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1 **Children's exposure to hazardous brominated flame retardants in plastic toys**

2

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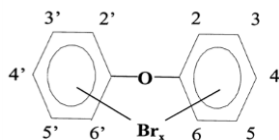
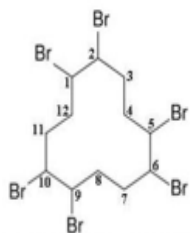
17 **Abstract**

18 We report concentrations of brominated flame retardants (BFRs) in 23 plastic samples from 20
19 new and second-hand children's toys sourced from the UK that had been previously shown to be
20 Br-positive by XRF. The results reinforce existing evidence that the recycling of BFR-treated
21 electronic plastics has led to the unintentional BFR contamination of articles not required to be
22 flame-retarded. The principal BFRs detected were PBDEs (and in particular BDE-209), HBCDD
23 and TBBP-A. PBDEs were detected in all samples with a maximum concentration of BDE-209
24 of 2500 mg/kg, and while TBBP-A was detected in 11 samples with a maximum concentration
25 of 3100 mg/kg. HBCDD was detected in 14 cases and was present in four toys at concentrations
26 (139-840 mg/kg) that would currently prevent their sale on the EU market. While estimated
27 exposures to PBDEs via accidental ingestion of toy plastic fell well below USEPA reference
28 doses, a child weighing 8.67 kg and ingesting 8 mg/day of a toy (the default assumption of the
29 European Commission's Toy Safety Directive for scraped-off toy material) contaminated at our
30 arithmetic mean concentration would be exposed to 0.2 ng/kg bw/day BDE-99. This compares
31 closely to a health-based limit value (HBLV) proposed in The Netherlands of 0.23-0.30 ng/kg
32 bw/day BDE-99. Of greater concern, the same child playing with a toy contaminated at the
33 maximum concentration in this study would be exposed to 1.4 ng/kg bw/day BDE-99, thereby
34 exceeding the HBLV. This paper is the first to consider BFR exposure via incidental ingestion of
35 plastic from both contemporary and historical toys, revealing it to be considerable and for some
36 children their most significant pathway of exposure.

37

38

39 **Graphical abstract**



40

41 **Keywords**

42 recycled plastics; human exposure; POPs; PBDEs; HBCDD; TBBP-A

43

44 **Highlights**

45 Br and BFRs measured in children's plastic toys

46 HBCDD present in 4 toys at concentrations above the EU UTC limit value

47 PBDE concentrations in 2 toys exceed proposed limit of 500 mg/kg

48 TBBP-A detected in 11 samples

49 Children's exposure via unintentional ingestion of toy plastic may be substantial

50

51 **1. INTRODUCTION**

52 Due to evidence of their adverse health effects, brominated flame retardants (BFRs) such as
53 polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) are subject to
54 global bans and restrictions. As a consequence, reports of the presence of BFRs in plastic
55 children's toys as a result of the use of recycled polymers containing such chemicals are of concern
56 (Chen et al., 2009; Ionas et al., 2014; DiGangi et al., 2017; Guzzonato et al., 2017; Puype et al.,
57 2019; Straková and Petrlík, 2017). Children are particularly vulnerable to the adverse health effects
58 of contaminants because of their behavioural tendencies (*e.g.*, mouthing of objects and hand to
59 mouth activities) that differ from adults and result in higher levels of exposure (Landrigan et al.,
60 2011).

61

62 To protect children from migratable substances in toys, the Toy Safety Directive (TSD)
63 2009/48/EC was introduced by the EU (European Commission, 2009). It stipulates that substances
64 classified as carcinogenic, mutagenic, or toxic for reproduction (category 1A, 1B, or 2 and referred
65 to as CMRs) shall not be used in toys or components thereof. While Br and BFRs are not specified
66 under the TSD, in an effort to prevent the contamination of new plastic articles with PBDEs and
67 HBCDD, the EU introduced low persistent organic pollutant (POP) concentration limits (LPCLs).
68 Waste articles, such as plastic casings of end-of-life electronic equipment, that contain HBCDD
69 or PBDEs present in the Penta- and Octa- (and, since 2019, Deca-) BDE formulations at
70 concentrations exceeding the LPCL of 1,000 mg/kg cannot be recycled until their PBDE and
71 HBCDD content has been destroyed or irreversibly transformed (European Commission, 2014;
72 2016). However, the LCPL is currently being reviewed with the aim of adopting legislative limits
73 that are lower than 500 mg/kg as quickly as possible and no later than 2021 (European
74 Commission, 2019). Moreover, the EU has also introduced an Unintentional Trace Contaminant
75 (UTC) limit for HBCDD of 100 mg/kg (European Commission, 2016), with UTC limits for tetra-

76 , penta-, hexa-, hepta- and decaBDE of 10 mg/kg each to be introduced in July 2021 (European
77 Commission, 2019).

