For adults, estimated exposure via inhalation to  $\sum\text{EFRs}$ ,  $\sum\text{PBDEs}$  and  $\sum\text{HBCDDs}$   
400 was 4.3, 2.0 and 2.1 ng/day, respectively. Our estimated inhalation exposures to  $\sum\text{tri-hexa BDEs}$   
401 (Table 3) were comparable to those reported previously for UK toddlers and adults (0.16 and  
402 0.82 ng/day respectively)<sup>72</sup>. There are no previous estimates of human exposure to BDE-209 via  
403 inhalation in the UK. For  $\sum\text{HBCDDs}$ , Abdallah et al.<sup>57</sup> reported median daily intakes of 0.8 and  
404 3.9 ng/day for UK toddlers and adults, slightly exceeding our estimates (Table 3).

405 The main contributors to adult inhalation exposures were:  $\alpha$ -DBE-DBCH,  $\beta$ -DBE-DBCH, BDE-  
406 209 and  $\gamma$ -HBCDD, contributing 21%, 14%, 17% and 24% to the median  $\Sigma$ FRs exposure,  
407 respectively. Similar findings were observed for toddlers (Figure 1 and Figure S5 and 6).

408 Estimates of “typical” exposure for toddlers (i.e. dust contaminated at the median concentration  
409 ingested at the mean dust ingestion rate) to  $\Sigma$ EFRs,  $\Sigma$ PBDEs and  $\Sigma$ HBCDDs were 38, 280 and  
410 25 ng/day, respectively. For adults, “typical” exposures to  $\Sigma$ EFRs,  $\Sigma$ PBDEs and  $\Sigma$ HBCDDs  
411 were 27, 100 and 15 ng/day, respectively. Harrad et al.<sup>13</sup> reported “typical” exposure of toddlers  
412 and adults to  $\Sigma$ PBDEs via dust ingestion to be 234.3 and 612.6 ng/day respectively, which in  
413 both instances exceeds our exposure estimates (Table 3). A previous study reported UK exposure  
414 to  $\Sigma$ HBCDDs via dust ingestion to be 86.9 and 32.5 ng/day for toddlers and adults respectively,  
415 which also exceeds our current estimates (Table 3)<sup>57</sup>. For EFRs, our “typical” exposure estimates  
416 for UK adults and toddlers via dust ingestion exceed substantially those reported in a previous  
417 study<sup>12</sup> (Table 3). While this suggests UK exposure to LFRs via dust ingestion is decreasing, a  
418 substantial increase in exposure to EFRs is a potential cause for concern.

419 Under our “typical” exposure scenario, dust ingestion is a more significant contributor to human  
420 exposure to  $\Sigma$ EFRs,  $\Sigma$ PBDEs and  $\Sigma$ HBCDDs than air inhalation (Table 3). This is similar to  
421 the findings of previous studies on PBDEs and HBCDDs<sup>57,72</sup>. For toddlers’ intake of FRs, dust  
422 ingestion was more important than air inhalation for all target compounds. However, for FRs  
423 with relatively high vapor pressures (e.g. DBE-DBCH and BDE-28), inhalation contributes more  
424 to UK adult exposure than dust ingestion (Table 3).

