

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

Wemken, Nina; Drage, Daniel; Abdallah, Mohamed; Harrad, Stuart; Coggins, Marie

DOI:

[10.1021/acs.est.9b02059](https://doi.org/10.1021/acs.est.9b02059)

License:

Other (please specify with Rights Statement)

Document Version

Peer reviewed version

Citation for published version (Harvard):

Wemken, N, Drage, D, Abdallah, M, Harrad, S & Coggins, M 2019, 'Concentrations of brominated flame retardants in indoor air and dust from Ireland reveal elevated exposure to decabromodiphenyl ethane', *Environmental Science and Technology*, vol. 53, no. 16, pp. 98269836. <https://doi.org/10.1021/acs.est.9b02059>

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

Publisher Rights Statement:

This document is the Accepted Manuscript version of a Published Work that appeared in final form in *Environmental Science and Technology*, copyright © American Chemical Society after peer review and technical editing by the publisher. To access the final edited and published work see <https://pubs.acs.org/doi/10.1021/acs.est.9b02059>

General rights

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

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

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

When citing, please reference the published version.

Take down policy

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

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

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

4
5 *Nina Wemken*[†], *Daniel Simon Drage*[‡], *Mohamed Abou-Elwafa Abdallah*[‡], *Stuart Harrad*[‡],
6 *Marie Ann Coggins*^{*†}

7 [†]*School of Physics and the Ryan Institute, National University of Ireland, Galway, H91TK33,*
8 *Ireland*

9 [‡]*School of Geography, Earth & Environmental Sciences, University of Birmingham,*
10 *Birmingham B15 2TT, U.K*

11
12 **Corresponding author: marie.coggins@nuigalway.ie*

13
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/m³) 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 pg/m^3) and dust (7,100 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 ng/g), and in 81% of air
28 samples (median: 24 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 ng/day) and toddlers (210
30 ng/day) as well as to BDE-209 (220 ng/day and 650 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 Σ 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 1960s¹ 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 bromination², 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))³. Deca-
42 BDE was mainly used in high impact polystyrene (HIPS) for electrical housings and in fabric
43 and soft furnishing applications⁴. 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 textiles⁵; while Octa-BDE was used mainly
46 in hard plastic acrylonitrile butadiene styrene (ABS) casings, and to a lesser degree HIPS and
47 EEE^{5,6}.

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 textiles⁷ and in HIPS used for
50 EEE⁵. Both HBCDDs and PBDEs are categorised as additive BFRs and are therefore easily
51 released by volatilisation⁸⁻¹⁰ as well as abrasion and direct source-to-dust transfer^{11,12} 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^{13,14}. Animal studies have further shown
55 neurodevelopmental and behavioural outcomes of exposure to PBDEs such as hepatic
56 abnormality, endocrine disruption and possibly cancer^{13,15-17}. 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^{13,18,19}. 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 Convention²⁰ 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 2016²¹. 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-BDE²². Limited toxicological data exists for DBDPE, however, it is
67 structurally very similar to BDE-209 and may therefore display comparable adverse effects²³.
68 BFRs have been detected in human tissues such as breast milk, adipose tissue, blood, serum,
69 placenta and liver samples²⁴⁻²⁷. Moreover, BFR concentrations in food products, household
70 dust and air have been recorded worldwide²⁷⁻³³. Results suggest multiple external exposure
71 pathways for BFRs including ingestion of dust, diet, dermal exposure and inhalation^{27, 29, 34}.
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^{25, 31, 33} and dermal contact with flame-retarded fabrics³⁵.

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 2010²⁷ and ongoing surveillance of
77 dietary exposure^{36,37}. 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 ignition³⁸. 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 ignition³⁹⁻⁴¹. This legislation is
85 specific to the UK and Ireland, and is not a requirement in other European countries³⁸. 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), 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 methodologies²⁹. 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 samplers⁴². 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 cm², density: 0.02 g cm⁻³; PACS Leicester, UK), pre-cleaned via Soxhlet extraction
110 with dichloromethane (DCM) for 8 hours⁴³. The PUF was partially enclosed in two stainless-
111 steel housings (top \varnothing : 26 cm, bottom : \varnothing : 18 cm) and mounted according to previous studies⁴⁴.

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/m³ to 3.1 pg/m³ for PBDEs, 7.5 pg/m³ for BDE-
122 209, 15 pg/m³ for DBDPE and 0.3 pg/m³ 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 Australia⁴⁵⁻⁴⁸.

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)³⁹⁻⁴¹. 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 sectors⁴⁹. DBDPE registrations under REACH⁵⁰
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 2014⁴⁹. Concentrations of DBDPE in UK
160 indoor dust increased between 2006-07 and 2015^{47,51}. 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 2016⁴⁷, cars (sampled in 2003-2005)⁵¹, schools (sampled in 2007-2008)³¹ and offices (sampled
165 in 2016)⁴⁷ respectively.

166 Median concentrations of DBDPE reported here for Irish homes exceed those reported for
167 Swedish homes⁵² Australian homes (also sampled in 2016)⁴⁸ and homes in Beijing⁵³. Median
168 DBDPE concentrations reported for Irish cars also exceed those in Greece⁵⁴, while those in
169 Irish offices exceed those in Australia⁴⁸. Moreover, median concentrations in Irish schools
170 exceed those recently reported for low energy Swedish preschools (300 times higher)⁵⁵ and
171 preschools in Stockholm (100 times higher)⁴⁶.