78

79 In addition to PBDEs and HBCDD, other BFRs have been used in a variety of applications to
80 impart flame retardancy to polymers such as electrical items and fabrics. The most widely used of
81 these is tetrabromobisphenol-A (TBBP-A) (Abdallah, 2016), while others such as
82 decabromodiphenyl ethane (DBPDE), pentabromobenzene (PBBz), hexabromobenzene (HBB),
83 pentabromotoluene (PBT), and pentabromoethylbenzene (PBEB) are also reported to have been
84 used (Covaci et al., 2011). As use of the latter five BFRs is thought to have increased in recent
85 years in response to restrictions on “legacy” BFRs like PBDEs and HBCDD, compounds such as
86 DBDPE, PBBz, HBB, PBT, and PBEB are collectively referred to here as “novel” BFRs (or
87 NBFRs). While TBBP-A and these NBFRs are not subject to restriction, their presence in, for
88 example, indoor air has been demonstrated (Abdallah et al., 2008; Cequier et al., 2014; Newton et
89 al., 2015), and concerns have emerged over their potential adverse health effects (Covaci et al.,
90 2009; Nakari and Huhtala, 2009; Ezechiáš et al., 2012). Given the widespread use of such BFRs,
91 similar concerns exist that they may be present in items containing recycled plastics, with DBDPE
92 being reported to be present in children’s plastic toys purchased in China (Chen et al., 2009).

93

94 Previously, 200 second-hand plastic toys (encompassing plastic components from multi-material
95 toys) sourced in the UK were analysed by X-ray fluorescence (XRF) spectrometry for the
96 presence of hazardous elements (As, Ba, Cd, Cr, Hg, Pb, Sb, Se) regulated in children toys under
97 the TSD, with Br as a proxy metric of BFRs also analysed and detected in many cases (Turner,
98 2018a). A subsequent study also found Br in many plastic toys that had been purchased new,
99 with the majority of Br-positive items black in colour and consistent with the recycling of
100 electronic waste plastic that is often black for cosmetic and economic purposes (Turner, 2018b).

101 The overall aim of this study, therefore, was to measure concentrations of various legacy (*i.e.*
102 PBDEs and HBCDD) and NBRFRs in a selection of toys sourced in the UK that had been shown
103 to be Br-positive (Turner, 2018b), and to use these data to conduct an assessment of exposure of
104 children playing with such toys and the associated health risk. As previous studies of this topic
105 had not considered exposure via inadvertent ingestion of plastic particles, we further aimed to
106 examine such exposure to test the hypothesis that this pathway is a significant source of exposure
107 to infants. Subsidiary aims were to: (a) evaluate the extent to which measurements of elemental
108 Br in toys are attributable to our target BFRs, and (b) identify any exceedances of the EU's
109 current and impending LPCL and UTC limit values for PBDEs and HBCDD.

110

111 **2. MATERIALS AND METHODS**

112 *2.1. Sampling and screening for total Br via XRF*

113 Twenty three plastic components (samples) from 20 new and second-hand toys (Table 1)
114 previously shown to contain Br that is believed to be derived in whole or in part from the
115 recycling of electronic waste plastic (Turner, 2018a; 2018b) were selected for this study. Sample
116 #s 4, 10, and 23 originated from new toys, with all other samples taken from second-hand items.
117 Toys included game pieces, vehicles, parts of figures, items of jewellery and the handle of a
118 dummy. As an additional check on the total Br content of each sample, and to assess the
119 homogeneity of the distribution of Br, a NITON XL3t 700XRF spectrometer was used to make
120 measurements at between two and four points on each sample (Table 1). Before analysis, the
121 surface of each sample was wiped with a clean non-fibrillating tissue to remove any surface dust.
122 The instrument window was then placed as flat as possible against the sample surface and a
123 measurement of Br content conducted for 60 seconds. The limit of quantification for Br was 5
124 mg/kg. Calibration of the XRF was performed by Niton UK using proprietary standards

125 containing varying concentrations of relevant inorganic compounds in a polymer matrix. The
126 instrument was operated in a low density “plastics” mode and with thickness correction.

127

128 *2.2. BFR measurement methods*

129 *2.2.1. Chemicals and reagents*

130 HPLC-grade solvents were used for sample extraction and LC–MS/MS analysis (Fisher
131 Scientific, Loughborough, UK). Concentrated sulphuric acid was purchased from Sigma–Aldrich
132 (St. Louis, MA, USA). Individual α -, β - and γ -HBCDD standards, $^{13}\text{C}_{12}$ α -, β - and γ -HBCDD,
133 d_{18} - γ -HBCDD, individual standards of PBDEs 17, 28, 47, 49, 77, 99, 100, 153, 154, 183,196,
134 197, 209 and 128, $^{13}\text{C}_{12}$ -BDE-209,TBBP-A, $^{13}\text{C}_{12}$ -TBBP-A, PBBz, PBT, PBEB, HBB, and $^{13}\text{C}_6$ -
135 HBB and DBDPE were purchased from Wellington Laboratories (Guelph, ON, Canada).
136 Polychlorinated biphenyl (PCB)-129 was obtained from Qmx laboratories (Thaxted, UK). A
137 certified reference material (CRM) for polypropylene (ERM-EC591), containing certified
138 concentrations of PBDEs, was purchased from IRMM (Brussels, Belgium).

