

# Concentrations of brominated flame retardants in indoor air and dust from Ireland reveal elevated exposure to decabromodiphenyl ethane

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1 Concentrations of Brominated Flame Retardants in  
2 Indoor Air and Dust from Ireland reveal elevated  
3 exposure to Decabromodiphenyl Ethane

4  
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14 KEYWORDS

15 BFRs, indoor dust, indoor air, cars, homes, offices, schools, human exposure, DBDPE,  
16 PBDEs, HBCDD

17 ABSTRACT

18 Concentrations of decabromodiphenyl ethane (DBDPE), 13 polybrominated diphenyl ethers  
19 (PBDEs) and hexabromocyclododecane (HBCDD), were measured in indoor air and dust  
20 collected from Irish homes, cars, offices and primary schools during 2016-17. Median  
21 concentrations of DBDPE in air (88 pg/m<sup>3</sup>) and dust (6,500 ng/g) exceed significantly those  
22 previously reported internationally, with concentrations highest in offices and schools,  
23 suggesting DBDPE is widely used in Ireland. Median concentrations of BDE-209 in air (340

24  $\text{pg/m}^3$ ) and dust (7,100  $\text{ng/g}$ ) exceed or are within the range of concentrations reported recently  
25 for the same microenvironments in the UK, and exceed those reported in many other countries.  
26 Concentrations of BDE-209 in cars exceeded significantly ( $p < 0.05$ ) those in other micro-  
27 environments. HBCDD was detected in all dust samples (median: 580  $\text{ng/g}$ ), and in 81% of air  
28 samples (median: 24  $\text{pg/m}^3$ ) at concentrations similar to those reported recently for the UK and  
29 elsewhere. Estimates of exposure to DBDPE of Irish adults (92  $\text{ng/day}$ ) and toddlers (210  
30  $\text{ng/day}$ ) as well as to BDE-209 (220  $\text{ng/day}$  and 650  $\text{ng/day}$  for adults and toddlers respectively)  
31 exceed substantially those reported for the UK population. Moreover, our estimates of exposure  
32 of the Irish population to  $\Sigma$ tri-deca-PBDEs exceed previous estimates for Ireland via dietary  
33 exposure.

34

## 35 INTRODUCTION

36 Brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs) and  
37 hexabromocyclododecane (HBCDD) were first marketed in the late 1960s<sup>1</sup> and subsequently  
38 widely applied in many jurisdictions to a variety of soft furnishings, building insulation foams,  
39 electrical and electronic equipment (EEE). There are three main commercial PBDE mixtures  
40 with varying degrees of bromination<sup>2</sup>, namely Penta-BDE (primarily BDE-99 (45-50%) and  
41 BDE-47 (38-42%), Octa-BDE (primarily BDE-183) and Deca-BDE (>90% BDE-209))<sup>3</sup>. Deca-  
42 BDE was mainly used in high impact polystyrene (HIPS) for electrical housings and in fabric  
43 and soft furnishing applications<sup>4</sup>. Penta-BDE was widely used in treatment of polyurethane  
44 foam, especially for transport and for soft furnishings such as upholstery, mattresses and  
45 textiles, as well as in circuit boards, packaging and textiles<sup>5</sup>; while Octa-BDE was used mainly  
46 in hard plastic acrylonitrile butadiene styrene (ABS) casings, and to a lesser degree HIPS and  
47 EEE<sup>5,6</sup>.

48 HBCDD has primarily been used as a flame retardant (FR) in expanded and extruded  
49 polystyrene (EPS/XPS) used in building insulation foam, in textiles<sup>7</sup> and in HIPS used for  
50 EEE<sup>5</sup>. Both HBCDDs and PBDEs are categorised as additive BFRs and are therefore easily  
51 released by volatilisation<sup>8-10</sup> as well as abrasion and direct source-to-dust transfer<sup>11,12</sup> from  
52 treated products into the environment. Human exposure to BFRs is associated with many  
53 adverse effects such as endocrine disruption, liver microsomal enzyme induction,  
54 immunotoxicity, neurotoxicity and carcinogenicity<sup>13,14</sup>. Animal studies have further shown  
55 neurodevelopmental and behavioural outcomes of exposure to PBDEs such as hepatic  
56 abnormality, endocrine disruption and possibly cancer<sup>13,15-17</sup>. In animals, HBCDD was found  
57 to induce hepatic cytochrome P450 enzymes and alter the normal uptake of neurotransmitters,  
58 while in humans it has been reported to trigger cancer through non-mutagenic mechanisms and  
59 disruption of the thyroid hormone system<sup>13,18,19</sup>. This evidence of their toxicity, combined with  
60 their environmental persistence and bioaccumulation potential, has resulted in the listing of

61 Penta- and Octa-BDE (2009), HBCDD (2013) and Deca-BDE (2017) as persistent organic  
62 pollutants (POPs) under the Stockholm Convention<sup>20</sup> leading to restrictions on their  
63 manufacture and use. Whilst banned in 2013, exemptions existed for HBCDD in Europe for  
64 use in EPS/XPS insulation until August 2016<sup>21</sup>. The restrictions on PBDEs have created a  
65 market demand for replacement FRs, such as decabromodiphenyl ethane (DBDPE), marketed  
66 as a replacement for Deca-BDE<sup>22</sup>. Limited toxicological data exists for DBDPE, however, it is  
67 structurally very similar to BDE-209 and may therefore display comparable adverse effects<sup>23</sup>.  
68 BFRs have been detected in human tissues such as breast milk, adipose tissue, blood, serum,  
69 placenta and liver samples<sup>24-27</sup>. Moreover, BFR concentrations in food products, household  
70 dust and air have been recorded worldwide<sup>27-33</sup>. Results suggest multiple external exposure  
71 pathways for BFRs including ingestion of dust, diet, dermal exposure and inhalation<sup>27, 29, 34</sup>.  
72 These data indicate non-occupational human exposure to HBCDDs and PBDEs occurs  
73 primarily by a combination of contact with indoor dust (dermal and/or ingestion), indoor air  
74 and diet<sup>25, 31, 33</sup> and dermal contact with flame-retarded fabrics<sup>35</sup>.

75 Currently, knowledge of exposure of the Irish population to BFRs is confined to a single  
76 biomonitoring study using human breast milk conducted in 2010<sup>27</sup> and ongoing surveillance of  
77 dietary exposure<sup>36,37</sup>. Given the known importance in other countries of indoor exposures, this  
78 study measures PBDEs, HBCDD and DBDPE in indoor air and settled floor dust samples  
79 collected from Irish homes, cars, offices and schools. These data are used to calculate the first  
80 exposure estimates for Irish adults and toddlers to PBDEs, HBCDD and DBDPE via inhalation  
81 and ingestion of indoor dust. The European Union does not have a universal requirement for  
82 upholstered furniture to be resistant to ignition<sup>38</sup>. The UK and Ireland both have specific  
83 stringent fire safety legislation on the flammability of furniture, stating that filling materials  
84 and upholstered covers must be resistant to various sources of ignition<sup>39-41</sup>. This legislation is  
85 specific to the UK and Ireland, and is not a requirement in other European countries<sup>38</sup>. We  
86 therefore tested the hypothesis that concentrations of DBDPE, PBDEs, and HBCDD in Ireland

87 would more closely resemble those in the UK than those in mainland Europe.