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 Σ 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^{28,32,33}, contributing to >99% of Σ 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 2008⁵⁶ after which its use started to decline
184 following restrictions, its inclusion in the RoHs Directive and its classification as a SVHC
185 under REACH⁴⁹. An estimated 10% of the total Deca-BDE imported into the EU between 2000
186 and 2005, was imported into Ireland⁵⁶. 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 furniture⁵⁷ 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)⁵⁸. When
191 compared with UK data^{31,47}, 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^{31,47}. In contrast,
194 concentrations in UK cars⁵⁹ 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 Greece⁵⁴, Germany⁶⁰ and the Czech Republic²⁸. BDE-209
199 concentrations in Irish homes, offices and cars exceed recent values for Australia⁴⁸ as well as
200 those reported for Beijing homes and offices⁵³. BDE-209 median concentrations in Irish
201 schools also exceed those in Brazil (median: 420 ng/g)⁶¹.

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 America⁷ than Europe. Concentrations of both congeners in Irish offices, homes and cars are
206 exceeded by those in Australia, USA and Canada^{33,48} but are comparable to those for the Czech
207 Republic and the UK^{33,59}.

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

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^{28,64,65}.

214 Σ 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)^{31,47} for samples collected in 2015 and 2008, which may reflect a downward trend
217 in HBCDD use in response to recent restrictions.

218 Σ HBCDD median concentrations in office dust are exceeded by those in France in 2016
219 (median: 4,700 ng/g)⁶⁶. 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)⁶⁷, 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 Republic²⁸
223 and 8 times higher than in Greece⁵⁴.

224 In this study, α -HBCDD was the dominant isomer in homes, offices and schools, with γ -
225 HBCDD dominant in cars. The average isomer profile for homes (46% α -HBCDD, 32% γ -
226 HBCDD and 22% β -HBCDD and offices (57% α -HBCDD, 26% β -HBCDD and 17% γ -
227 HBCDD) is similar to that previously reported in the UK⁴⁷ and schools followed the same
228 pattern (52% α -HBCDD, 24% γ -HBCDD and 24% β -HBCDD)³¹.

229 In contrast, similar to a Greek study⁵⁴ γ -HBCDD was the most abundant isomer in Irish cars
230 for which the average profile was 45% γ -HBCDD, 38% α -HBCDD and 18% β -HBCDD.
231 Previous researchers have also reported γ -HBCDD to dominate in car dust from the Czech
232 Republic²⁸ and the UK⁵⁹.

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^{18,68,69}. 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%)³³ but higher than the UK in 2016⁴⁷ (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)⁴⁷.

247 Concentrations of DBDPE in Irish homes are comparable to those in US homes (median: 42
248 pg/m³), but exceed those reported for Canadian and Czech homes³³. Concentrations in Irish
249 offices exceed those in Spain⁷⁰, while those in schools exceed those reported for Swedish pre-
250 schools in 2016-18⁵⁵ and Norwegian schools sampled in 2012 (median: 8.3 pg/m³)⁷¹.

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/m³ and 300 pg/m³ for Σ 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 Σ PBDE for homes, cars, offices and schools respectively, similar to the UK⁴⁷, Sweden⁵² and
261 Germany⁷². Concentrations of BDE-209 in Irish homes are consistent with those for the UK in
262 2015⁴⁷, but those in Irish offices exceed by a factor of two those recently recorded in the UK⁴⁷.
263 Concentrations of BDE-209 in Irish homes and cars are lower than those in Sweden^{52,73}.

264 It is difficult to make comparisons between the Σ tri-octa BDE concentrations reported here
265 and elsewhere due to the different congener compositions studied. Σ tri-octa BDE median
266 concentrations in Irish homes (5.6-330 pg/m³) were lower than those reported in the UK

267 (median: 13-2,600 pg/m³)⁴⁷, whereas those in Irish offices (median: 15 pg/m³, range 5.7-6200
268 pg/m³) exceed those in the UK (median: 20-150 pg/m³)⁴⁷. Lowest median concentrations in
269 this study were in schools (7.0-150 pg/m³), and were lower than those in South Korea (<dl-
270 33,500 pg/m³)⁷⁴ and Norway (<dl-150 pg/m³)⁷¹.

271 Median concentrations of BDE-47 (2.1 pg/m³) and BDE-99 (6.1 pg/m³) are exceeded by those
272 in the US (median: 52 pg/m³, 15 pg/m³, respectively)³³. This likely reflects greater use of the
273 Penta BDE formulation in the US than Europe⁷.

274 **HBCDD**

275 HBCDDs were detected in 81% of all air samples. Σ HBCDD concentrations in Irish homes
276 (median: 20 pg/m³) were 5 times lower than those in the UK (median: 110 pg/m³)⁴⁷.
277 Concentrations in Irish cars (median: 25 pg/m³) are almost 500 times lower, and office samples
278 (14 pg/m³) less than half those in the UK in 2008²⁹ (median: 13,000 pg/m³) and 2015⁴⁷ (median:
279 41 pg/m³) 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/m³)
281 in 2006⁵².

282 Unlike the HBCDD isomer pattern in dust, the most abundant isomer in air samples was γ -
283 HBCDD across all microenvironments, followed by α -HBCDD. The HBCDD stereoisomer
284 concentration profile for homes (γ : 62%, α : 26%, β : 12%), cars (γ : 71%, α : 22%, β : 7%),
285 offices (γ : 66%, α : 27%, β : 7%) and schools (γ : 62%, α : 27%, β : 11%) is similar to the profiles
286 in UK homes and offices⁴⁷.