139

140 *2.2.2. BFR extraction and extract purification*

141 Samples were analysed for concentrations of BFRs using a validated in-house method (Abdallah
142 et al., 2017). Briefly, accurately weighed 0.2 g aliquots of each sample where Br was detected by
143 XRF were transferred into 15 mL glass centrifuge tubes and spiked with 20 ng of internal
144 standards ($^{13}\text{C}_{12}$ α -, β - and γ -HBCDD, $^{13}\text{C}_{12}$ -BDE 77, $^{13}\text{C}_{12}$ -BDE-128, $^{13}\text{C}_{12}$ -TBBP-A and $^{13}\text{C}_6$ -
145 HBB) as well as 40 ng $^{13}\text{C}_{12}$ -BDE 209. Samples were extracted with 3 mL CH_2Cl_2 by vortexing
146 for 2 min and sonicating for 5 min. This was repeated with two further 3 mL aliquots of fresh
147 CH_2Cl_2 . Extracts were collected and combined in a separate centrifuge tube and evaporated to
148 near dryness at 40 °C under a gentle stream of nitrogen before being reconstituted in 2 mL of
149 hexane and vortexed to precipitate dissolved plastics. The hexane supernatant was collected and

150 washed with 2 mL of >98 % concentrated sulphuric acid before vortexing for 30 s. Samples
151 were left for 2 h followed by centrifugation at 3000 rpm for 5 min to ensure complete separation
152 of the organic layer. The clean supernatant hexane layer was collected in a glass tube and
153 concentrated to near dryness before reconstitution in 200 μ L of toluene containing 0.1 ng/ μ L
154 PCB-129 and d₁₈- γ -HBCDD for recovery determination (or syringe) standards. The extracts were
155 transferred to auto-sampler vials with glass inserts for quantitative analysis of PBDEs, PBBz,
156 and DBDPE on GC/MS. After GC-MS analysis, the same extracts were solvent-exchanged to
157 methanol ready for determination of HBCDDs and TBBP-A via LC-MS/MS.

158

159 2.2.3. Instrumental Analysis

160 Determination of PBDEs and NBFs was conducted on a ThermoFisher Trace 1310 gas
161 chromatograph coupled to a ThermoFisher ISQ mass spectrometer operated in electron
162 ionization mode and using selective ion monitoring. With a programmable temperature
163 vaporizer, 1 μ L of extracts were injected onto a Restek Rxi-5Sil MS column (15 m \times 0.25 mm \times
164 0.25 μ m film thickness) with He as the carrier gas at a flow rate of 1.5 mL/min (Abdallah et al.,
165 2017).

166

167 HBCDDs and TBBP-A were quantified on a Shimadzu LC-20AB prominence binary pump
168 liquid chromatograph equipped with a SIL-20A auto-sampler, and a DGU-20A3 vacuum
169 degasser coupled to an AB Sciex API 2000 triple quadrupole MS (Abdallah et al., 2017). An
170 Agilent Pursuit XRS3 C18 column (150 mm \times 2 mm id, 3 μ m particle size) eluted with a mobile
171 phase of (i) 1:1 methanol/water with 2 mM ammonium acetate and (ii) methanol at a flow rate of
172 180 μ L/min. The mass spectrometer was operated in negative ESI mode. MS/MS detection
173 operated in the multiple reaction monitoring mode was used for quantitative determination of
174 HBCDD isomers based on m/z 640.6 \rightarrow 79, m/z 652.4 \rightarrow 79, and m/z 657.7 \rightarrow 79 for the native,

175 ¹³C-labelled and d₁₈-labelled HBCD diastereomers, respectively and *m/z* 540.8→79, *m/z*
176 552.8→79 for the native and ¹³C-labelled TBBP-A, respectively.

177

178 2.2.4. QA/QC

179 Average recoveries of internal standards were between 65 % and 78 %. Table SI-1 shows that
180 the concentrations of PBDEs detected in an aliquot of the certified reference material ERM-
181 EC591 compared favourably with the certified values. Limits of quantification (LOQs) were
182 estimated from a signal to noise ratio of 10:1; target compounds were not detected above LOQs
183 in the blanks and results were therefore not corrected for blank residues. Table SI-2 gives the
184 LOQs for all target compounds.

185

186 2.3. Data analysis

187 For the purposes of calculating descriptive statistics, <LOQ values were replaced by *f* × LOQ,
188 where *f* = the detection frequency of a given BFR expressed as a decimal fraction.

189

190 2.4. Exposure estimation methods

191 2.4.1. Exposure via oral ingestion

192 Exposure via oral ingestion (*E*_{oral ingestion} in ng/kg body weight/day) was estimated using the
193 following algorithm:

$$194 \quad E_{\text{oral ingestion}} = C_{\text{toy}} \times m \times (BA/BW)$$

195 where *C*_{toy} is the BFR concentration in the toy (in ng/g), *m* is the mass of toy ingested per day
196 which, by default, is 8 mg/day according to the Toy Safety Directive (Lenzner et al., 2018), *BA* is
197 the bioavailability of the BFR (%), assumed conservatively to equal the bioaccessibility of BFRs
198 of dust in simulated gastrointestinal tract fluid (Abdallah et al., 2012), and *BW* is body weight
199 (kg).