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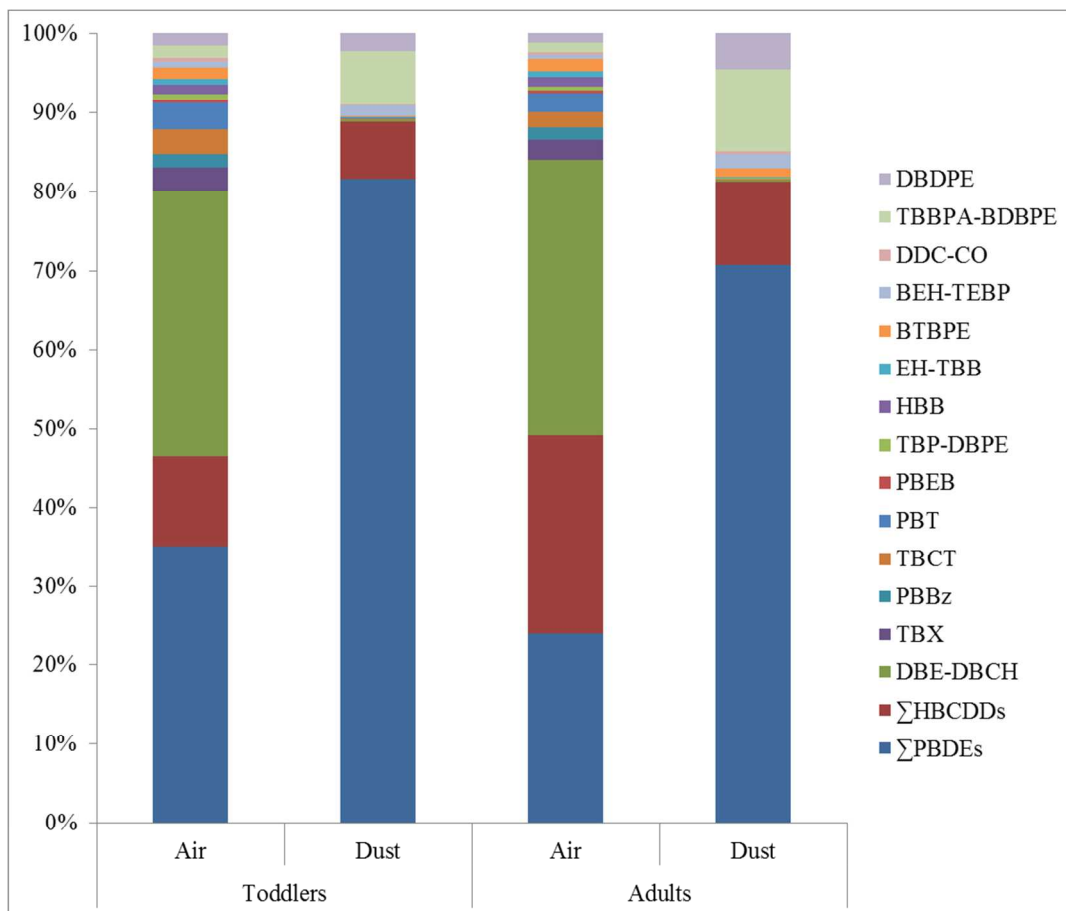
429 **Supporting Information**

430 Full details of the analytical protocols, QA/QC measurements and human exposure assessment  
431 models are provided as supporting information. This material is available free of charge via the  
432 Internet at <http://pubs.acs.org>.

433

434 **Figures**

435 **Figure 1.** Relative contribution (expressed as %) of different target FRs to the overall daily  
 436 exposure (ng/day) of UK toddlers and adults via inhalation and dust ingestion.



437



438 **Tables**439 **Table 1.** Descriptive Statistics for Concentrations (pg/m<sup>3</sup>) of target FRs in UK indoor air