287 Previous studies have also observed the γ -HBCDD isomer to make a greater contribution to
288 Σ HBCDD in air than in dust²⁹. This was shown to arise from a photolytically-mediated shift
289 from γ -HBCDD to α -HBCDD in dust⁷⁵.

290

291 **Comparisons between microenvironments**

292 Several studies observed differences in concentrations of BFRs between different
293 microenvironments^{47,48,73}. 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.²⁹ and McGrath et al.⁴⁸ 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 ventilation⁴⁸. 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/m³) exceeded significantly those in homes (median: 48 pg/m³) ($p<0.05$); while
315 BDE-209 concentrations (median: 410 pg/m³) in schools exceeded significantly those in cars
316 (median: 200 pg/m³) ($p<0.05$).

317 Concentrations of BDE-47 in homes (median: 2.1 pg/m³) were significantly lower compared

318 to those in offices (median: 3.4 pg/m³) ($p<0.05$). BDE-99 concentrations in homes (median:
319 6.1 pg/m³) exceeded significantly higher those in schools and cars (median: 3.1 pg/m³ and 2.1
320 pg/m³ 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^{29,48,75}. 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/m³) were significantly lower than Galway (median: 300 pg/m³)
346 and Limerick (median: 530 pg/m³) 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 Asia⁷⁶. 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/m³) and significant lower concentrations in Dublin schools (median: 150
355 pg/m³). 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^{28,30,66}.
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 Σ HBCDD ($p < 0.05$) were found in
368 homes (13 out of 32) with carpets. Similar observations have been made in two UK studies^{29,47}.

369 As HBCDD was not prevalent in Irish waste electronics or carpets⁷⁶, 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 Ireland⁷⁶.

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 Σ 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 pg/m^3) compared to pre 2013 ($n=10$; average:
386 250 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 Σ 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 Σ 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 ng/g , c.f. 8.6 ng/g), BDE-99 ($p<0.01$; 89

395 ng/g, c.f. 31 ng/g) and Σ 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^{47,52,77}, as BFRs partition
400 between the particulate and the gaseous phase⁷⁸. 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 Σ 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 95th 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 UK⁴⁷ (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, Σ 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)⁴⁷.

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

423 Σ 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 respectively³¹.

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/day⁷⁹. Those for

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

429 USEPA guidelines^{79,80}. 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 Σ PBDEs (2.4 ng/kg bw/day)⁸¹.

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 wt⁸². 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)	Median	4,200	13,000	7.6	13	1	49	200	100	200	490
	Range	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
	Mean	39,000	58,000	26	45	4.1	130	1,500	680	670	2900
	UK ^{47*}	<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)
	USA ^{33,65*}	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)
	Czech Republic ^{28*}	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)
	Sweden ^{64*}	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)
Beijing ^{53*}	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)	
Australia ^{48*}	1,600 (nd-9,000)	1,100 (290-13,000)	56 (nd-2,800)	74 (16-58,000)	<MQL (nd-26)	(18-11,000)					
Brazil ^{61*}	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)	Median	7,700	26,000	24	50	4.1	150	330	250	490	1,300
	Range	<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
	Mean	23,000	82,000	31	70	9.8	200	650	410	180	2,800
	UK ^{32,51*}	100 (<dl-2,900) ⁵¹	100,000 (12,000-2,600,000)	54 (19-7,500)	100 (23-80,00)	7.8 (<dl-67)		20,00 (54-88,00) ²⁹	740 (16-5,200)	9,600 (27-56,000)	13,000 (190-69,000)
	Germany ^{60*}	13,00 (110-6,500)	940 (220-3,100)	17 (2.1-43)	32 (1.3-88)	3.7 (1.3-<0.2-17)					
	Czech Republic ^{28*}	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)
Greece ^{54*}	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)	
Brazil ^{61*}	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
	Median	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
UK ^{47*}		<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)
France ^{66*}								2,700 (540-6,400)	440 (140-1,500)	1,300 (312-6,400)	4,700 (1,100-10,000)
Beijing ^{53*}		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)
Australia ^{48*}		1,900 (nd-10,000)	1,500 (nd-7,200)	220 (40-540,0000)	230 (46-1,000,000)	<MQL	(nd-920)				
Brazil ^{61*}		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
UK ^{31*}		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)
Japan ^{67**}		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)
Sweden ^{55*}		34 (<2.2-420)	54 (<4.1-1,200)								
Stockholm ^{46*}		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)
Brazil ^{61*}		300 (210-700)	420 (94-1,200)	8.2 (3.2-30)	33 (30-36)						