200

201 2.4.2. Exposure via dermal uptake

202 We estimated exposure via dermal uptake (E_{dermal} in ng/kg body weight/day) as follows:

$$203 E_{\text{dermal}} = C_{\text{toy}}^* \times PSA \times IEF \times (AF/BW)$$

204 Here, C_{toy}^* is the BFR concentration of BFR in toy the (in ng/m²) ($C_{\text{toy}}^* = 1.4 \times 0.05 \times C_{\text{toy}}$
205 assuming a 0.05 cm depth of surface and a density of 1.4 g/cm³ equivalent to acrylonitrile
206 butadiene styrene) (Kuang et al., 2018), PSA is the palm surface area exposed dermally to the toy
207 (assumed to be 0.0986 m²) (Chen et al., 2009), IEF is the indoor exposure fraction, or the
208 number of hours per day for which dermal contact with toys occurs (assumed to be 2; Chen et al.,
209 2009), and AF is the absorbed fraction. The latter is based on measured data for the dermal
210 uptake of PBDEs and HBCDD from fabrics over a 24 h contact (Abdallah and Harrad, 2018) and
211 measured data for 24 h contact with solutions of TBBP-A (Abdallah et al., 2015), and is
212 normalised for a 2 h period.

213

214 3. RESULTS AND DISCUSSION

215 3.1. Concentrations of Br and BFRs in children's plastic toys

216 Table 2 reports the average of the replicate measurements of total Br made by XRF for each toy,
217 including components thereof, while Table SI-3 lists each individual Br measurement for these
218 samples. In some cases, total Br appears to be uniformly distributed in the plastic, while in other
219 cases there is evidence for its heterogeneous dispersion in the toy. Table 2 also lists
220 concentrations of the target BFRs in each sample, as well as the median, arithmetic mean, and
221 maximum concentrations for the whole dataset.

222

223 BFRs were detected in all toys tested with summed concentrations ranging from 1.4 mg/kg to
224 about 6140 mg/kg. The principal BFRs detected in the toys were either PBDEs (in particular
225 BDE-209), HBCDD or TBBP-A, with only low concentrations detected of our target NBFRs.

226 The newest toys, purchased in 2017, contained < 20 mg/kg of BFRs but displayed a wide range
227 in compounds historical and new BFRs. Overall, the BFR pattern and absolute concentrations in
228 our samples are within the range of those previously reported for other studies of plastic items
229 containing recycled polymers, including toys and food contact articles (Chen et al., 2009;
230 Guzzonato et al., 2017; Ionas et al., 2014; Kuang et al., 2018; Puype et al., 2015, 2017).

231

232 If our study has quantified all of the Br-containing compounds in a sample, then the total Br and
233 Σ BFR concentrations should be broadly equal, with the latter slightly exceeding the former
234 because Br only constitutes a proportion of the mass of any BFR. Moreover, as we measured
235 BFRs in a single small aliquot of each toy or component thereof and our replicate XRF
236 measurements of Br revealed varying degrees of inhomogeneity of Br distribution within the
237 sample, Br and Σ BFR measurements will deviate in some instances. Specifically, the
238 heterogeneous distribution of Br and BFRs likely explains why Σ BFRs > average Br in sample #s
239 5 (Σ BFRs = 847 mg/kg c.f. Br = 90 mg/kg) and 6 (Σ BFRs = 468 mg/kg c.f. Br = 289 mg/kg);
240 thus, in sample #5, while Br was <LOQ at 2 measurement points, it was 269 mg/kg at a third,
241 and in sample #6, while Br was <LOQ at 1 measurement point, it was 578 mg/kg in the other.
242 Conversely, where Br exceeds Σ BFR substantially for a given sample (*e.g.*, sub-samples #15, 16
243 and 17 from the same toy) this implies that there is another source or sources of Br in that
244 sample. This may either be an organobromine compound, like the NBFR, BTBPE, which was
245 not measured here but was detected at 1,100 mg/kg in a UK plastic kitchen utensil (Kuang et al.,
246 2018), a polymeric BFR (Gouteux et al, 2008; Puype et al, 2017) or an inorganic Br compound.
247 Despite these discrepancies, however, there was a significant relationship between Br and Σ BFR
248 among the samples, with linear regression analysis returning a best fit line of $\text{Br} = 1.39 \Sigma\text{BFR} +$
249 208 ($r^2 = 0.840$, $p < 0.05$).

250

251 *3.2. Do concentrations of PBDEs and HBCDD in toys exceed LPCL and/or UTC limit values?*

252 Two of the samples analysed (#s 2 and 8) exceed the proposed LPCL value of 500 mg/kg for the
253 summed concentrations of PBDEs (including Deca-BDE), and three additional samples (#s 6, 18
254 and 20) exceed the UTC limit set for July 2021 of 10 mg/kg for Deca-BDE alone. With regard to
255 HBCDD, four samples (#s 2, 5, 11 and 12) exceed the UTC limit of 100 mg/kg, although all
256 were purchased before the limit was introduced (March 2016). Overall, eight out of the 23 sub-
257 samples analysed exceed current or impending limit values for restricted BFRs. In all cases, the
258 items exceeding limits were second-hand and manufactured before 2016. This may suggest that
259 measures to eliminate BFRs from toys containing recycled plastic have been effective; however,
260 we only studied three toys manufactured after these measures were introduced and a much larger
261 study is required to fully evaluate the efficacy of these measures.