## 88 MATERIALS AND METHODS

### 89 *Sampling strategy*

90 Project ethical approval was obtained from the Research Ethics Committee of the National  
91 University of Ireland, Galway (Ref 16/May/02). Project participants were recruited via the  
92 project website ([www.nuigalway.ie/elevate](http://www.nuigalway.ie/elevate)), through articles in the national press, and through  
93 acquaintances of the authors. Paired samples of indoor air and dust were collected from 4  
94 different microenvironment categories (homes (n=32), cars (n=32), offices (n=32) and primary  
95 schools (n=32)) between August 2016 and January 2017 in 3 Irish counties (Galway, Limerick  
96 and Dublin). Prior to the commencement of the study, participants completed a questionnaire  
97 to collect information on the year of construction of the building, the age and presence of  
98 putative sources like electrical items, car age and model etc.

### 99 *Sampling methods*

100 *Indoor dust sampling* - Project sampling protocols followed previously published  
101 methodologies<sup>29</sup>. Home and office participants were asked not to clean their cars for two weeks  
102 or living rooms for two days, prior to sampling. Due to school policy, classroom floors are  
103 cleaned on a daily basis, so samples were taken at the end of the school day from classrooms.  
104 Further details of sample collection protocols are provided as Supplementary data.

105 *Air Sampling* - Air samples were collected by deploying passive air samplers for approximately  
106 60 days in order to sample the maximum volume of air while remaining in the linear uptake  
107 phase of the PUF disk samplers<sup>42</sup>. The sampling apparatus consisted of two parts: a sorbent  
108 (XAD-3) impregnated polyurethane foam disk (PUF) ( $\varnothing$ : 140 mm, thickness: 12 mm, surface  
109 area: 360.6 cm<sup>2</sup>, density: 0.02 g cm<sup>-3</sup>; PACS Leicester, UK), pre-cleaned via Soxhlet extraction  
110 with dichloromethane (DCM) for 8 hours<sup>43</sup>. The PUF was partially enclosed in two stainless-  
111 steel housings (top  $\varnothing$ : 26 cm, bottom :  $\varnothing$ : 18 cm) and mounted according to previous studies<sup>44</sup>.

112 Samplers were placed on elevated surfaces in homes, offices, and schools, and on the floor  
113 behind the passenger or driver's seat in cars.

#### 114 ***Quality Assurance/Quality Control***

115 A reagent blank was analysed with every batch of samples. Instrumental analysis is described  
116 in detail in Supplementary data. None of the target compounds were detected in blank samples  
117 at concentrations above the limit of detection. Therefore, results were not corrected for blank  
118 residues and method limits of detection (LOD) and limits of quantification (LOQ) were  
119 estimated based on S/N = 3:1 and 10:1, respectively. Average LOQs ranged from 0.1 ng/g to  
120 0.4 ng/g for PBDEs, 2.5 ng/g for BDE-209, 13 ng/g for DBDPE and 0.1 ng/g for HBCDDs in  
121 dust (Table SI-1). For air, LOQs were: 0.43 pg/m<sup>3</sup> to 3.1 pg/m<sup>3</sup> for PBDEs, 7.5 pg/m<sup>3</sup> for BDE-  
122 209, 15 pg/m<sup>3</sup> for DBDPE and 0.3 pg/m<sup>3</sup> for HBCDDs (Table SI-2). For non-detects (nd) ½  
123 LOQ was used for statistical analysis. Method accuracy and precision was determined by  
124 analysis of an aliquot of standard reference material SRM-2585 (NIST) with every 10 samples.  
125 Measured concentrations were close to the certified levels with a relative standard deviation  
126 (RSD) of <15% (Table SI -3).

#### 127 ***Statistical Analysis***

128 Statistical analysis was performed using SPSS 24.0. BFR concentration data was log normally  
129 distributed (Kolmogorov-Smirnov test, ( $p>0.05$ )). A one-way ANOVA was used to test the  
130 significance of observed differences in BFR concentrations between microenvironment  
131 categories and regional differences in BFR concentrations for different microenvironments  
132 ( $p<0.05$ ). A two-tailed Pearson's correlation coefficient was used to investigate associations  
133 between air and dust BFR concentrations in homes, cars, offices and schools and factors such  
134 as year of building construction, car age, and number of electronics present. Differences in air  
135 and dust concentrations in offices, schools and homes with electronic goods purchased before  
136 and after 2009 or 2013 or in the presence or absence of room ventilation or presence or absence  
137 of carpets were examined using an independent sample t-test ( $p<0.05$ ).

138

## 139 RESULTS AND DISCUSSION

140 **Concentrations of DBDPE, PBDEs and HBCDDs in indoor dust**

141 All 13 PBDE congeners, DBDPE and HBCDD were detected in all microenvironments (Table  
142 1, and Table SI-6, SI-7, SI-8). After BDE-209 (DF 100%), and HBCDD (99% DF) DBDPE  
143 had the highest detection frequency (98%) (Table SI-9, SI-10).

144 **DBDPE**

145 DBDPE was detected in 98% of all dust samples analysed (n=120) across all  
146 microenvironments ranging from <LOD to concentrations of 540,000 ng/g (median: 6500  
147 ng/g). To the authors' knowledge, our data on concentrations of DBDPE in indoor dust are  
148 both the most recent and contain the highest concentrations reported globally.

149 Highest median concentrations were detected in schools (10,000 ng/g), followed by cars (7,700  
150 ng/g), offices (6,100 ng/g) and homes (4,200 ng/g). Concentration data exceed markedly those  
151 reported for similar microenvironments across Europe, China and Australia<sup>45-48</sup>.

152 These substantially elevated DBDPE concentrations suggest that DBDPE may have been used  
153 as a flame retardant in soft furnishings to meet Irish fire safety requirements for domestic  
154 furniture, which differ from other EU member states (except for the UK)<sup>39-41</sup>. DBDPE is  
155 thought to have replaced Deca-BDE in plastics and textiles with a wide range of uses in the  
156 transport, building, construction, and domestic sectors<sup>49</sup>. DBDPE registrations under REACH<sup>50</sup>  
157 now exceed in weight BDE-209 registrations; 13 importers or manufacturers report a combined  
158 annual tonnage of between 10,000 and 100,000 tonnes of DBDPE in 2018, compared to the  
159 total range of 1,000 and 10,000 for BDE-209 in 2014<sup>49</sup>. Concentrations of DBDPE in UK  
160 indoor dust increased between 2006-07 and 2015<sup>47,51</sup>. While no comparative Irish data exist,  
161 we hypothesise similar increasing temporal trends are occurring in Ireland and thus compared  
162 our Irish data with those in the most recent UK survey. Median DBDPE concentrations in Irish  
163 homes, cars, schools and offices exceed by 100, 77, 610 and 80 times those in UK homes in



164 2016<sup>47</sup>, cars (sampled in 2003-2005)<sup>51</sup>, schools (sampled in 2007-2008)<sup>31</sup> and offices (sampled  
165 in 2016)<sup>47</sup> respectively.

166 Median concentrations of DBDPE reported here for Irish homes exceed those reported for  
167 Swedish homes<sup>52</sup> Australian homes (also sampled in 2016)<sup>48</sup> and homes in Beijing<sup>53</sup>. Median  
168 DBDPE concentrations reported for Irish cars also exceed those in Greece<sup>54</sup>, while those in  
169 Irish offices exceed those in Australia<sup>48</sup>. Moreover, median concentrations in Irish schools  
170 exceed those recently reported for low energy Swedish preschools (300 times higher)<sup>55</sup> and  
171 preschools in Stockholm (100 times higher)<sup>46</sup>.