449

450

451

452

453

454

455

456 **Table 2.** Summary of Concentrations of BFRs (pg/m³) 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 ³)	Median	48	410	2.1	6.1	<1.1	19	<0.3	<0.3	10	20
	Range	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
	Mean	390	880	4.9	37	0.8	50	10	3.5	100	110
	UK⁴⁷	<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)
	Sweden⁵²		310 (43-1,100)								2 (<1.6-33)
	Canada³³	9.2 (na-74)	49 (nd-220)	39 (54-760)	5.3 (1.3-73)	1.3 (nd-1.6)					
	Czech³³	-	9.4 (nd-15)	1.6 (0.56-16)	0.29 (0.16-1.4)	0.12 (nd-0.23)					
	USA³³	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 ³)	Median	160	200	1.9	2.1	<1.1	11	8.9	<0.3	18	25
	Range	<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
	Mean	340	660	3.2	9.1	0.58	19	13	6.2	180	200
	UK^{44,51}			14.8 (2.9-4,700) ⁵¹	12 (0.0-2,300) ⁵¹			2,000 (54-8,800)	740 (16-5,200)	9,600 (27-56,000)	13,000 (190-69,000) ⁴⁴
	Sweden⁵²		400 (160-2,500)								0.0 (<1.6)
	<i>n</i>	31	31	31	31	31	31	32	32	32	32
Offices (pg/m ³)	Median	<15	240	3.4	4.2	0.55	15	<0.3	<0.3	9.6	14
	Range	<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
	Mean	240	420	160	48	0.54	230	86	42	90	220
	UK⁴⁷	<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)
	Sweden⁵²		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 ³)	Median	220	410	2.3	3.1	<1.1	12	16	<0.3	22	38
	Range	<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
	Mean	460	1,600	5.2	9.5	0.54	21	33	160	96	280
	Sweden⁵⁵	<7.6	<32	17	<14	<2.1		<0.57	<0.41	<1.0	<2.0
	Norway⁷¹	8.3	<MLD	130	23	<MLD	180				
	South Korea⁷⁴	-	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^a and high-end^b exposure scenarios^c.

			α -HBCDD	β -HBCDD	γ -HBCDD	Σ -HBCDD	DBDPE	BDE-209	BDE-47	BDE-99	BDE-183	Σ tri-octa-BDEs
adult	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>UK⁴⁷</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>
toddler	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>UK⁴⁷</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>
school child	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>UK³¹</i>	<i>dust</i>	<i>high</i>			<i>330</i>		<i>13,000</i>		<i>4.3</i>			
RfDs^{79,80}			200,000				7,000	2,000	100	3,000		

460

^a 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 m³/d²⁹ for adults and 3.8 m³/d toddlers and school children; dust: 20 mg/day for adults and 50 mg/day for toddlers and school children).

^b High-end exposure scenario suggests adult and toddler exposure to air and dust ingestion at the 95th percentile concentration using high ingestion rates (adult 50 mg/day, toddlers and school children 200 mg/day²⁹).

^c 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.

461 AUTHOR INFORMATION

462 **Corresponding Author**463 *Corresponding author: marie.coggins@nuigalway.ie464 **Present Addresses**465 †*School of Physics and the Ryan Institute, National University of Ireland, Galway, H91TK33,*
466 *Ireland*467 ‡*School of Geography, Earth & Environmental Sciences, University of Birmingham,*
468 *Birmingham B15 2TT, U.K*469 **Author Contributions**470 The manuscript was written through contributions of all authors. All authors have given
471 approval to the final version of the manuscript.472 **Funding Sources**473 ELEVATE is funded by the Environmental Protection Agency of Ireland (Grant 2015-HW-
474 MS-4).475 **ACKNOWLEDGEMENTS**476 This project (Grant 2015-HW-MS-4) is funded under the EPA Research Programme 2014-
477 2020. The EPA Research Programme is a Government of Ireland initiative funded by the
478 Department of Communications, Climate Action and Environment. We gratefully
479 acknowledge all the participants for their permission to collect air and dust samples in this
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>

487

488 REFERENCES

- 489 (1) Secretariat of the Stockholm Convention. *Guidance for the inventory, identification and*
490 *substitution of Hexabromocyclododecane (HBCD) under the Stockholm Convention on*
491 *Persistent Organic Pollutants, Draft*; 2015.
- 492 (2) Kirk, R.; Othmer, D. *Kirk-Othmer encyclopedia of chemical technology*, 5th ed.; Wiley:
493 New York, 2007.
- 494 (3) WHO (World Health Organisation). *Environmental Health Criteria 192. Flame*
495 *retardants: a general introduction*; Geneva, 1997.
- 496 (4) Weil, E.; Levchik, S. Polystyrenes and Thermoplastic Styrene Copolymers. In *Flame*
497 *Retardants for Plastics and Textiles: Practical Applications*; Hanser Publishers:
498 Munich, 2009; pp 35–57.
- 499 (5) European Commission. *Final report: Study on waste related issues of newly listed POPs*
500 *and candidate POPs*; Munich, 2011; Vol. 2011.
- 501 (6) UNEP. Technical review of the implications of recycling commercial penta and
502 octabromodiphenyl ethers. *Unep/Pops/Poprc.6/2 1. 2010, August 201* (August), 3–41.
- 503 (7) Bromine Science and Environmental Forum (BSEF); BSEF. Major Brominated Flame
504 Retardants Volume Estimates. Total Market Demand by Region in 2001. www.bsef.com
505 (accessed Feb 12, 2019).
- 506 (8) Alaei, M.; Arias, P.; Sjödin, A.; Bergman, Å. An overview of commercially used
507 brominated flame retardants, their applications, their use patterns in different
508 countries/regions and possible modes of release. *Environ. Int.* **2003**, 29 (6), 683–689.
- 509 (9) Darnerud, P. Toxic effects of brominated flame retardants in man and in wildlife.

