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Formal Waste Treatment Facilities as a Source of Halogenated Flame Retardants and Organophosphate Esters to the Environment: A Critical Review with Particular Focus on Outdoor Air and Soil

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1 Abstract: Extensive use of halogenated flame retardants (HFRs) and organophosphate esters (OPEs) 2 has generated great concern about their adverse effects on environmental and ecological safety and 3 human health. As well as emissions during use of products containing such chemicals, there are 4 mounting concerns over emissions when such products reach the waste stream. Here, we review the 5 available data on contamination with HFRs and OPEs arising from formal waste treatment facilities 6 (including but not limited to e-waste recycling, landfill, and incinerators). Evidence of the transfer 7 of HFRs and OPEs from products to the environment shows that it occurs via mechanisms such as: 8 volatilisation, abrasion, and leaching. Higher contaminant vapour pressure, increased temperature, 9 and elevated concentrations of HFRs and OPEs in products contribute greatly to their emissions to 10 air, with highest emission rates usually observed in the early stages of test chamber experiments. 11 Abrasion of particles and fibres from products is ubiquitous and likely to contribute to elevated FR 12 concentrations in soil. Leaching to aqueous media of brominated FRs (BFRs) is likely to be a 13 second-order process, with elevated dissolved humic matter and temperature of leaching fluids 14 likely to facilitate such emissions. However, leaching characteristics of OPEs are less well-15 understood and require further investigation. Data on the occurrence of HFRs and OPEs in outdoor 16 air and soil in the vicinity of formal e-waste treatment facilities suggests such facilities exert a 17 considerable impact. Waste dumpsites and landfills constitute a potential source of HFRs and OPEs 18 to soil, and improper management of waste disposal might also contribute to HFR contamination in 19 ambient air. Current evidence suggests minimal impact of waste incineration plants on BFR 20 contamination in outdoor air and soil, but further investigation is required to confirm this.

21

22 Keywords: brominated flame retardants, OPFRs, WEEE, incinerators, atmosphere.

24 Halogenated flame retardants (HFRs) and organophosphate esters (OPEs) have been used 25 extensively in various commercial and household products to meet fire safety regulations (Liu et al., 26 2016; Stubbings and Harrad, 2014). Global consumption of brominated flame retardants (BFRs, sum of tetrabromobisphenol A (TBBP-A), hexabromocyclododecane (HBCDD), and three 27 28 commercial mixtures of polybrominated diphenyl ethers (PBDEs)) increased from 204,300 tonnes 29 in 1999 to 410,000 tonnes in 2008 (Bromine Science and Environmental Forum, 2000; Shaw et al., 30 2010). With the global phase-out of PBDEs and HBCDDs, demand for alternative FRs climbed 31 considerably. For instance, the global consumption of OPEs increased sharply from 100,000 tonnes 32 to 1,050,000 tonnes between 1999 and 2018 (Li et al., 2019), and the global production of novel 33 BFRs (NBFRs) was estimated to be 100,000-180,000 tonnes annually (Papachlimitzou et al., 2012), 34 making OPEs and NBFRs the mainstream in current organic FR market. More detailed information 35 on global production/consumption of various HFRs and OPEs is summarised in section 1 and table 36 S1 in Supplementary Material. Due to their persistence and toxicity, HFRs and OPEs may 37 accumulate in sediment and soil, and bioaccumulate in fish, birds, and mammals, thereby exerting 38 adverse impacts on aquatic and terrestrial ecosystem (Anh et al., 2017; Igbal et al., 2017b; Lam et 39 al., 2009; McKinney et al., 2011; Pittinger and Pecquet, 2018; Ross et al., 2009; Tongue et al., 2019; 40 Xie et al., 2018). Therefore, despite the targeted analyses of HFRs and OPEs, novel untargeted 41 screening strategies were also developed to identify unknown brominated compounds and OPEs 42 (Meng et al., 2020; Peng et al., 2016; Ye et al., 2021). In addition, human exposure to HFRs and 43 OPEs is of great concern. Generally, waste recycling and treatment sites (especially those handling 44 electrical/electronic waste, or e-waste, and waste plastics), HFR and OPE production areas, and 45 urban areas (especially in low- and middle-income countries) are the most contaminated areas (Anh 46 et al., 2017; Awasthi et al., 2018; Cao et al., 2014; Huang et al., 2013; Innocentia et al., 2019; Iqbal 47 et al., 2017a; Li et al., 2018a; Ma et al., 2017a, 2017b, 2021; Muenhor et al., 2018; Wang et al., 48 2010a, 2018a; Zeng et al., 2020) – see also section 2 in Supplementary Material. As a result, workers 49 and residents inhabiting these areas are likely to experience high exposure to HFRs and OPEs, with 50 toddlers and infants being of special concern (Die et al., 2019; Ge et al., 2020; Gravel et al., 2019; 51 Li et al., 2018a, 2018c; Ma et al., 2017a, 2017b, 2021; Schecter et al., 2018; Tao et al., 2016; Zeng 53