## 172 **PBDEs**

173 BDE-209 had a detection frequency (DF) of 100% while BDE-47 was detected in 98% of  
174 samples. The following PBDE congeners had DFs > 60%: -196, -197, -183 and -99, while  
175 congeners BDE-17, -28, -49, -66, -100, -154 and -153 had DFs between 3–50%. In terms of  
176 concentrations, BDE-209 was the most abundant congener in all dust samples, followed by  
177 BDE-99>BDE-47>BDE-183. The median concentration of  $\Sigma$ tri-octa BDEs (consisting of  
178 BDE-17, -28, -47, -49, -66, -99, -100, -153, -154, -183, -196 and -197) across all  
179 microenvironments was 43 ng/g dust.

180 In agreement with other international studies, BDE-209 was the most abundant PBDE  
181 congener detected across all microenvironments<sup>28,32,33</sup>, contributing to >99% of  $\Sigma$ PBDEs for  
182 homes, cars and schools, and 98% in offices. This is unsurprising given BDE-209 was the most  
183 commonly used PBDE congener from 2000 until 2008<sup>56</sup> after which its use started to decline  
184 following restrictions, its inclusion in the RoHs Directive and its classification as a SVHC  
185 under REACH<sup>49</sup>. An estimated 10% of the total Deca-BDE imported into the EU between 2000  
186 and 2005, was imported into Ireland<sup>56</sup>. Although no Irish statistics on use of Deca-BDE in  
187 textiles exist, UK data is thought to closely represent usage patterns in Ireland, and an estimated  
188 three quarters of the Deca-BDE used to treat UK textiles was used in domestic furniture<sup>57</sup> while  
189 95% of all upholstered materials used in the UK were treated with flame retardants to comply

190 with the UKFFFSR (UK Furniture and Furnishings (Fire Safety) Regulation)<sup>58</sup>. When  
191 compared with UK data<sup>31,47</sup>, median BDE-209 concentrations in Irish homes (13,000 ng/g) are  
192 higher, while concentrations in Irish offices (3500 ng/g) and schools (8100 ng/g) are consistent  
193 with UK median concentrations reported in 2015 and 2007/08 respectively<sup>31,47</sup>. In contrast,  
194 concentrations in UK cars<sup>59</sup> between 2003-2005 are four times higher than in Irish cars  
195 (median: 26,000 ng/g), perhaps reflecting a downward trend in Deca-BDE use in vehicle  
196 upholstery.

197 Comparisons with other countries reveal BDE-209 concentrations in Irish indoor environments  
198 exceed those reported for Greece<sup>54</sup>, Germany<sup>60</sup> and the Czech Republic<sup>28</sup>. BDE-209  
199 concentrations in Irish homes, offices and cars exceed recent values for Australia<sup>48</sup> as well as  
200 those reported for Beijing homes and offices<sup>53</sup>. BDE-209 median concentrations in Irish  
201 schools also exceed those in Brazil (median: 420 ng/g)<sup>61</sup>.

202 Detection frequencies across all microenvironments for BDE-47 and BDE-99 (97% and 71%)  
203 were high but these congeners were present in lower concentrations than BDE-209. BDE-47  
204 and BDE-99 are typically associated with the Penta-BDE mixture more widely used in North  
205 America<sup>7</sup> than Europe. Concentrations of both congeners in Irish offices, homes and cars are  
206 exceeded by those in Australia, USA and Canada<sup>33,48</sup> but are comparable to those for the Czech  
207 Republic and the UK<sup>33,59</sup>.

208 With respect to  $\Sigma$ tri-octa BDEs, concentrations in Irish homes exceed slightly those in the UK,  
209 Portugal and China<sup>47,62,63</sup>, but are similar to those reported for Brazilian primary schools  
210 (median: 41 ng/g)<sup>61</sup>.

### 211 **HBCDDs**

212 HBCDDs were detected in all samples, with concentrations in Irish homes (median: 490 ng/g)  
213 exceeding those for the UK (110 ng/g) in 2015 and in other international studies<sup>28,64,65</sup>.

214  $\Sigma$ HBCDD concentrations in office, school and car dust (median: 380 ng/g, 800 ng/g, 490 ng/g  
215 respectively) are lower than those for the UK (median: 4,100 ng/g, 4,100 ng/g, 13,000 ng/g

216 respectively)<sup>31,47</sup> for samples collected in 2015 and 2008, which may reflect a downward trend  
217 in HBCDD use in response to recent restrictions.

218  $\Sigma$ HBCDD median concentrations in office dust are exceeded by those in France in 2016  
219 (median: 4,700 ng/g)<sup>66</sup>. There are few published data on concentrations of HBCDD in schools;  
220 results from this study (median: 800 ng/g) are consistent with those for Japan (510 ng/g)<sup>67</sup>, but  
221 the Japanese study was conducted before HBCDD's listing under the Stockholm Convention  
222 in 2013. Concentrations in Irish cars are nearly 40 times higher than in the Czech Republic<sup>28</sup>  
223 and 8 times higher than in Greece<sup>54</sup>.

224 In this study,  $\alpha$ -HBCDD was the dominant isomer in homes, offices and schools, with  $\gamma$ -  
225 HBCDD dominant in cars. The average isomer profile for homes (46%  $\alpha$ -HBCDD, 32%  $\gamma$ -  
226 HBCDD and 22%  $\beta$ -HBCDD and offices (57%  $\alpha$ -HBCDD, 26%  $\beta$ -HBCDD and 17%  $\gamma$ -  
227 HBCDD) is similar to that previously reported in the UK<sup>47</sup> and schools followed the same  
228 pattern (52%  $\alpha$ -HBCDD, 24%  $\gamma$ -HBCDD and 24%  $\beta$ -HBCDD)<sup>31</sup>.

229 In contrast, similar to a Greek study<sup>54</sup>  $\gamma$ -HBCDD was the most abundant isomer in Irish cars  
230 for which the average profile was 45%  $\gamma$ -HBCDD, 38%  $\alpha$ -HBCDD and 18%  $\beta$ -HBCDD.  
231 Previous researchers have also reported  $\gamma$ -HBCDD to dominate in car dust from the Czech  
232 Republic<sup>28</sup> and the UK<sup>59</sup>.

233

#### 234 **Concentrations of DBPDE, PBDEs and HBCDDs in indoor air**

235 There is a dearth of data regarding concentrations of BFRs in indoor air published over the last  
236 five years and so limited comparisons can be made with our data<sup>18,68,69</sup>. Seven of our target  
237 PBDEs, along with DBDPE and HBCDD were detected in all MEs (Table 2, Table SI-11, SI-  
238 12, SI-13). BDE-209 had the highest detection frequency (DF 96%), followed by HBCDD  
239 (81% DF) and DBDPE (65 %) (Table SI-14, SI-15).

#### 240 **DBDPE**

241 This study reports the most recent indoor air data anywhere for DBDPE in homes, schools and  
242 offices and the first data for cars. DBDPE was detected in 65% of air samples, lower than in  
243 Canada (85%)<sup>33</sup> but higher than the UK in 2016<sup>47</sup> (DF: 20%). Similar to our indoor dust data,  
244 concentrations in Irish indoor air are also mostly higher than those reported internationally.  
245 Concentrations of DBDPE in Irish home and offices are >10 and >30 times higher than 2016  
246 UK data (cars and schools were not included in the UK study)<sup>47</sup>.