Statistical Parameter	Homes					Offices				
	This study, n=15				UK previous studies	This study, n=20				UK previous studies
	DF	Mean	Median	Range	Range	DF	Mean	Median	Range	Range
$\alpha$ -DBE-DBCH	100%	99	64	17-350	--	100%	180	160	74-440	--
$\beta$ -DBE-DBCH	100%	74	45	13-250	--	100%	140	120	41-300	--
TBX	100%	31	9.7	1.6-190	--	100%	16	14	2.6-38	--
PBBz	93%	6.6	5.3	<0.10-22	--	100%	16	11	3.1-47	--
TBCT	60%	22	9.9	<0.10-240	--	65%	1.6	<0.10	<0.10-11	--
PBT	100%	17	11	2.3-63	--	65%	15	4.7	<0.10-200	--
PBEb	100%	1.6	1.3	0.41-5.4	--	100%	4.2	1.7	0.31-35	--
TBP-DBPE	93%	3.5	2	<0.40-14	--	85%	18	1.9	<0.40-280	--
HBB	73%	11	4.2	<0.20-91	--	85%	19	9.4	<0.20-170	--
EH-TBB	100%	4.8	2	0.05-44	--	100%	22	5.3	0.67-240	--
BTBPE	73%	11	5	<1.0-50	--	100%	32	11	0.7-220	--
BEH-TEBP	93%	10	2.1	<0.10-130	--	90%	2.2	1.4	<0.10-11	--
<i>syn</i> -DDC-CO	7%	<2.0	<2.0	<2.0-4.6	--	5%	1.3	<2.0	<2.0-7.7	--
<i>anti</i> -DDC-CO	20%	2.2	<1.2	<1.2-20	--	5%	1.8	<1.2	<1.2-24	--
TBBPA-BDBPE	20%	13	<10	<10-87	--	5%	7.3	<10	<10-50	--
DBDPE	40%	26	<10	<10-97	--	5%	7.5	<10	<10-54	--
BDE28	100%	22	1.2	0.21-310	<dl-60.8 <sup>b</sup>	100%	4.3	2.7	0.81-13	<dl-43.5 <sup>b</sup>
BDE47	100%	120	13	0.15-1700	45-1330 <sup>a</sup> ; 1.9-106.9 <sup>b</sup>	100%	44	6	0.15-380	58-7140 <sup>a</sup> ; 4.0-567.6 <sup>b</sup>
BDE100	87%	44	1.5	<0.10-600	2.6-82 <sup>a</sup> ; <dl-26.5 <sup>b</sup>	85%	3	1.5	<0.10-17	4.1-1450 <sup>a</sup> ; <dl-142.4 <sup>b</sup>
BDE99	100%	130	12	0.05-1700	8.7-209 <sup>a</sup> ; <dl-80.4 <sup>b</sup>	100%	9.2	7.9	1.2-42	9.0-6510 <sup>a</sup> ; 3.9-633.5 <sup>b</sup>
BDE154	53%	14	1.2	<0.50-180	<1.0-6.0 <sup>a</sup> ; <dl-16.5 <sup>b</sup>	40%	0.78	<0.50	<0.50-6	<1.0-228 <sup>a</sup> ; <dl-12.8 <sup>b</sup>
BDE153	67%	24	1.8	<0.4-180	<1.0-6.1 <sup>a</sup> ; <dl-20.5 <sup>b</sup>	55%	0.6	<0.4	<0.4-3.6	<1.0-181 <sup>a</sup> ; <dl-7.0 <sup>b</sup>
BDE183	27%	2.8	<1.0	<1.0-12	--	10%	<1.0	<1.0	<1.0-3.8	--
BDE209	100%	660	170	23-3800	--	100%	74	26	2.3-350	--
$\alpha$ -HBCDD	40%	43	<2.6	<2.6-400	14-430 <sup>c</sup>	59%	8.8	5.4	<2.6-31	18-87 <sup>c</sup>
$\beta$ -HBCDD	47%	17	<2.2	<2.2-160	5-54 <sup>c</sup>	53%	5.2	<2.2	<2.2-15	14-34 <sup>c</sup>
$\gamma$ -HBCDD	100%	270	110	16-1400	39-710 <sup>c</sup>	100%	55	34	3.1-320	43-370 <sup>c</sup>
$\Sigma$ HBCDDs	--	320	110	19-1500	67-1300 <sup>c</sup>	--	69	41	5.5-360	70-460 <sup>c</sup>

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446

447 **Table 2.** Descriptive Statistics for Concentrations (ng/g) of target FRs in UK indoor dust

Statistical Parameter	Homes					Offices				
	This study, n=45				UK previous studies	This study, n=47				UK previous studies
	DF	Mean	Median	Range	Range	DF	Mean	Median	Range	Range
$\alpha$ -DBE-DBCH	100%	9.4	5.9	1.4-52	--	100%	23	15	2.5-130	--
$\beta$ -DBE-DBCH	100%	12	6.2	1.5-77	--	100%	18	13	2.7-120	--
TBX	80%	18	1.4	<0.010-410	--	98%	3.2	2.1	<0.010-19	--
PBBz	87%	3.3	2.2	<0.010-12	--	91%	4.5	3.3	<0.010-23	--
TBCT	40%	21	<0.010	<0.010-300	--	67%	11	3.4	<0.010-68	--
PBT	67%	7.1	1.8	<0.010-90	--	87%	6	2.5	<0.010-59	--
PBEb	87%	2.3	0.78	<0.010-21	--	100%	2	1.4	0.36-10	--
TBP-DBPE	84%	6.6	1.8	<0.050-47	--	98%	24	4.7	<0.050-370	--
HBB	75%	1.8	<0.030	<0.030-12	--	91%	14	9.9	<0.030-84	--
EH-TBB	94%	21	10	<0.010-85	--	91%	120	31	<0.010-2000	--
BTBPE	100%	14	5.6	0.01-110	<dl-1900 <sup>a</sup>	100%	100000	160	0.019-4700000	<dl-40 <sup>a</sup>
BEH-TEBP	100%	240	65	16-3500	--	100%	1000	160	54-25000	--
<i>syn</i> -DDC-CO	63%	3.6	0.77	<0.26-28	--	98%	60	11	<0.26-640	--
<i>anti</i> -DDC-CO	84%	21	4.9	<0.15-170	--	98%	210	62	<0.15-2100	--
TBBPA-BDBPE	100%	5800	1000	71-49000	--	100%	3400	2300	310-14000	--
DBDPE	60%	240	41	<1.2-2300	<dl-3400 <sup>a</sup>	96%	1600	440	<1.2-17000	<dl-860 <sup>a</sup>
BDE28	57%	1.9	0.16	<0.03-15	<dl-2.1 <sup>a</sup>	82%	3.9	2.6	<0.03-22	<dl-11 <sup>a</sup>
BDE47	77%	14	13	<0.04-50	1.2-58 <sup>a</sup>	100%	83	37	7.1-660	2.6-380 <sup>a</sup>
BDE100	100%	4.2	3	0.75-16	<dl-17 <sup>a</sup>	100%	18	12	1.9-120	<dl-79
BDE99	100%	31	22	5-92	2.8-180 <sup>a</sup>	100%	100	77	15-480	4.2-490 <sup>a</sup>
BDE154	94%	2	1.2	<0.06-9.3	<dl-16 <sup>a</sup>	100%	7.7	3.9	0.8-68	<dl-38 <sup>a</sup>
BDE153	100%	4.8	3	0.025-24	<dl-110 <sup>a</sup>	100%	28	9.2	0.025-190	<dl-99 <sup>a</sup>
BDE183	90%	7.4	3.5	<0.13-51	<dl-550 <sup>a</sup>	100%	29	9.8	0.065-220	<dl-24 <sup>a</sup>
BDE209	100%	34000	4500	160-370000	<dl-2200000 <sup>a</sup>	100%	9100	2700	200-110000	620-280000 <sup>a</sup>
$\alpha$ -HBCDD	100%	2300	320	21-28000	22-66000 <sup>b</sup>	100%	1100	980	100-2800	15-2900 <sup>b</sup>
$\beta$ -HBCDD	100%	1100	85	6.1-12000	9-26000 <sup>b</sup>	100%	300	330	22-590	11-1300 <sup>b</sup>
$\gamma$ -HBCDD	100%	4900	93	23-71000	70-75000 <sup>b</sup>	100%	630	350	31-3700	36-3700 <sup>b</sup>
$\Sigma$ HBCDDs	--	8300	610	50-110000	140-140000 <sup>b</sup>	--	2000	1700	150-6400	90-6600 <sup>b</sup>