262

263 A recent report revealed the presence of HBCDD in various toys and Rubik's cubes purchased in
264 the Czech Republic, up to a maximum of 91 mg/kg in a toy shoe (Straková et al., 2017). Of even
265 greater concern, an earlier survey of 95 Rubik's cubes and 16 additional child-related items
266 sourced from 26 countries around the world (DiGangi et al., 2017) revealed a maximum HBCDD
267 concentration of 1,586 mg/kg, with two items exceeding the LPCL value at the time of 1,000
268 mg/kg and seven exceeding the UTC limit value of 100 mg/kg.

269

270 *3.3. The presence of TBBP-A and NBRs in toys*

271 Relatively few reports exist of the presence of BFRs other than PBDEs and HBCDD in plastic
272 toys, and we believe this study to be only the second report of TBBP-A in such products. Our
273 data for concentrations of TBBP-A (range not detected to 3,140 mg/kg) suggest a broadly similar
274 level of contamination to that observed in toys from the Czech Republic, Germany, and Italy
275 (range 210 mg/kg – 7,800 mg/kg) (Guzzonato et al., 2017). Only trace quantities of the target

276 NBFRs were detected in our samples, providing reassurance that their presence in plastic toys is
277 not currently of significant concern.

278

279 *3.4. What are the human exposure implications of the presence of BFRs in toys?*

280 The potential for human exposure arising from the presence of PBDEs in toys has previously
281 been evaluated (Chen et al., 2009; Ionas et al., 2014). These studies identified potential for
282 exposure via inhalation arising from volatilization of PBDEs, mouthing, dermal contact and oral
283 ingestion (transfer of PBDEs to hands and subsequent oral exposure). As both studies employed
284 the same exposure assessment algorithms, the relative importance of the four exposure pathways
285 was identical in both instances. In summary, exposure via mouthing was estimated to
286 predominate for pre-school children, with exposures via the other pathways making relatively
287 minor contributions. While the absolute estimated exposure levels varied due to the differences
288 in BFR concentrations in toys between the two studies, neither study identified exposures via
289 contact with toys to be of significant toxicological concern.

290

291 Detailed examination of the input data employed to estimate exposure via inhalation, dermal
292 contact, and oral ingestion (Chen et al., 2009; Ionas et al., 2014) reveals the key input parameter
293 in each case to be an emission factor derived for volatilization of PBDEs from flame-retarded
294 items like television sets (Kemmlin et al., 2003). While constituting a useful approach to
295 scoping exposure via these pathways, volatilization emission factors from source items
296 containing PBDEs at concentrations well in excess of those present in plastic toys are predicted
297 to overestimate inhalation exposures and are less appropriate when extrapolated to the estimation
298 of dermal and oral ingestion pathways. Similar considerations apply to the volatilization
299 emission factors reported previously (Kemmlin et al., 2003) for HBCDD (for which the
300 emission factors are from flame-retarded expanded and extruded polystyrene), while published

301 emission factors for TBBP-A and DBDPE do not appear to be available (Kemmlein et al., 2003).
302 Regarding mouthing exposure, Chen et al. (2009) based their estimates on experimentally-
303 derived measurements of BFR migration from two toy samples into human saliva. However, the
304 data obtained are expressed as $\text{pg}/\text{cm}^2/\text{minute}$ rather than as a proportion of the BFR mass
305 present in these toys and cannot thus be extrapolated to estimate migration from toys containing
306 different BFR concentrations such as ours. More recently, exposure to a variety of organic
307 contaminants (but not BFRs) present in plastic children's toys has been evaluated, based on
308 inadvertent oral ingestion of small quantities of plastic and subsequent uptake via the
309 gastrointestinal tract (Lenzner et al., 2018).

310

311 Given the aforementioned considerations, we evaluate for the first time exposure to BFRs
312 measured in plastic children's toys for: (a) the oral ingestion of 8 mg/day of toy plastic (in line
313 with the default assumption of the Toy Safety Directive for scrapable toy material), and (b)
314 dermal uptake arising from a child handling toys. As Chen et al. (2009) identified infants aged
315 between 3 and 18 months (and of body weight 8.67 kg) to be at greatest risk, we have evaluated
316 exposure for this age group only.

317

318 Table 3 summarizes our estimates of typical and high-end exposure via both pathways identified
319 above ($E_{\text{oral ingestion}}$ and E_{dermal}) and obtained using both the arithmetic mean and the maximum
320 BFR concentrations, respectively, for the samples shown in Table 2. It is very clear that while
321 dermal exposure does occur for young children, exposure arising from accidental ingestion of
322 plastic from toys is orders of magnitude greater. Also shown in Table 2 are previously published
323 typical and high-end exposure estimates to BFRs for UK children arising from other pathways;
324 namely: diet (Tao et al, 2017), inhalation (Tao et al, 2016), dust ingestion (Tao et al, 2016),
325 dermal contact with BFR-containing fabrics and indoor dust (Abdallah and Harrad, 2018), and

326 breast milk consumption (Tao et al, 2017). Typical and high-end estimates arising from each
327 pathway for Σ PBDEs and Σ HBCDDs are also compared in Figures 1 and 2, respectively.