247 Concentrations of DBDPE in Irish homes are comparable to those in US homes (median: 42  
248 pg/m<sup>3</sup>), but exceed those reported for Canadian and Czech homes<sup>33</sup>. Concentrations in Irish  
249 offices exceed those in Spain<sup>70</sup>, while those in schools exceed those reported for Swedish pre-  
250 schools in 2016-18<sup>55</sup> and Norwegian schools sampled in 2012 (median: 8.3 pg/m<sup>3</sup>)<sup>71</sup>.

#### 251 **PBDEs**

252 BDE-209, -99 and -47 had DFs >90% in all microenvironments, whereas BDE-100, -28, -183,  
253 -154 and -153 were detected in <85 % of air samples, with BDE-197, -196, -49 and -17 not  
254 detected in any sample. The relative abundance of individual congeners in indoor air was:  
255 BDE-209>BDE-99>BDE-47>BDE-183. The median concentrations of PBDEs in indoor air  
256 for all microenvironments were 7.0 pg/m<sup>3</sup> and 300 pg/m<sup>3</sup> for  $\Sigma$ tri-octa BDE (consisting of  
257 BDE-17, -28, -47, -49, -66, -99, -100, -153, -154 -183, -196 and -197) and BDE-209  
258 respectively.

259 BDE-209 was the predominant PBDE congener in all MEs representing 95, 97, 64 and 99% of  
260  $\Sigma$ PBDE for homes, cars, offices and schools respectively, similar to the UK<sup>47</sup>, Sweden<sup>52</sup> and  
261 Germany<sup>72</sup>. Concentrations of BDE-209 in Irish homes are consistent with those for the UK in  
262 2015<sup>47</sup>, but those in Irish offices exceed by a factor of two those recently recorded in the UK<sup>47</sup>.  
263 Concentrations of BDE-209 in Irish homes and cars are lower than those in Sweden<sup>52,73</sup>.

264 It is difficult to make comparisons between the  $\Sigma$ tri-octa BDE concentrations reported here  
265 and elsewhere due to the different congener compositions studied.  $\Sigma$ tri-octa BDE median  
266 concentrations in Irish homes (5.6-330 pg/m<sup>3</sup>) were lower than those reported in the UK

267 (median: 13-2,600 pg/m<sup>3</sup>)<sup>47</sup>, whereas those in Irish offices (median: 15 pg/m<sup>3</sup>, range 5.7-6200  
268 pg/m<sup>3</sup>) exceed those in the UK (median: 20-150 pg/m<sup>3</sup>)<sup>47</sup>. Lowest median concentrations in  
269 this study were in schools (7.0-150 pg/m<sup>3</sup>), and were lower than those in South Korea (<dl-  
270 33,500 pg/m<sup>3</sup>)<sup>74</sup> and Norway (<dl-150 pg/m<sup>3</sup>)<sup>71</sup>.

271 Median concentrations of BDE-47 (2.1 pg/m<sup>3</sup>) and BDE-99 (6.1 pg/m<sup>3</sup>) are exceeded by those  
272 in the US (median: 52 pg/m<sup>3</sup>, 15 pg/m<sup>3</sup>, respectively)<sup>33</sup>. This likely reflects greater use of the  
273 Penta BDE formulation in the US than Europe<sup>7</sup>.

#### 274 **HBCDD**

275 HBCDDs were detected in 81% of all air samples.  $\Sigma$ HBCDD concentrations in Irish homes  
276 (median: 20 pg/m<sup>3</sup>) were 5 times lower than those in the UK (median: 110 pg/m<sup>3</sup>)<sup>47</sup>.  
277 Concentrations in Irish cars (median: 25 pg/m<sup>3</sup>) are almost 500 times lower, and office samples  
278 (14 pg/m<sup>3</sup>) less than half those in the UK in 2008<sup>29</sup> (median: 13,000 pg/m<sup>3</sup>) and 2015<sup>47</sup> (median:  
279 41 pg/m<sup>3</sup>) respectively, which may reflect a decreasing trend in HBCDD use. Nonetheless,  
280 median concentrations in all Irish microenvironments still exceed those in Sweden (<2 pg/m<sup>3</sup>)  
281 in 2006<sup>52</sup>.

282 Unlike the HBCDD isomer pattern in dust, the most abundant isomer in air samples was  $\gamma$ -  
283 HBCDD across all microenvironments, followed by  $\alpha$ -HBCDD. The HBCDD stereoisomer  
284 concentration profile for homes ( $\gamma$ : 62%,  $\alpha$ : 26%,  $\beta$ : 12%), cars ( $\gamma$ : 71%,  $\alpha$ : 22%,  $\beta$ : 7%),  
285 offices ( $\gamma$ : 66%,  $\alpha$ : 27%,  $\beta$ : 7%) and schools ( $\gamma$ : 62%,  $\alpha$ : 27%,  $\beta$ : 11%) is similar to the profiles  
286 in UK homes and offices<sup>47</sup>.

287 Previous studies have also observed the  $\gamma$ -HBCDD isomer to make a greater contribution to  
288  $\Sigma$ HBCDD in air than in dust<sup>29</sup>. This was shown to arise from a photolytically-mediated shift  
289 from  $\gamma$ -HBCDD to  $\alpha$ -HBCDD in dust<sup>75</sup>.

290

#### 291 **Comparisons between microenvironments**

292 Several studies observed differences in concentrations of BFRs between different  
293 microenvironments<sup>47,48,73</sup>. We therefore used one-way ANOVA to establish if there were any  
294 significant ( $p<0.05$ ) differences in BFR concentrations between the microenvironments (ME)  
295 sampled followed by a SNK post-hoc test.

### 296 **Indoor dust**

297 There were no significant differences in DBDPE concentrations between different MEs. For  
298 PBDEs, concentrations of BDE-209 were significantly lower in offices (median: 3,500 ng/g)  
299 than in homes (median: 13,000 ng/g) ( $p<0.05$ ), cars (median: 26,000 ng/g) ( $p<0.05$ ) and  
300 schools (median: 8,100 ng/g) ( $p<0.05$ ). This may suggest declining use of this FR in offices  
301 (and to a lesser extent schools), where faster turnover of electronic and electrical goods than in  
302 homes is anticipated. Concentrations of BDE-99 in cars (median: 50 ng/g) exceed significantly  
303 those in offices ( $p<0.05$ ) and schools ( $p<0.05$ ) but are statistically indistinguishable from those  
304 in homes. Moreover, BDE-183 concentrations are significantly higher in cars (median: 4.1  
305 ng/g) than in offices (median: 3.2 ng/g) ( $p<0.05$ ). Abdallah et al.<sup>29</sup> and McGrath et al.<sup>48</sup> made  
306 similar observations. Higher concentrations of PBDEs have been associated with interiors of  
307 vehicles, due to the increased volume of synthetic surfaces, increased volatilisation of BFRs  
308 due to high temperatures in unoccupied cars as well as to smaller air volume within cars and  
309 reduced ventilation<sup>48</sup>. No other significant differences were observed between MEs.