54 Formal waste treatment activities, which are typically conducted under government licenses that 55 ensure appropriate worker protection and environmental emission safeguards, have been practised 56 widely in high-income countries (Ceballos and Dong, 2016; McGrath et al., 2018). Recycling and recovering, landfilling, and incineration are usually the most common final waste treatments. 57 58 According to the latest data available, in the UK, 221 million tonnes of total waste were generated 59 in 2016. Of this, 48.5% was recycled/recovered, 24.4% landfilled, and 6.1% incinerated, while the 60 remaining 21.0% was disposed of via backfilling (7.8%) or land treatment and release into water 61 bodies (13.2%) (Department for Environment, Food & Rural Affairs, 2020). Generally, formal 62 waste treatment activities have been well-organised and regulated in high-income countries from 63 the early years of the 21st century. For instance, disposal of waste electrical and electronic equipment 64 (WEEE) to landfills has been restricted within the EU since 2003 as a result of the WEEE directive 65 (European Commission, 2003; Harrad et al., 2020b). Related to this, co-disposal of hazardous and 66 non-hazardous wastes in landfills has been abandoned in the UK since 2004, and wastes are 67 categorised into three distinct classes (i.e., hazardous, non-hazardous, and inert waste) prior to their 68 disposal in separate landfills (Stubbings and Harrad, 2014). Similar measures have been taken in 69 the US, where landfills are divided into different categories, namely: 1) municipal solid waste 70 landfills, specifically designed to receive household waste, as well as other types of non-hazardous 71 wastes; 2) industrial waste landfills, designed to collect commercial and institutional waste; 3) 72 hazardous waste landfills, used specifically for the disposal of hazardous waste; and 4) 73 polychlorinated biphenyl (PCB) landfills (U.S. Environmental Protection Agency, 2020). In 74 addition to landfills, formal e-waste recycling has also become a rapidly growing industry in high-75 income countries in response to the need for responsible management of this potentially hazardous 76 waste stream (Gravel et al., 2019). Environmentally-sound techniques have been generally used in 77 these formal e-waste recycling facilities, and controls to reduce exposures, including ventilation and 78 personal protective equipment, are common (Ceballos and Dong, 2016; McGrath et al., 2018; 79 Tomko and McDonald, 2013), though not always implemented (Nguyen et al., 2019).

80

81 In low- and middle-income countries, formal waste treatment facilities are limited, with countries 82 like China and Colombia establishing formal waste treatment facilities in the last few years only 83 (Ceballos and Dong, 2016). In general, it appears that government supervision of the operation of 84 existing facilities is insufficient. For instance, where regulations governing collection of municipal 85 waste is insufficiently enforced, many HFR- and OPE-containing products and materials such as e-86 waste and furniture are landfilled when they reach their end of life (Innocentia et al., 2019; Olukunle 87 and Okonkwo, 2015; Qi et al., 2019). Another example of insufficient government supervision of 88 regulations designed to limit the environmental impacts of waste handling is the coexistence of 89 formal waste recycling companies and informal waste recycling workshops run by individuals or 90 families in certain areas in China (Wang et al., 2018b; Wu et al., 2019).