328

329 It is evident that exposure via incidental oral ingestion of toy plastic can make a very substantial
330 contribution to overall exposure of young children to our target BFRs. Specifically, under the
331 typical scenarios (where the plastic ingested is assumed to contain BFRs at the arithmetic mean
332 concentration determined in this study, along with typical estimates for other pathways),
333 ingestion contributes 31.8 % of overall exposure to Σ PBDEs and 58 % of overall exposure to
334 Σ HBCDD. Under the high-end exposure scenarios (where the plastic ingested is assumed to
335 contain the maximum BFR concentration determined, along with high-end estimates for other
336 pathways), the contribution made by ingestion of toy plastic to overall exposure falls to 17.7 %
337 and 41 % of Σ PBDEs and Σ HBCDD, respectively.

338

339 We also compared exposures to PBDEs via oral ingestion of and dermal uptake from plastic toys
340 with the reference doses (RfDs) promulgated by the USEPA for BDEs 47 and 99 (= 100 ng/kg
341 bw/day) and BDE 209 (= 7,000 ng/kg bw/day) (US EPA, 2019a; 2019b; 2019c). Reassuringly,
342 even under the maximum exposure scenario estimated exposures arising from toys alone or the
343 combined pathways are well below the respective RfD values. However, our maximum exposure
344 estimate arising from toys alone for BDE-99 (1.4 ng/kg bw/d) exceeds the health-based limit
345 value (HBLV) proposed in the Netherlands (Bakker et al., 2008) of 0.23-0.30 ng/kg bw/day.
346 Moreover, the typical exposure estimate from toys (0.2 ng/kg bw/day) is very close to this
347 HBLV.

348

349 **4. Conclusions**

350 This study provides the first evidence of the presence of a range of BFRs in both new and
351 second-hand toys sourced from the UK. These data add to previous evidence from elsewhere in
352 the world that suggest that recycling of BFR-treated plastics has led to the unintentional but
353 widespread contamination of articles not required to meet flame retardancy regulations. Eight out
354 of the twenty plastic toys examined contained concentrations of PBDEs or HBCDD that would
355 now or in the near future prevent their sale on the EU market. Exposure of young children to
356 BFRs via incidental ingestion of plastic from toys has also been evaluated for the first time.
357 Exposure via this route appears to be considerable and for some individuals and BFRs may
358 represent the most significant pathway via which they are exposed.

359

360 **SUPPORTING INFORMATION**

361 Tables showing: (1) concentrations of PBDEs detected in a certified reference material compared
362 to certified values, (2) limits of quantification for target BFRs, and (3) concentrations of total Br
363 detected in replicate measurements made for each toy.

364

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369

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518 **Table 1: Plastic toy samples selected for analysis**

Sample #	Toy Description	Approximate Date of Purchase or manufacture	# total Br measurements (by XRF)
1	Wind-up bug	2005	3
2	Big Eyes spectacles	2005	2
3	Tic Tac Toe game box	2001	2
4	Black Fidget Spinner with metallic Finish	2017	2
5	Motorcycle	2004	3
6	Magnetic compass	2005	2
7	Car chassis I	2004	3
8 ^a	Beads painted gold	1997	2
9	Action Man Binoculars	2006	2
10	Black Fidget spinner	2017	2
11	Circular dice	2005	2
12	Othello games counter	1997	2
13 ^a	Beads painted red	1997	2
14	Entry gate for garage	2004	3
15 ^a	Bead from necklace	2005	3
16 ^a	Link from necklace	2005	2
17 ^a	Pendant from necklace	2005	4
18 ^a	Wheels of jeep	2007	3
19	Car chassis II	2005	4
20	Wheels of airplane	2004	4
21	Piece from board game	2006	4
22	Case of board game	2006	4
23	Handle of child's dummy	2017	3

519 ^a sample #s 8 and 13, and 15, 16, and 17 are sub-samples from the same toys (beads and
520 necklace, respectively)

Table 2: Concentrations (mg kg⁻¹) of selected BFRs and total Br (obtained using XRF) in the plastic toys