### 310 **Indoor air**

311 Differences in BFR concentrations in air were observed between the different  
312 microenvironments studied. DBDPE and BDE-209 concentrations in schools were  
313 significantly higher than in offices ( $p<0.05$  for both). DBDPE concentrations in schools  
314 (median: 220 pg/m<sup>3</sup>) exceeded significantly those in homes (median: 48 pg/m<sup>3</sup>) ( $p<0.05$ ); while  
315 BDE-209 concentrations (median: 410 pg/m<sup>3</sup>) in schools exceeded significantly those in cars  
316 (median: 200 pg/m<sup>3</sup>) ( $p<0.05$ ).

317 Concentrations of BDE-47 in homes (median: 2.1 pg/m<sup>3</sup>) were significantly lower compared

318 to those in offices (median: 3.4 pg/m<sup>3</sup>) ( $p < 0.05$ ). BDE-99 concentrations in homes (median:  
319 6.1 pg/m<sup>3</sup>) exceeded significantly higher those in schools and cars (median: 3.1 pg/m<sup>3</sup> and 2.1  
320 pg/m<sup>3</sup> respectively) ( $p < 0.05$ ).

321 The number of putative sources, the cleaning pattern and the location of the sampler relative to  
322 putative sources may influence the sample concentration<sup>29,48,75</sup>. The contextual information  
323 recorded for participating schools, offices, homes and cars was thus examined but provided no  
324 insights into the concentration trends observed.

325

### 326 **Regional differences between microenvironments**

327 The study included samples collected from different regions (Limerick, Galway and Dublin)  
328 in Ireland. We examined our data to establish if there were any differences in BFR  
329 concentrations between each of the microenvironments (ME) between regions in sampled  
330 using one-way ANOVA followed by a Tukey post-hoc test.

### 331 **Indoor dust**

332 Concentrations of DBDPE in dust from Limerick schools (median: 24,000 ng/g) exceeded  
333 significantly those in Galway schools (median: 1,500 ng/g;  $p < 0.001$ ) but were not significantly  
334 higher than in Dublin schools (median: 14,000 ng/g). Two notably high concentrations of  
335 DBDPE (median: 230,000 ng/g and 540,000 ng/g) were found in two Limerick schools,  
336 however the density of electronic and electrical equipment like interactive white boards,  
337 laptops, CD players and foam containing furniture within all participating schools were similar.  
338 Moreover, the purchase of school furniture and electrical equipment is governed by Irish  
339 national policy and not by individual schools or regions. We are therefore unable to explain  
340 the significantly higher DBDPE concentrations in Limerick schools which may be attributable  
341 to the small sample numbers involved (~10 schools from each region).

### 342 **Indoor air**

343 Some statistically significant regional differences in car and school BDE-209 concentrations  
344 were observed. In relation to cars, 10-12 cars were sampled in each region, Dublin car  
345 concentrations (median: 3.8 pg/m<sup>3</sup>) were significantly lower than Galway (median: 300 pg/m<sup>3</sup>)  
346 and Limerick (median: 530 pg/m<sup>3</sup>) cars ( $p < 0.001$ ,  $p < 0.001$ ), which could not be explained by  
347 the contextual data. Most cars in this study were either manufactured in Germany or Asia. A  
348 recent survey of BFRs in Irish waste detected high BDE-209 concentrations in end of life  
349 vehicles manufactured in both Germany and Asia<sup>76</sup>. The cleaning pattern of the cars did not  
350 influence the BDE-209 air concentration and neither did the presence of a child seat or air  
351 conditioning.

352 Some significant ( $p < 0.05$ ) regional differences were observed in concentrations of BDE-209  
353 in schools. Significantly higher BDE-209 concentrations were detected in Galway schools  
354 (median: 930 pg/m<sup>3</sup>) and significant lower concentrations in Dublin schools (median: 150  
355 pg/m<sup>3</sup>). Higher concentrations (not statistically significantly), were observed in Galway  
356 schools in older school buildings (built before 1983) whereas we could not see this trend in  
357 Limerick and Dublin. Nearly all classrooms contained one or more foam containing chairs,  
358 although the age of the chairs was difficult to establish.

359

### 360 **Sources of BFRs in indoor air and dust**

361 Our data was statistically analysed to explore associations between BFR concentrations and  
362 factors such as the number of electronics present in the room and type of floor surface etc.  
363 However, similar to several other international studies, few obvious trends were found<sup>28,30,66</sup>.  
364 This is most likely due to the convenience sampling approach used and the likely variable BFR  
365 presence in putative sources.

366 Concentrations of HBCDD in office dust were positively correlated with the number of  
367 electronics present ( $p < 0.01$ ). Higher air concentrations of  $\Sigma$ HBCDD ( $p < 0.05$ ) were found in  
368 homes (13 out of 32) with carpets. Similar observations have been made in two UK studies<sup>29,47</sup>.



369 As HBCDD was not prevalent in Irish waste electronics or carpets<sup>76</sup>, the cause of these  
370 correlations is unclear. A positive significant correlation ( $p<0.01$ ) was found between  
371 concentrations of BDE-209 in air and the number of electronics in schools. BDE-209 was  
372 widely used in electronic and electrical items up until 2008, and high levels of this FR have  
373 also been detected in waste IT and telecommunication items in Ireland<sup>76</sup>.

374

### 375 **Temporal trends of BFR concentrations in air and dust data**

376 This is the first study of BFR concentrations in Irish indoor air and dust, therefore no  
377 comparisons can be made with previous Irish data. Correlations between BFR concentrations  
378 and year of building construction, car registration and the age of electronics present in the  
379 environment were examined (Pearson correlation, Table SI-16, SI-17). Furthermore,  
380 independent t-tests were used to investigate differences in concentrations from different age  
381 categories. Year of home construction was significantly negatively correlated with  
382 concentrations of  $\Sigma$ HBCDD ( $p<0.01$ ), which possibly reflects the impact of recent restrictions  
383 on the use of HBCDD in building insulation materials.

384 Significantly higher concentrations in air of BDE-209 ( $p<0.05$ ) were found in offices which  
385 had electronics purchased after 2013 ( $n=16$ ;  $540 \text{ pg/m}^3$ ) compared to pre 2013 ( $n=10$ ; average:  
386  $250 \text{ pg/m}^3$ ). Given the recent restrictions on BDE-209 use, this observation is puzzling.

387 Less surprisingly, homes with a greater number of electronics purchased before 2009 (pre 2009  
388  $n=8$ , post 2009  $n=21$ ) had significantly higher concentrations of  $\Sigma$ tri-octa BDE, suggesting a  
389 positive impact from legislative restrictions on octa- and penta-BDE.

390 Year of car registration and concentrations in dust of BDE-47 ( $p<0.01$ ), BDE-99 ( $p<0.05$ ) and  
391  $\Sigma$ tri-octa BDE ( $p<0.05$ ) were negatively correlated. Moreover, concentrations in dust collected  
392 from cars ( $n=19$ ) registered after the listing of Penta and Octa-BDE under the Stockholm  
393 Convention in 2009, were significantly lower than those in dust from cars registered pre 2009  
394 ( $n=10$ ) for congeners BDE-47 ( $p<0.01$ ; average  $42 \text{ ng/g}$ , c.f.  $8.6 \text{ ng/g}$ ), BDE-99 ( $p<0.01$ ;  $89$

395 ng/g, c.f. 31 ng/g) and  $\Sigma$ tri-octa BDE ( $p<0.01$ ; average concentration pre 2009: 255 ng/g, post  
396 2009: 76 ng/g).