- 510 *Environ. Int.* **2003**, 29 (6), 841–853.
- 511 (10) Covaci, A.; Voorspoels, S.; Ramos, L.; Neels, H.; Blust, R. Recent developments in the
512 analysis of brominated flame retardants and brominated natural compounds. *Journal of*
513 *Chromatography A*. 2007, pp 145–171.
- 514 (11) Rauert, C.; Harrad, S.; Stranger, M.; Lazarov, B. Test chamber investigation of the
515 volatilization from source materials of brominated flame retardants and their subsequent
516 deposition to indoor dust. *Indoor Air* **2015**, 25 (4), 393–404.
- 517 (12) Rauert, C.; Harrad, S. Mass transfer of PBDEs from plastic TV casing to indoor dust via
518 three migration pathways - A test chamber investigation. *Sci. Total Environ.* **2015**, 536,
519 568–574.
- 520 (13) Darnerud, P. O. Brominated flame retardants as possible endocrine disrupters. *Int. J.*
521 *Androl.* **2008**, 31 (2), 152–160.
- 522 (14) Vonderheide, A. P.; Mueller, K. E.; Meija, J.; Welsh, G. L. Polybrominated diphenyl
523 ethers: Causes for concern and knowledge gaps regarding environmental distribution,
524 fate and toxicity. *Sci. Total Environ.* **2008**, 400 (1–3), 425–436.
- 525 (15) Hakk, H. Different HBCD stereoisomers are metabolized differently. *Toxicol. Lett.*
526 **2010**, 196, S33–S34.
- 527 (16) Wikoff, D. S.; Birnbaum, L. Human Health Effects of Brominated Flame Retardants. In
528 *Brominated Flame Retardants*; Eljarrat, E., Barceló, D., Eds.; Springer Berlin
529 Heidelberg: Berlin, Heidelberg, 2011; pp 19–53.
- 530 (17) Birnbaum, L. S.; Staskal, D. F. Brominated flame retardants: Cause for concern?
531 *Environ. Health Perspect.* **2004**, 112 (1), 9–17.
- 532 (18) Covaci, A.; Gerecke, A. C.; Law, R. J.; Voorspoels, S.; Kohler, M.; Heeb, N. V.; Leslie,
533 H.; Allchin, C. R.; De Boer, J. Hexabromocyclododecanes (HBCDs) in the environment
534 and humans: A review. *Environ. Sci. Technol.* **2006**, 40 (12), 3679–3688.
- 535 (19) Law, R. J.; Kohler, M.; Heeb, N. V.; Gerecke, A. C.; Schmid, P.; Voorspoels, S.; Covaci,

- 536 A.; Becher, G.; Janák, K.; Thomsen, C. Hexabromocyclododecane challenges scientists
537 and regulators. *Environ. Sci. Technol.* **2005**, *39* (13), 281A-287A.
- 538 (20) Secretariat of the Stockholm Convention. *Stockholm Convention on Persistent Organic*
539 *Pollutants*; United Nations Environment Programme, 2010.
- 540 (21) European Commission. Commission Regulation (EU) 2016/293. *Off. J. Eur. Union*
541 **2016**.
- 542 (22) Arias, P. A. Brominated flame retardants- An overview. The Second International
543 Workshop on Brominated Flame Retardants, BFR 2001. In *The Second International*
544 *Workshop on Brominated Flame Retardants, BFR 2001*; Stockholm, 2001; pp 17–19.
- 545 (23) Hardy, M. L.; Margitich, D.; Ackerman, L.; Smith, R. L. The subchronic oral toxicity
546 of ethane, 1,2-bis(pentabromophenyl) (Saytex 8010) in rats. *Int. J. Toxicol.* **2002**, *21* (3),
547 165–170.
- 548 (24) Abdallah, M. A. E.; Harrad, S. Tetrabromobisphenol-A, hexabromocyclododecane and
549 its degradation products in UK human milk: Relationship to external exposure. *Environ.*
550 *Int.* **2011**, *37* (2), 443–448.
- 551 (25) Abdallah, M. A. E.; Harrad, S. Polybrominated diphenyl ethers in UK human milk:
552 Implications for infant exposure and relationship to external exposure. *Environ. Int.*
553 **2014**, *63*, 130–136.
- 554 (26) Frederiksen, M.; Thomsen, C.; Frøshaug, M.; Vorkamp, K.; Thomsen, M.; Becher, G.;
555 Knudsen, L. E. Polybrominated diphenyl ethers in paired samples of maternal and
556 umbilical cord blood plasma and associations with house dust in a Danish cohort. *Int. J.*
557 *Hyg. Environ. Health* **2010**, *213* (4), 233–242.
- 558 (27) Pratt, I.; Anderson, W.; Crowley, D.; Daly, S.; Evans, R.; Fernandes, A.; Fitzgerald, M.;
559 Geary, M.; Keane, D.; Morrison, J. J.; Reilly, A.; Tlustos, C. Brominated and fluorinated
560 organic pollutants in the breast milk of first-time Irish mothers: is there a relationship to
561 levels in food? *Food Addit. Contam. A, Chem. Anal. Control. Expo. risk Assess.* **2013**,