Sample #	BDE-47	BDE-100	BDE-99	BDE-153	BDE-183	BDE-197	BDE-196	BDE-209	TBBP-A	HBCDD	PBBz	PBT	PBEB	HBB	DBDPE	ΣBFRs	Average Br
1	0.19	<LOQ	0.21	0.89	4.3	1.8	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.0002	0.0028	7.5	27
2	0.28	<LOQ	0.32	74	97	43	20	570	15	130	0.0003	<LOQ	<LOQ	0.0002	<LOQ	950	1700
3	0.30	<LOQ	0.15	<LOQ	0.21	0.15	<LOQ	2.2	<LOQ	25	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	28	52
4	0.30	0.13	0.39	<LOQ	0.46	<LOQ	<LOQ	2.0	2.3	9.1	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	15	590
5	2.40	0.72	3.7	0.46	0.50	<LOQ	<LOQ	1.8	<LOQ	840	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	850	90
6	0.26	<LOQ	0.31	2.0	13	6.3	4.3	250	190	1.1	0.0004	<LOQ	<LOQ	<LOQ	0.02	470	290
7	0.34	<LOQ	0.66	<LOQ	0.37	<LOQ	<LOQ	<LOQ	<LOQ	0.25	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1.6	21
8	1.4	0.93	2.3	63	360	180	91	290	2500	9.6	0.0022	<LOQ	<LOQ	0.0039	0.22	3500	8100
9	<LOQ	<LOQ	0.49	<LOQ	0.39	<LOQ	<LOQ	<LOQ	<LOQ	0.52	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	1.4	78
10	0.66	<LOQ	0.73	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	5.5	1.2	0.0003	<LOQ	0.0004	<LOQ	<LOQ	8.0	33
11	0.35	0.26	0.41	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	11	360	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	380	380
12	0.56	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	550	0.0003	<LOQ	<LOQ	<LOQ	0.0005	550	510
13	2.0	0.64	3.2	110	<LOQ	280	140	2500	3100	<LOQ	0.0023	<LOQ	<LOQ	<LOQ	0.25	6100	7300
14	0.64	<LOQ	0.74	<LOQ	0.22	<LOQ	<LOQ	<LOQ	28	20	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	49	9
15	<LOQ	<LOQ	<LOQ	<LOQ	7.4	<LOQ	<LOQ	<LOQ	1.9	<LOQ	<LOQ	0.33	<LOQ	0.23	<LOQ	10	960
16	<LOQ	<LOQ	<LOQ	<LOQ	2.7	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.30	0.07	<LOQ	0.12	<LOQ	3.2	840
17	<LOQ	<LOQ	<LOQ	<LOQ	5.2	<LOQ	<LOQ	<LOQ	1.6	<LOQ	0.49	0.13	<LOQ	0.22	<LOQ	7.8	1000
18	<LOQ	<LOQ	<LOQ	<LOQ	2.5	0.35	0.39	12	<LOQ	<LOQ	1.5	<LOQ	<LOQ	<LOQ	0.19	17	76
19	<LOQ	<LOQ	<LOQ	<LOQ	2.0	<LOQ	<LOQ	<LOQ	<LOQ	2.7	0.36	0.04	<LOQ	0.09	<LOQ	5.5	4.3
20	<LOQ	<LOQ	<LOQ	0.24	3.6	1.5	1.3	112	84	1.9	0.61	0.07	<LOQ	0.15	<LOQ	210	850
21	<LOQ	<LOQ	<LOQ	<LOQ	2.2	<LOQ	<LOQ	0.42	<LOQ	<LOQ	0.07	<LOQ	<LOQ	<LOQ	<LOQ	2.8	4.5
22	<LOQ	<LOQ	<LOQ	<LOQ	1.2	0.33	0.56	<LOQ	<LOQ	<LOQ	0.11	<LOQ	<LOQ	<LOQ	<LOQ	2.3	4.0
23	<LOQ	<LOQ	<LOQ	<LOQ	2.4	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.28	<LOQ	<LOQ	0.09	<LOQ	3.0	0.0
Median ^a	0.26	0.00027	0.21	0.0005	2.0	0.0009	0.0007	0.1	0.8	1.1	0.0004	0.0002	0.00002	0.00004	0.0008	15	90
Average ^a	0.42	0.12	0.59	11	22	23	11	160	260	85	0.16	0.03	0.00003	0.04	0.03	570	1000
Maximum	2.4	0.93	3.7	110	360	280	140	2500	3100	840	1.5	0.33	0.0004	0.23	0.25	6100	8100
LPCL ^b								500 ^b									
UTC ^c								10									
UTC ^d										100							

<LOQ denotes not detected

^a for purposes of calculating descriptive statistics, where concentration <LOQ the value has been replaced with $f \times \text{LOQ}$ where f = detection frequency of BFR expressed as a decimal fraction

- ^b Low POP Concentration Limit for Σ PBDEs – to be enforced from 2021
- ^c Unintentional Trace Contaminant limit value for BDE-209 – to be enforced from 2021
- ^d Unintentional Trace Contaminant limit value for HBCDD – current since 2016

Table 3: Estimated exposures (ng/kg bw/day) to BFRs of young children associated with plastic toys and other pathways

Exposure pathway/scenario	BDE-47	BDE-99	BDE-209	ΣPBDEs	ΣHBCDD	ΣNBFRs
<i>E</i> _{oral} ingestion (typical) ^a	0.2	0.2	21	35	64	0.0001
<i>E</i> _{oral} ingestion (high-end) ^a	1.3	1.4	318	520	634	160
<i>E</i> _{dermal} (typical) ^b	0.001	0.001	- ⁱ	0.01	0.2	- ⁱ
<i>E</i> _{dermal} (high-end) ^b	0.007	0.006	- ⁱ	0.092	2.0	- ⁱ
Diet (typical) ^c	nr	nr	nr	4.8	1.0	3.0
Diet (high-end) ^c	nr	nr	nr	26	6.2	19
Breast milk (typical) ^d	17	5.9	0.65	35	17	18
Breast milk (high-end) ^d	41	10	2.8	80	34	350
Dermal contact with dust and fabrics ^e	nr	nr	- ^j	3.9	24	- ^j
Indoor air inhalation and dust ingestion (typical) ^f	0.10	0.17	31	33	2.9	32
Indoor air inhalation and dust ingestion (high-end) ^f	18	28	2200	2300	870	290
RfD ^g	100	100	7000	-	-	-
HBLV ^h	-	0.23-0.30	-	-	-	-

^a assuming that child ingests 8 mg day⁻¹ of toy plastic contaminated at arithmetic mean and maximum concentration for “typical” and high-end exposure

^b assuming that dermal contact occurs with toys contaminated at the arithmetic mean and maximum concentration for “typical” and high-end exposure

^c “typical” and high-end dietary exposures for UK toddlers (Tao et al., 2017)

^d “typical” and high-end exposures for breast-fed UK infants (Tao et al., 2017)

^e sum of exposures via dermal contact with indoor dust and BFR-containing fabrics (Abdallah and Harrad, 2018). Note ΣPBDE exposure for this estimate covers only those congeners present in the Penta-BDE formulation detected in a fabric covering from a US sofa

^f sum of estimates of exposure of UK toddlers via indoor air inhalation and dust ingestion (Tao et al., 2016). For dust ingestion, “typical” exposure assumes median BFR concentration and mean dust ingestion, high-end exposure assumes 95th percentile BFR concentration and high dust ingestion.