397

### 398 **Correlations between air and dust concentrations**

399 We hypothesised concentrations of BFRs in air and dust correlate<sup>47,52,77</sup>, as BFRs partition  
400 between the particulate and the gaseous phase<sup>78</sup>. This hypothesis was tested by examining the  
401 relationship between concentrations of BFRs in air and dust samples collected from the same  
402 MEs using a Spearman's rho test (Table SI-18). Significant positive correlations were observed  
403 for  $\Sigma$ HBCDD (in schools only) ( $p<0.01$ ), for BDE-99 (in homes only) ( $p<0.05$ ), and for BDE-  
404 209 (in homes only), ( $p<0.01$ ). It is unclear why correlations are only evident in certain  
405 microenvironments. This may be due to the limited number of samples in each individual  
406 microenvironment, and therefore further sampling would be required for further investigation.

407

### 408 **Exposure assessment**

409 BFR concentrations in indoor air and dust were used to estimate exposures of Irish adults,  
410 toddlers and school children, via inhalation of airborne BFRs and ingestion of BFRs in dust  
411 (Table 3) (a summary of all the assumptions and algorithms used in exposure calculations are  
412 presented in the supporting information). Two exposure scenarios were considered; a "typical"  
413 exposure scenario using median BFR concentrations and the second a 'high-end exposure  
414 scenario', assuming ingestion/inhalation of the 95<sup>th</sup> percentile BFR concentrations. In addition,  
415 a higher dust ingestion rate was used for the high-end scenario calculation. High-end exposure  
416 scenario estimates for DBDPE (adult: 120 ng/kg bw/day, toddler: 2,500 ng/kg bw/day) and  
417 BDE-209 (adult: 100 ng/kg bw/day, toddler 2,500 ng/kg bw/day) exceed the equivalent high-  
418 end exposures reported recently for the UK<sup>47</sup> (adult: 3.4 ng/kg bw/day, 57 ng/kg bw/day;  
419 toddler: 33 ng/kg bw/day, 1,900 ng/kg bw/day for DBDPE and BDE-209 respectively). By  
420 comparison,  $\Sigma$ HBCDD exposure estimates for adults (7.8 ng/kg bw/day) and toddlers (170

421 ng/kg bw/day) are below UK results (adult: 22 ng/kg bw/day, toddler: 750 ng/kg bw/day)<sup>47</sup>.

422 The high-end exposure estimates (1,100 ng/kg bw/day) for BDE-209 and (86 ng/kg bw/day)

423  $\Sigma$ HBCDD calculated for school children (age 4-6), are below UK values of 330 ng/kg bw/day

424 and 1,300 ng/kg bw/day respectively<sup>31</sup>.

425 High-end estimates of exposure to BDE-209 for Irish adults, toddlers and school children

426 (Table 3) are 100 ng/kg bw/day, 2,500 ng/kg bw/day and 1,100 ng/kg/day respectively and

427 below the USEPA reference dose (RfD) value for adults of 7,000 ng/kg bw/day<sup>79</sup>. Those for

428 Octa-BDE (BDE-183), Penta-BDE (BDE-47 and BDE-99) and  $\Sigma$ HBCDD are also below

429 USEPA guidelines<sup>79,80</sup>. Our estimates of typical adult exposure via inhalation and dust

430 ingestion exceed Irish dietary exposure estimates for BDE-209 (0.3 ng/kg bw/day) but fall

431 below those for  $\Sigma$ PBDEs (2.4 ng/kg bw/day)<sup>81</sup>.

432

433 The limitations of this study are the convenience nature of the sampling that means that the

434 samples analysed are not necessarily representative of Ireland. Moreover, samples taken

435 represent a snapshot of contamination in time and space. Its strengths are that it reveals the

436 presence of elevated concentrations of the legacy BFR BDE-209 and its replacement DBDPE

437 in air and dust from various Irish indoor environments. BDE-209 was the main PBDE congener

438 detected in homes and cars, suggesting substantial use of Deca-BDE to comply with fire safety

439 regulations. In striking contrast, DBDPE was the most abundant BFR detected in Irish offices

440 and school classrooms, suggesting widespread use in Ireland, likely as a replacement for BDE-

441 209 – which is supported by the knowledge that DBDPE has been offered as a direct

442 replacement for DecaBDE, with application at the same concentration of 10-15% by wt<sup>82</sup>. To

443 the authors' knowledge, concentrations of DBDPE in this study were the highest reported in

444 indoor environments anywhere to date. Detailed study of the health implications of exposure

445 to DBDPE are thus recommended.

446 TABLES

447 **Table 1.** Summary of Concentrations of BFRs (ng/g) in Indoor Dust from Irish Homes, Cars, Offices and Schools, together with Median  
 448 Concentrations from selected other studies (\*Median, \*\*Mean).