- 562 30 (10), 1788–1798.
- 563 (28) Kalachova, K.; Hradkova, P.; Lankova, D.; Hajslova, J.; Pulkrabova, J. Occurrence of
564 brominated flame retardants in household and car dust from the Czech Republic. *Sci.*
565 *Total Environ.* **2012**, *441* (441), 182–193.
- 566 (29) Abdallah, M. A. E.; Harrad, S.; Covaci, A. Hexabromocyclododecanes and
567 Tetrabromobisphenol-A in Indoor Air and Dust in Birmingham, UK: Implications
568 for Human Exposure. *Environ. Sci. Technol.* **2008**, *42* (18), 1–7.
- 569 (30) Cunha, S. C.; Kalachova, K.; Pulkrabova, J.; Fernandes, J. O.; Oliveira, M. B. P. P.;
570 Alves, A.; Hajslova, J. Polybrominated diphenyl ethers (PBDEs) contents in house and
571 car dust of Portugal by pressurized liquid extraction (PLE) and gas chromatography-
572 mass spectrometry (GC-MS). *Chemosphere* **2010**, *78* (10), 1263–1271.
- 573 (31) Harrad, S.; Goosey, E.; Desborough, J.; Abdallah, M. A. E.; Roosens, L.; Covaci, A.
574 Dust from U.K. primary school classrooms and daycare centers: The significance of dust
575 as a pathway of exposure of young U.K. children to brominated flame retardants and
576 polychlorinated biphenyls. *Environ. Sci. Technol.* **2010**, *44* (11), 4198–4202.
- 577 (32) Harrad, S.; Ibarra, C.; Abdallah, M. A. E.; Boon, R.; Neels, H.; Covaci, A.
578 Concentrations of brominated flame retardants in dust from United Kingdom cars,
579 homes, and offices: Causes of variability and implications for human exposure. *Environ.*
580 *Int.* **2008**, *34* (8), 1170–1175.
- 581 (33) Venier, M.; Audy, O.; Vojta, Š.; Bečanová, J.; Romanak, K.; Melymuk, L.; Krátká, M.;
582 Kukučka, P.; Okeme, J.; Saini, A.; Diamond, M. L.; Klanova, J. Brominated flame
583 retardants in the indoor environment — Comparative study of indoor contamination
584 from three countries. *Environ. Int.* **2016**, *94*, 150–160.
- 585 (34) Abdallah, M. A. E.; Pawar, G.; Harrad, S. Effect of Bromine Substitution on Human
586 Dermal Absorption of Polybrominated Diphenyl Ethers. *Environ. Sci. Technol.* **2015**,
587 *49* (18), 10976–10983.

- 588 (35) Abdallah, M. A. E.; Harrad, S. Dermal contact with furniture fabrics is a significant
589 pathway of human exposure to brominated flame retardants. *Environ. Int.* **2018**, *118*
590 (May), 26–33.
- 591 (36) (FSAI) Food Safety Authority of Ireland. Monitoring and surveillance series.
592 *Investigation into levels of chlorinated and brominated organic pollutants in carcass*
593 *fat, offal, eggs and milk produced in Ireland*; Dublin, January 2010.
- 594 (37) Tlustos, C.; McHugh, B.; Pratt, I.; Tyrell, L.; McGovern, E. Investigation into Levels of
595 Dioxins, Furans, Polychlorinated Biphenyls and Brominated Flame Retardants in
596 Fishery Products in Ireland. *Mar. Environ. Heal. Ser.* **2006**, *26*, 1–58.
- 597 (38) Alliance for Flame Retardant Free Furniture in Europe. Competitiveness
598 <http://www.safefurniture.eu/competitiveness.html> (accessed Jun 14, 2019).
- 599 (39) Hagen, R.; Karemaker, M.; Larsson, I.; Hörnqvist, A.; Brants, D.; Elorza, J.; Watts, D.;
600 Van Mierlo, E.; Maloziec, D.; Paloluoma, P.; de Witte, L. *Fire safety of upholstered*
601 *furniture and mattresses in the domestic area: European fire services recommendations*
602 *on test methods*; 2017.
- 603 (40) Fire Safety Advice Center. Furniture and Furnishings (Fire Safety) Regulations
604 1988/1989, 1993 and 2010 [http://www.firesafe.org.uk/furniture-and-furnishings-fire-](http://www.firesafe.org.uk/furniture-and-furnishings-fire-safety-regulations-19881989-and-1993/)
605 [safety-regulations-19881989-and-1993/](http://www.firesafe.org.uk/furniture-and-furnishings-fire-safety-regulations-19881989-and-1993/) (accessed Feb 25, 2019).
- 606 (41) Irish Statute Book (eISB). S.I. No. 316/1995 - Industrial Research and Standards (Fire
607 Safety) (Domestic Furniture) Order, 1995
608 <http://www.irishstatutebook.ie/eli/1995/si/316/made/en/print#> (accessed Jul 19, 2018).
- 609 (42) Hazrati, S.; Harrad, S. Calibration of polyurethane foam (PUF) disk passive air samplers
610 for quantitative measurement of polychlorinated biphenyls (PCBs) and polybrominated
611 diphenyl ethers (PBDEs): Factors influencing sampling rates. *Chemosphere* **2007**, *67*,
612 448–455.
- 613 (43) Shoeib, M.; Harner, T.; Sum, C. L.; Lane, D.; Zhu, J.; Lee, S. C.; Lane, D.; Zhu, J.