^g USEPA Reference dose (US EPA, 2019a; 2019b; 2019c)

^h Health based limit value proposed by Bakker et al. (2008)

ⁱ dermal absorption not detected for BDE-209 and not studied for our target NBFRs

nr = not reported

Figure 1: Relative contribution (expressed as % of total exposure) of selected pathways to exposure of UK young children to Σ PBDEs under (a) typical and (b) high-end scenarios (note no high-end estimate available of dermal exposure via dust and fabrics)

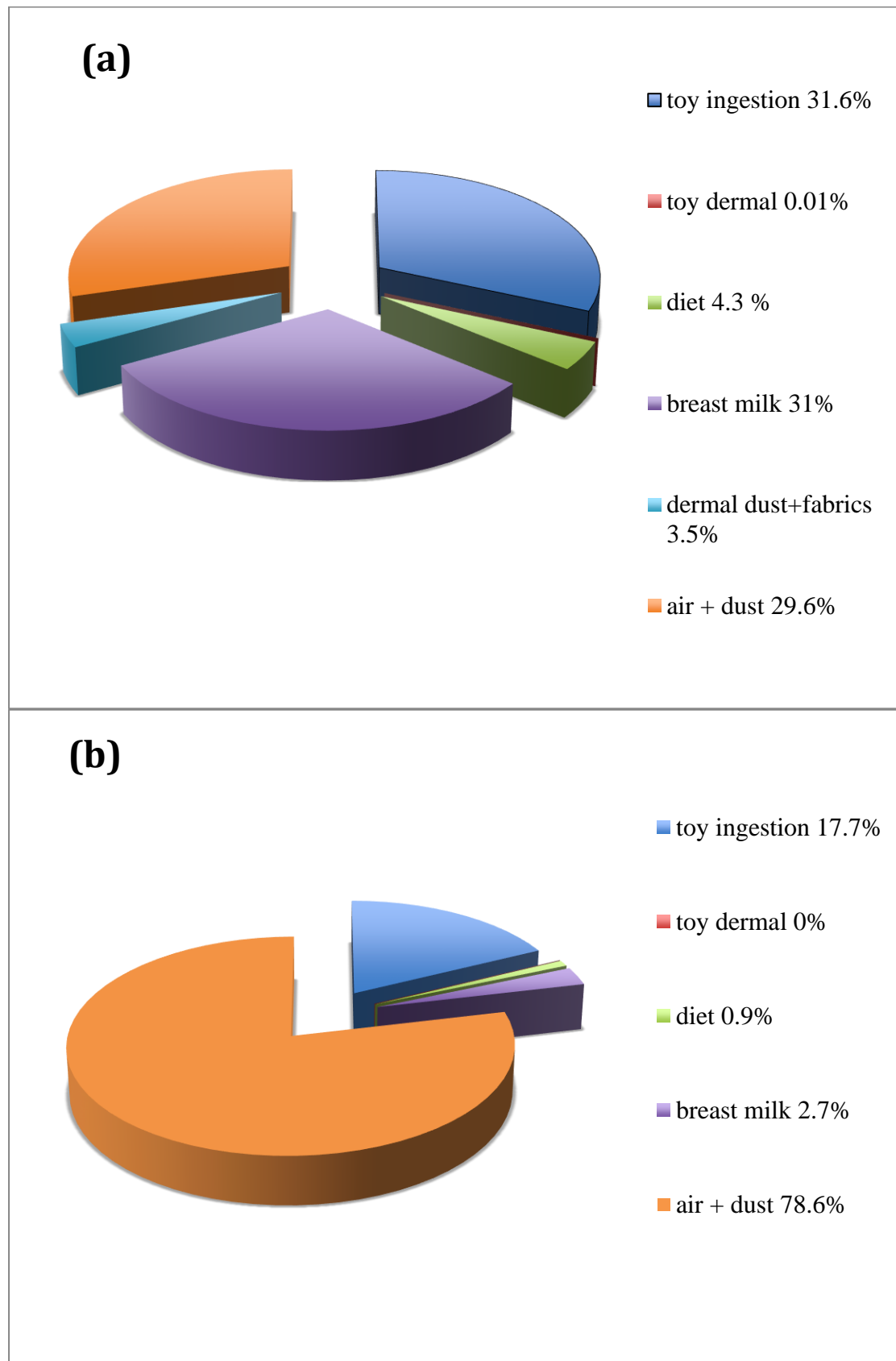


Figure 2: Relative contribution (expressed as %of total exposure) of selected pathways to exposure of UK young children to Σ HBCDD under (a) typical and (b) high-end scenarios (note no high-end estimate available of dermal exposure via dust and fabrics)