location	statistical parameter	DBDPE	BDE-209	BDE-47	BDE-99	BDE-183	∑tri-octa-BDEs	α-HBCDD	β-HBCDD	γ-HBCDD	∑-HBCDD
	<i>n</i>	29	29	29	29	29	29	26	26	26	26
homes (ng/g)	<b>Median</b>	4,200	13,000	7.6	13	1	49	200	100	200	490
	<b>Range</b>	410-460,000	140-650,000	0.6-240	<0.2-500	<0.3-33	10-940	0.31-28,000	0.12-12,000	0.83-5,600	1.3-43,000
	<b>Mean</b>	39,000	58,000	26	45	4.1	130	1,500	680	670	2900
	<b>UK</b> <sup>47*</sup>	<10 (<10-97)	4,500 (160-370,000)	13 (0.15-1,700)	12 (0.05-1,700)	<1.0 (<1.0-12)		<2.6 (<2.6-400)	<2.2 (<2.2-160)	110 (16-1,400)	110 (19-1,500)
	<b>USA</b> <sup>33,65*</sup>	150 (nd-3,100)	2,200 (75-7,500)	270 (20-1,300)	340 (20-2,800)	11 (nd-37)		62 (17-910)	16 (7-230)	73 (13-790)	160 (39-1,800)
	<b>Czech Republic</b> <sup>28*</sup>	140 (<20-1,700)	375 (41-5,500)	8.9 (<0.1-11)	11.6 (<0.1-95)	3.9 (<0.8-460)		26 (<0.3-280)	7.1 (<0.3-57)	61 (<0.3-740)	93 (<0.3-950)
	<b>Sweden</b> <sup>64*</sup>	150 (943-1,500)	310 (140-310,000)	21 (6.5-460)	17 (<0.74-300)	-		56 (14-1,400)	18 (3.4-730)	37 (2.5-4,000)	110 (20-6,000)
<b>Beijing</b> <sup>53*</sup>	560 (220-3,100)	150 (69-410)	1.3 (0.67-4.2)	4.9 (1.2-25)	0.37 (0.21-3.7)	8.0 (5.1-37)	64 (34-510)	21 (9.8-120)	64 (30-370)	160 (74-1,000)	
<b>Australia</b> <sup>48*</sup>	1,600 (nd-9,000)	1,100 (290-13,000)	56 (nd-2,800)	74 (16-58,000)	<MQL (nd-26)	(18-11,000)					
<b>Brazil</b> <sup>61*</sup>	400 (150-740)	410 (160-1,200)	8.0 (4.5-1,400)	153 (20-290)							
	<i>n</i>	28	28	28	28	28	28	29	29	29	29
cars (ng/g)	<b>Median</b>	7,700	26,000	24	50	4.1	150	330	250	490	1,300
	<b>Range</b>	<13-190,000	14-680,000	<0.1-130	<0.2-270	<0.3-92	0.094-690	3.4-3,700	4.8-2,600	2.4-17,000	2,400-20,000
	<b>Mean</b>	23,000	82,000	31	70	9.8	200	650	410	180	2,800
	<b>UK</b> <sup>32,51*</sup>	100 (<dl-2,900) <sup>51</sup>	100,000 (12,000-2,600,000)	54 (19-7,500)	100 (23-80,00)	7.8 (<dl-67)		20,00 (54-88,00) <sup>29</sup>	740 (16-5,200)	9,600 (27-56,000)	13,000 (190-69,000)
	<b>Germany</b> <sup>60*</sup>	13,00 (110-6,500)	940 (220-3,100)	17 (2.1-43)	32 (1.3-88)	3.7 (1.3-<0.2-17)					
	<b>Czech Republic</b> <sup>28*</sup>	99 (<20-3,600)	170 (<5-33,000)	2.2 (<0.1-280)	<0.1 (<0.1-280)	<0.8 (<0.8-15)		9 (<0.3-45)	<0.3 (<0.3-44)	25 (<0.3-240)	33 (<0.3-240)
<b>Greece</b> <sup>54*</sup>	856 (33-5,200)	2,800 (110-38,000)	9.1 (0.63-9,000)	12 (1.4-11,000)	1.4 (LOD-1,200)	(23-17,000)	90 (<LOQ-1,300)	16 (<LOQ-290)	46 (<LOQ-260)	155 (<LOQ-1,800)	
<b>Brazil</b> <sup>61*</sup>	1,400 (420-3,800)	1,600 (300-4,000)	31 (4.3-190)	100 (8.5-350)							
location	statistical parameter	DBDPE	BDE-209	BDE-47	BDE-99	BDE-183	∑tri-octa-BDEs	α-HBCDD	β-HBCDD	γ-HBCDD	∑-HBCDD
	<i>n</i>	31	31	31	31	31	31	32	32	32	32
	<b>Median</b>	6,100	3,500	7.7	7	3.2	30	220	96	630	380

offices (ng/g)	Range Mean	<13-1300,00 12,000	560-150,000 4,200	0.8-130 16	<0.2-160 26	<0.3-190 11	2.8-770 77	<0.1-4,400 520	18-710 160	8.8-3,300 170	84-5,200 850
<b>UK</b> <sup>47*</sup>		<10 (<10-54)	26 (2.3-350)	6 (0.15-380)	7.9 (1.2-42)	<1.0 (<1.0-3.8)		5.4 (<2.6-31)	<2.2 (<2.2-15)	34 (3.1-320)	41 (5.5-360)
<b>France</b> <sup>66*</sup>								2,700 (540-6,400)	440 (140-1,500)	1,300 (312-6,400)	4,700 (1,100-10,000)
<b>Beijing</b> <sup>53*</sup>		1,000 (580-1,600)	490 (220-2,900)	1.5 (0.49-17)	1.6 (0.82-32)	0.69 (0.58-5.4)	10 (6.2-64)	100 (60-160)	35 (18-54)	93 (32-190)	260 (110-390)
<b>Australia</b> <sup>48*</sup>		1,900 (nd-10,000)	1,500 (nd-7,200)	220 (40-540,0000)	230 (46-1,000,000)	<MQL	(nd-920)				
<b>Brazil</b> <sup>61*</sup>		2000 (840-5000)	4200 (1800-25000)	13 (7.5-34)	30 (12-53)						
<i>n</i>		32	32	32	32	32	32	30	30	30	30
schools (ng/g)	Median Range Mean	10,000 620-540,000 48,000	8,100 200-71,000 17,000	5 1.3-35 9	5.1 <0.2-240 20	<0.3 <0.3-26 2	30 2.5-290 50	420 12-4,100 680	180 71-2,300 350	130 28-6,700 620	800 250-10,000 1700
<b>UK</b> <sup>31*</sup>		98 (<20-2,500)	5,000 (49-88,000)	26 (1.6-120)	36 (1.1-270)	1.2 (<2-48)	100 (23-1,000)	1,400 (24-100,000)	550 (14-67,000)	1,700 (34-72,000)	4,100 (72-89,000)
<b>Japan</b> <sup>67**</sup>		125 (9.0-800)	1,000 (200-4,800)	8.9 (0.68-73)	7.9 (0.58-60)	13 (0.22-110)		340 (18-1,700)	64 (2.4-340)	104 (0-500)	510 (20-2,300)
<b>Sweden</b> <sup>55*</sup>		34 (<2.2-420)	54 (<4.1-1,200)								
<b>Stockholm</b> <sup>46*</sup>		150 (<0.58-300)	69 (<1.9-130)	30 (11-49)	44 (19-68)	5.7 (<2.2-<9.2)		310 (170-4,500)	72 (52-88)	129 (98-160)	510 (380-640)
<b>Brazil</b> <sup>61*</sup>		300 (210-700)	420 (94-1,200)	8.2 (3.2-30)	33 (30-36)						

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456 **Table 2.** Summary of Concentrations of BFRs (pg/m<sup>3</sup>) in Indoor Air from Irish Homes, Cars, Offices and Schools and Median Concentrations  
 457 from selected previous studies