91

We have previously reviewed the evidence for environmental contamination with HFRs and OPEs as a result of unregulated treatment of e-waste (Ma et al., 2021). Here, we review the available data on contamination with such chemicals arising from formal waste treatment facilities. The present review aims to: 1) identify the pathways of HFR and OPE transfer from FR-containing wastes to the outdoor environment, e.g, volatilisation, abrasion, and leaching; 2) summarise current state-ofknowledge on the occurrence of HFRs and OPEs in outdoor air and soil in the vicinity of formal waste treatment facilities; and 3) highlight substantial research gaps that require investigation.

99

100 2. Methods

101 Data collection was conducted between 17/06/2020 and 20/03/2021, and two electronic databases 102 (ScienceDirect and Web of Science Core Collection) were searched for relevant publications. 103 Various keywords were used in data collection, and the keywords were combined as "flame retardant 104 + keyword 1 + keyword 2". Table S2 (Supplementary Material) shows the keywords that were used 105 as well as the number of publications located on ScienceDirect and Web of Science Core Collection. 106 Specifically, a total of 738 publications were found on ScienceDirect, with a further 1846 papers 107 located on Web of Science Core Collection. These publications were initially rated for relevance by 108 screening titles and abstracts, leaving 192 papers from ScienceDirect and 247 papers from Web of 109 Science Core Collection. After removal of duplicates (n=83), 356 publications remained for further

110 screening.

111

The remaining 356 publications were further identified by screening sampling methodology, statistical data presented, and conclusions. Via this process, 249 papers were removed (including 114 243 irrelevant papers and 6 articles not written in English), leaving 107 articles that were reviewed in this study.

116

117 3. Formal waste treatment facilities as a source of HFRs and OPEs to the outdoor 118 environment

119 **3.1 Emissions of FRs to air**

120 Emission chamber tests have been conducted to study the emissions of HFRs and OPEs from 121 electronics, furnishings, and other commercial products via volatilisation (Kajiwara and Takigami, 122 2013; Kemmlein et al., 2003; Ni et al., 2007; Poppendieck et al., 2017; Rauert and Harrad, 2015; 123 Rauert et al., 2015; Sun et al., 2018; Tokumura et al., 2019). Such emissions likely explain the 124 observed elevated concentrations of HFRs and OPEs in outdoor air in the vicinity of formal waste 125 treatment facilities (Cahill et al., 2007; Hong et al., 2018; Park et al., 2014; Wang et al., 2018a). On 126 the one hand, HFR and OPE emissions to the atmosphere are expected through transport, outdoor 127 storage, and outdoor landfilling of FR-containing wastes; on the other hand, emissions to indoor air 128 could result from indoor waste treatment activities (e.g., indoor storage, classification, testing, 129 recycling, and dismantling, etc.), which then leads to outdoor air contamination through indoor-130 outdoor air exchange (ventilation and infiltration). However, in part due to differences in the design 131 of previous emission chamber tests, the specific emission rates (SER) reported in different studies 132 vary substantially (Table 1). In summary, FR physicochemical properties, chamber temperature, 133 timescale of emission experiments, and concentrations of FRs in the products or materials tested, 134 are possible factors influencing emissions of FRs to air.