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452 **Table 3.** Estimates of exposure (ng/day) of UK Adults and Toddlers to FRs via air and dust, and  
 453 relative significance (%) of each pathway under a typical exposure scenario <sup>a</sup>  
 454

Compounds	Intake (ng/day)			
	Toddlers		Adults	
	Air	Dust	Air	Dust
∑tri-hepta BDEs	0.11	10	0.6	2.1
BDE209	0.32	270	1.4	100
∑HBCDDs	0.14	25	2.1	15
∑EFRs	0.66	38	4.3	27
α-DBE-DBCH	0.24	0.31	1.7	0.16
β-DBE-DBCH	0.17	0.44	1.2	0.18
TBX	0.037	0.065	0.21	0.027
PBBz	0.02	0.11	0.13	0.047
TBCT	0.038	0.42	0.16	0.16
PBT	0.042	0.15	0.2	0.054
PBEB	0.005	0.017	0.028	0.009
TBP-DBPE	0.0075	0.058	0.039	0.031
HBB	0.016	0.083	0.11	0.07
EH-TBB	0.0075	0.35	0.053	0.19
BTBPE	0.019	0.31	0.13	1.5
BEH-TEBP	0.008	4.7	0.039	2.6
<i>syn</i> -DDC-CO	0.0038	0.014	0.02	0.058
<i>anti</i> -DDC-CO	0.0023	0.46	0.012	0.52
TBBPA-BDBPE	0.019	23	0.1	15
DBDPE	0.019	7.8	0.1	6.5

455 <sup>a</sup> typical exposure scenario denotes adults and toddlers assumed to inhale air and ingest dust contaminated at the  
 456 median concentration at the average ingestion rate (20 mg/day and 50 mg/day for adults and toddlers respectively)

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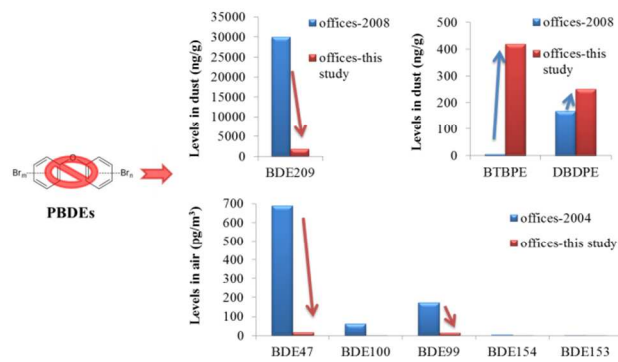


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