- 614 Sorbent-Impregnated Polyurethane Foam Disk for Passive Air Sampling of Volatile
615 Fluorinated Chemicals. *Anal. Chem.* **2008**, *80* (3), 675–682.
- 616 (44) Abdallah, M. A. E.; Harrad, S. Modification and calibration of a passive air sampler for
617 monitoring vapor and particulate phase brominated flame retardants in indoor air:
618 Application to car interiors. *Environ. Sci. Technol.* **2010**, *44* (8), 3059–3065.
- 619 (45) Qi, H.; Li, W.; Liu, L.; Zhang, Z.; Zhu, N.; Song, W.; Ma, W.; Li, Y. Levels, distribution
620 and human exposure of new non-BDE brominated flame retardants in the indoor dust of
621 China. *Environ. Pollut.* **2014**, *195*, 1–8.
- 622 (46) Persson, J.; Wang, T. Temporal trends of decabromodiphenyl ether and emerging
623 brominated flame retardants in dust, air and window surfaces of newly built low - energy
624 preschools. *Indoor Air* **2019**, *00*, 1–13.
- 625 (47) Tao, F.; Abdallah, M. A.-E.; Harrad, S. Emerging and Legacy Flame Retardants in UK
626 Indoor Air and Dust: Evidence for Replacement of PBDEs by Emerging Flame
627 Retardants? *Environ. Sci. Technol.* **2016**, *50* (23), 13052–13061.
- 628 (48) McGrath, T. J.; Morrison, P. D.; Ball, A. S.; Clarke, B. O. Concentrations of legacy and
629 novel brominated flame retardants in indoor dust in Melbourne, Australia: An
630 assessment of human exposure. *Environ. Int.* **2018**, *113*, 191–201.
- 631 (49) European Chemical Agency (ECHA). Bis(pentabromophenyl) ether
632 <https://echa.europa.eu/registration-dossier/-/registered-dossier/14217/1#> (accessed Feb
633 12, 2019).
- 634 (50) European Chemical Agency (ECHA). 1,1'-(ethane-1,2-diyl)bis[pentabromobenzene]
635 <https://echa.europa.eu/registration-dossier/-/registered-dossier/15001> (accessed Feb 12,
636 2019).
- 637 (51) Harrad, S.; Hazrati, S.; Ibarra, C. Concentrations of Polychlorinated Biphenyls in Indoor
638 Air and Polybrominated Diphenyl Ethers in Indoor Air and Dust in Birmingham, United
639 Kingdom: Implications for Human Exposure. *Environ. Sci. Technol.* **2006**, *40* (15),

- 640 4633–4638.
- 641 (52) Thuresson, K.; Björklund, J. A.; de Wit, C. A.; Wit, C. A. De. Tri-decabrominated
642 diphenyl ethers and hexabromocyclododecane in indoor air and dust from Stockholm
643 microenvironments 1: Levels and profiles. *Sci. Total Environ.* **2012**, *414* (1), 713–721.
- 644 (53) Wang, J.; Wang, Y.; Shi, Z.; Zhou, X.; Sun, Z. Legacy and novel brominated flame
645 retardants in indoor dust from Beijing, China: Occurrence, human exposure assessment
646 and evidence for PBDEs replacement. *Sci. Total Environ.* **2018**, *618*, 48–59.
- 647 (54) Besis, A.; Christia, C.; Poma, G.; Covaci, A.; Samara, C. Legacy and novel brominated
648 flame retardants in interior car dust – Implications for human exposure. *Environ. Pollut.*
649 **2017**, *230*, 871–881.
- 650 (55) Larsson, K.; Wit, C. A. De; Sellstro, U.; Sahlstro, L.; Lindh, C. H.; Berglund, M.
651 Brominated Flame Retardants and Organophosphate Esters in Preschool Dust and
652 Children’s Hand Wipes. *Environ. Sci. Technol.* **2018**, *52*, 4878–4888.
- 653 (56) European Chemical Agency (ECHA). *Annex XVI consultant report decabde*; London,
654 2014.
- 655 (57) European Chemicals Bureau. *European Union risk assessment report: Diphenyl ether,*
656 *octabromo derivative*; 2003.
- 657 (58) European Chemical Agency (ECHA). *Annex XV dossier Proposal for Identification of*
658 *a PBT / vPvB Substance*; 2012.
- 659 (59) Harrad, S.; Abdallah, M. A. E. Brominated flame retardants in dust from UK cars -
660 Within-vehicle spatial variability, evidence for degradation and exposure implications.
661 *Chemosphere* **2011**, *82* (9), 1240–1245.
- 662 (60) Brommer, S.; Harrad, S.; Van Den Eede, N.; Covaci, A. Concentrations of
663 organophosphate esters and brominated flame retardants in German indoor dust
664 samples. *J. Environ. Monit.* **2012**, No. 14, 2482.
- 665 (61) Cristale, J.; Aragão Belé, T. G.; Lacorte, S.; Rodrigues de Marchi, M. R. Occurrence

- 666 and human exposure to brominated and organophosphorus flame retardants via indoor
667 dust in a Brazilian city. *Environ. Pollut.* **2018**, *237*, 695–703.
- 668 (62) Coelho, S. D.; Sousa, A. C. A.; Isobe, T.; Kim, J.-W.; Kunisue, T.; Nogueira, A. J. A.;
669 Tanabe, S. Brominated, chlorinated and phosphate organic contaminants in house dust
670 from Portugal. *Sci. Total Environ.* **2016**, *569–570*, 442–449.
- 671 (63) Peng, C.; Tan, H.; Guo, Y.; Wu, Y.; Chen, D. Emerging and legacy flame retardants in
672 indoor dust from East China. *Chemosphere* **2017**, *186*, 635–643.
- 673 (64) Sahlström, L. M. O.; Sellström, U.; De Wit, C. A.; Lignell, S.; Darnerud, O. Estimated
674 intakes of brominated flame retardants via diet and dust compared to internal
675 concentrations in a Swedish mother–toddler cohort. *Int. J. Hyg. Environ. Health* **2015**,
676 *218*, 422–432.
- 677 (65) Dodson, R. E.; Perovich, L. J.; Covaci, A.; Dirtu, A. C.; Brody, J. G.; Rudel, R. A. After
678 the PBDE Phase-Out: A Broad Suite of Flame Retardants in Repeat House Dust Samples
679 from California. *Environ. Sci. Technol.* **2012**, *46*, 13056–13066.
- 680 (66) Abou-Elwafa Abdallah, M.; Bressi, M.; Oluseyi, T.; Harrad, S.
681 Hexabromocyclododecane and tetrabromobisphenol-A in indoor dust from France,
682 Kazakhstan and Nigeria: Implications for human exposure. *Emerg. Contam.* **2016**, *2*,
683 73–79.
- 684 (67) Mizouchi, S.; Ichiba, M.; Takigami, H.; Kajiwara, N.; Takamuku, T.; Miyajima, T.;
685 Kodama, H.; Someya, T.; Ueno, D. Exposure assessment of organophosphorus and
686 organobromine flame retardants via indoor dust from elementary schools and domestic
687 houses. *Chemosphere* **2015**, *123*, 17–25.
- 688 (68) Covaci, A.; Voorspoels, S.; Abdallah, M. A. E.; Geens, T.; Harrad, S.; Law, R. J.
689 Analytical and environmental aspects of the flame retardant tetrabromobisphenol-A and
690 its derivatives. *Journal of Chromatography A*. 2009, pp 346–363.
- 691 (69) Frederiksen, M.; Vorkamp, K.; Thomsen, M.; Knudsen, L. E. Human internal and