location	statistical parameter	DBDPE	BDE-209	BDE-47	BDE-99	BDE-183	∑tri-octa-BDEs	α-HBCDD	β-HBCDD	γ-HBCDD	∑-HBCDD
	<i>n</i>	28	28	28	28	28	28	32	32	32	32
Homes (pg/m <sup>3</sup> )	<b>Median</b>	48	410	2.1	6.1	<1.1	19	<0.3	<0.3	10	20
	<b>Range</b>	15-7,000	<7.5-5,500	<0.43-28	<0.43-330	<1.1-7.5	5.6-330	<0.3-57	<0.3-34	<0.3-3,500	0.9-25,00
	<b>Mean</b>	390	880	4.9	37	0.8	50	10	3.5	100	110
	<b>UK<sup>47</sup></b>	<10 (<10-97)	170 (23-3,800)	13 (0.15-1,700)	12 (0.05-1,700)	<1.0 (<1.0-12)		<2.6 (<2.6-400)	<2.2 (<2.2-160)	110 (16-1,400)	110 (19-1,500)
	<b>Sweden<sup>52</sup></b>		310 (43-1,100)								2 (<1.6-33)
	<b>Canada<sup>33</sup></b>	9.2 (na-74)	49 (nd-220)	39 (54-760)	5.3 (1.3-73)	1.3 (nd-1.6)					
	<b>Czech<sup>33</sup></b>	-	9.4 (nd-15)	1.6 (0.56-16)	0.29 (0.16-1.4)	0.12 (nd-0.23)					
	<b>USA<sup>33</sup></b>	42 (nd-71)	260 (nd-5,500)	52 (4.5-820)	15 (nd-1,300)	2.5 (nd-5)					
	<i>n</i>	29	29	29	29	29	29	32	32	32	32
Cars (pg/m <sup>3</sup> )	<b>Median</b>	160	200	1.9	2.1	<1.1	11	8.9	<0.3	18	25
	<b>Range</b>	<15-3,200	<7.5-7,100	0.79-19	<0.43-150	<1.1	6.5-200	0.3-170	0.3-62	0.3-2,300	0.9-2,300
	<b>Mean</b>	340	660	3.2	9.1	0.58	19	13	6.2	180	200
	<b>UK<sup>44,51</sup></b>			14.8 (2.9-4,700) <sup>51</sup>	12 (0.0-2,300) <sup>51</sup>			2,000 (54-8,800)	740 (16-5,200)	9,600 (27-56,000)	13,000 (190-69,000) <sup>44</sup>
	<b>Sweden<sup>52</sup></b>		400 (160-2,500)								0.0 (<1.6)
	<i>n</i>	31	31	31	31	31	31	32	32	32	32
Offices (pg/m <sup>3</sup> )	<b>Median</b>	<15	240	3.4	4.2	0.55	15	<0.3	<0.3	9.6	14
	<b>Range</b>	<15-2,800	<7.5-1,600	<0.43-4,800	<0.43-880	<1.1	5.7-6,200	0.3-1,500	0.3-710	0.3-1,500	0.9-2,800
	<b>Mean</b>	240	420	160	48	0.54	230	86	42	90	220
	<b>UK<sup>47</sup></b>	<10 (<10-54)	26 (2.3-350)	6 (0.15-380)	7.9 (1.2-42)	<1.0 (<1.0-3.8)		5.4 (<2.6-31)	<2.2 (<2.2-15)	34 (3.1-320)	41 (5.5-360)
	<b>Sweden<sup>52</sup></b>		3,200 (68-5,800)								0.0 (<1.6)
	<i>n</i>	31	31	31	31	31	31	32	32	32	32
Schools (pg/m <sup>3</sup> )	<b>Median</b>	220	410	2.3	3.1	<1.1	12	16	<0.3	22	38
	<b>Range</b>	<15-3,800	<7.5-21,000	1.5-29	<0.43-99	<1.1-1.4	7.0-150	0.3-210	0.3-4,600	0.3-1,500	0.9-6,300
	<b>Mean</b>	460	1,600	5.2	9.5	0.54	21	33	160	96	280
	<b>Sweden<sup>55</sup></b>	<7.6	<32	17	<14	<2.1		<0.57	<0.41	<1.0	<2.0
	<b>Norway<sup>71</sup></b>	8.3	<MLD	130	23	<MLD	180				
	<b>South Korea<sup>74</sup></b>	-	0.21 (nd-3.6)	0.40 (nd-17)	0.28 (nd-13)	0.015 (nd-0.15)					

458 **Table 3.** Estimates of exposure (ng/kg bw/day) of Irish adults, toddlers and school children to FRs via indoor air, inhalation and dust ingestion  
 459 under typical<sup>a</sup> and high-end<sup>b</sup> exposure scenarios<sup>c</sup>.

			$\alpha$ -HBCDD	$\beta$ -HBCDD	$\gamma$ -HBCDD	$\Sigma$ -HBCDD	DBDPE	BDE-209	BDE-47	BDE-99	BDE-183	$\Sigma$ tri-octa-BDEs
<b>adult</b>	air	median	0.00035	0.00025	0.0039	0.0071	0.016	0.13	0.0010	0.0022	0.00040	0.0067
		high	0.073	0.037	0.34	0.46	1.2	1.7	0.012	0.10	0.0023	0.10
	dust	median	0.075	0.029	0.085	0.14	1.3	3.1	0.0023	0.0036	0.00045	0.013
		high	2.4	0.83	4.1	7.4	120	99	0.13	0.14	0.017	0.35
	total	median	0.075	0.029	0.088	0.14	1.3	3.2	0.0033	0.0058	0.00084	0.020
		high	2.5	0.87	4.4	7.8	120	100	0.14	0.25	0.020	0.45
<i>UK<sup>47</sup></i>	<i>total</i>	<i>high</i>	<i>6.1</i>	<i>3.4</i>	<i>13</i>	<i>22</i>	<i>3.4</i>	<i>57</i>	<i>0.64</i>	<i>0.81</i>	<i>0.093</i>	<i>1.7</i>
<b>toddler</b>	air	median	0.00019	0.000055	0.0037	0.0073	0.019	0.14	0.0008	0.0021	0.00020	0.0067
		high	0.020	0.0066	0.35	0.37	1.4	1.9	0.0083	0.11	0.0025	0.11
	dust	median	0.98	0.51	1.0	2.5	21	64	0.039	0.069	0.0054	0.25
		high	49	18	98	170	2,500	2,500	3.2	3.3	0.31	7.6
	total	median	0.98	0.51	1.0	2.5	21	64	0.040	0.071	0.0056	0.26
		high	49	18	99	170	2,500	2,500	3.2	3.4	0.31	7.7
<i>UK<sup>47</sup></i>	<i>total</i>	<i>high</i>	<i>200</i>	<i>120</i>	<i>430</i>	<i>750</i>	<i>33</i>	<i>1,900</i>	<i>15</i>	<i>24</i>	<i>2.3</i>	<i>100</i>
<b>school child</b>	air	median	0.0008	0.00003	0.0028	0.0052	0.019	0.090	0.00048	0.0012	0.00012	0.0039
		high	0.017	0.074	0.17	0.25	0.62	1.2	0.0042	0.044	0.0010	0.045
	dust	median	0.61	0.30	0.47	1.42	14	29	0.018	0.030	0.0022	0.12
		high	25	11	48	86	1,400	1,100	1.3	1.4	0.14	3.6
	total	median	0.61	0.30	0.47	1.4	14	30	0.019	0.031	0.0023	0.12
		high	25	11	48	86	1,400	1,100	1.3	1.4	0.14	3.7
<i>UK<sup>31</sup></i>	<i>dust</i>	<i>high</i>			<i>330</i>		<i>13,000</i>		<i>4.3</i>			
<b>RfDs<sup>79,80</sup></b>			<b>200,000</b>				<b>7,000</b>	<b>2,000</b>	<b>100</b>	<b>3,000</b>		

460

<sup>a</sup> Typical exposure scenario suggests adult and toddler exposure to air inhalation and dust ingestion at the median concentration at the average ingestion rates (air: 20 m<sup>3</sup>/d<sup>29</sup> for adults and 3.8 m<sup>3</sup>/d toddlers and school children; dust: 20 mg/day for adults and 50 mg/day for toddlers and school children).

<sup>b</sup> High-end exposure scenario suggests adult and toddler exposure to air and dust ingestion at the 95<sup>th</sup> percentile concentration using high ingestion rates (adult 50 mg/day, toddlers and school children 200 mg/day<sup>29</sup>).

<sup>c</sup> All values expressed as ng/kg/bw/day, assuming body weight of 70 kg for adults, 10 kg for toddlers, and 20 kg for school children.

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480 study.481 **SUPPORTING INFORMATION**482 Full details of sampling (including air sampling rates), analytical (including QA/QC data),  
483 and exposure assessment methods; alongside detailed information about BFR concentrations



484 in samples of indoor air and dust from different microenvironment categories in different  
485 locations in Ireland are provided as supporting information. This material is available free of  
486 charge via the Internet at <http://pubs.acs.org>

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