- 692 external exposure to PBDEs - A review of levels and sources. *Int. J. Hyg. Environ.*
693 *Health* **2009**, *212* (2), 109–134.
- 694 (70) Reche, C.; Viana, M.; Querol, X.; Corcellas, C.; Barceló, D.; Eljarrat, E. Particle-phase
695 concentrations and sources of legacy and novel flame retardants in outdoor and indoor
696 environments across Spain. *Sci. Total Environ.* **2019**, *649*, 1541–1552.
- 697 (71) Cequier, E.; Ionas, A. C.; Covaci, A.; Marc, R. M.; Becher, G.; Thomsen, C.; Marce, R.
698 M.; Becher, G.; Thomsen, C. Occurrence of a broad range of legacy and emerging flame
699 retardants in indoor environments in Norway. *Environ. Sci. Technol.* **2014**, *48* (12),
700 6827–6835.
- 701 (72) Fromme, H.; Körner, W.; Shahin, N.; Wanner, A.; Albrecht, M.; Boehmer, S.; Parlar,
702 H.; Mayer, R.; Liebl, B.; Bolte, G. Human exposure to polybrominated diphenyl ethers
703 (PBDE), as evidenced by data from a duplicate diet study, indoor air, house dust, and
704 biomonitoring in Germany. *Environ. Int.* **2009**, *35* (8), 1125–1135.
- 705 (73) Newton, S.; Sellström, U.; de Wit, C. A.; Sellstro, U.; Wit, C. A. De. Emerging Flame
706 Retardants, PBDEs, and HBCDDs in Indoor and Outdoor Media in Stockholm, Sweden.
707 *Environ. Sci. Technol.* **2015**, *49*, 2912–2920.
- 708 (74) Lim, Y.-W.; Kim, H.-H.; Lee, C.-S.; Shin, D.-C.; Chang, Y.-S.; Yang, J.-Y. Exposure
709 assessment and health risk of poly-brominated diphenyl ether (PBDE) flame retardants
710 in the indoor environment of elementary school students in Korea. *Sci. Total Environ.*
711 **2014**, *470–471*, 1376–1389.
- 712 (75) Harrad, S.; Abdallah, M. A. E.; Covaci, A. Causes of variability in concentrations and
713 diastereomer patterns of hexabromocyclododecanes in indoor dust. *Environ. Int.* **2009**,
714 *35* (3), 573–579.
- 715 (76) Drage, D. S.; Sharkey, M.; Abdallah, M. A. E.; Berresheim, H.; Harrad, S. Brominated
716 flame retardants in Irish waste polymers: Concentrations, legislative compliance, and
717 treatment options. *Sci. Total Environ.* **2018**, *625*, 1535–1543.

- 718 (77) Shoeib, M.; Harner, T.; Ikonomou, M.; Kannan, K. Indoor and Outdoor Air
719 Concentrations and Phase Partitioning of Perfluoroalkyl Sulfonamides and
720 Polybrominated Diphenyl Ethers. *Environ. Sci. Technol.* **2004**, *38* (5), 1313–1320.
- 721 (78) Bennett, D. H.; Scheringer, M.; McKone, T. E.; Hungerbühler, K. Predicting long-range
722 transport: A systematic evaluation of two multimedia transport models. *Environ. Sci.*
723 *Technol.* **2001**, *35* (6), 1181–1189.
- 724 (79) United States Environmental Protection Agency (US EPA). Technical Fact Sheet –
725 Polybrominated Diphenyl Ethers (PBDEs) and Polybrominated Biphenyls (PBBs).
726 **2012**, No. May, 1–5.
- 727 (80) National Research Council (US). Hexabromocyclododecane. In *Toxicological Risks of*
728 *Selected Flame-Retardant Chemicals*; The National Academies Press: Washington,
729 D.C., 2000; pp 53–71.
- 730 (81) Trudel, D.; Tlustos, C.; Von Goetz, N.; Scheringer, M.; Hungerbühler, K. PBDE
731 exposure from food in Ireland: Optimising data exploitation in probabilistic exposure
732 modelling. *J. Expo. Sci. Environ. Epidemiol.* **2010**, *21* (6), 565–575.
- 733 (82) Dungey, S.; Akintoye, L. *Environmental risk evaluation report: 1,1'- (Ethane-1,2-*
734 *diyl)bis[penta-bromobenzene]*, CAS: 84852-53-9; Bristol, 2007.
- 735
- 736