135

136 3.1.1 Impact of physicochemical properties of FRs on emissions to air

137 A cubic stainless-steel container (10 cm \times 10 cm \times 5 cm height) was placed in a constant-138 temperature oven (20 °C) to allow the emission rates of tris(1,3-dichloroisopropyl) phosphate 139 (TDCIPP; vapour pressure: 4.1×10^{-4} Pa at 25 °C) and tricresvl phosphate (TCP; vapour pressure: 140 $1.1-5.1 \times 10^{-5}$ Pa at 25 °C) to be measured (Tokumura et al., 2019). Test results showed a substantially 141 higher emission rate of TDCIPP (0.17 μ g·m⁻²·h⁻¹) than TCP (0.060 μ g·m⁻²·h⁻¹), despite the slightly 142 lower concentration of TDCIPP (4,310 $\mu g \cdot g^{-1}$) than TCP (4,840 $\mu g \cdot g^{-1}$) in the curtains. The authors 143 concluded that the vapour pressure of an OPE exerted an important impact on its emission rate. 144 Similar results were also obtained for BFRs (Rauert et al., 2015; Sun et al., 2018). Specifically, 145 significant correlations between the PBDE mass captured on a flexible polyurethane foam (PUF) 146 cylinder (and deemed to have volatilised from curtains) and the vapour pressure (at 60°C, Table S3, 147 Supplementary Material) of PBDE congeners were observed in a cylindrical test chamber (r²=0.84, p=0.003) and a Micro-Chamber ($r^2=0.67$, p=0.024), which implied that the volatilisation emission 148 149 behaviour of PBDEs was strongly influenced by their vapour pressure (Rauert et al., 2015). Sun et 150 al. (2018) also observed higher emission rates of HBCDDs than of some NBFRs such as bis(2,4,6-151 tribromophenoxy) ethane (BTBPE) and bis(2-ethyl hexyl) tetrabromophthalate (BEH-TEBP or 152 TBPH). Moreover, HBCDD had higher sensitivity to the rise of temperature (i.e., greater increase 153 in emission rates per unit increase in temperature) than did NBFRs, which could possibly be 154 attributed to the higher vapour pressure of HBCDD than the studied NBFRs (Table S3, 155 Supplementary Material).

156

157 3.1.2 Impact of temperature on emissions of FRs to air

158 Tokumura et al. (2019) revealed a sharp (65-fold) increase in the emission rate of TDCIPP with 159 increasing temperature, from an average emission rate of 0.17 µg·m⁻²·h⁻¹ at 20 °C to 11 µg·m⁻²·h⁻¹ 160 at 60 °C. Similar results were reported for tris(1-chloro-2-propyl) phosphate (TCIPP) (Poppendieck 161 et al., 2017), NBFRs (Sun et al., 2018), HBCDD (Kajiwara and Takigami, 2013; Sun et al., 2018), 162 and PBDEs (Kajiwara and Takigami, 2013; Kemmlein et al., 2003). For instance, Kajiwara and 163 Takigami (2013) adapted a small cylindrical stainless steel chamber (7 cm diameter \times 5.5 cm height) 164 to conduct emission tests on three BFR-treated textile samples at temperature of 20-80 °C While the 165 emission rates of both HBCDDs and PBDEs remained stable over a temperature range of 20-60 °C, 166 a considerable increase in the emission rates of HBCDDs (~20 fold) and PBDEs (~6 fold) was 167 observed at 80 °C Notably, the relative abundance of α -HBCDD, which has the highest vapour 168 pressure among the three HBCDD diastereomers, continued to increase as the temperature was

- 169 raised, while that of γ -HBCDD (which has the lowest vapour pressure among the three HBCDD
- 170 diastereomers) continued to decline (vapour pressure at 25 °C: 1.05×10^{-8} for α -HBCDD, 5.82×10^{-9}

171 for β-HBCDD, 8.39×10^{-11} for γ-HBCDD). Moreover, the relative abundance of $\sum_{di-to octa-BDEs}$

- 172 compared to $\sum_{\text{nona-to deca-BDEs}}$ was noted to increase with temperature.
- 173

174 3.1.3 Impact on FR emissions to air of timescale of experiments

175 It has been suggested that emission rates will be greatest at the beginning of an experiment when a 176 material is tested in a ventilated chamber (Liang et al., 2015). This hypothesis is supported by Sun 177 et al. (2018), who reported a continuous decline of emission rates of BEH-TEBP (from approximately 6.0 pg·g⁻¹·h⁻¹ to less than 1.0 pg·g⁻¹·h⁻¹) and HBCDD (from approximately 80 pg·g⁻¹ 178 $^{1}\cdot h^{-1}$ to 10 pg·g⁻¹·h⁻¹) from new carpet during a 5-week experiment. Similar results were also 179 180 reported for TCIPP (Ni et al., 2007). A possible explanation for this temporal decline in emission 181 rate is that volatilisation of FRs present in the surface of the test material is more facile when the 182 test material is initially placed into a chamber, leading to an initially high emission rate, whereas 183 long-term emissions of FRs are likely limited by the inefficient diffusion of FRs from the inside of 184 the test material to the surface (Poppendieck et al., 2017). Another study conducted by Kemmlein 185 et al. (2003) evaluated emission behaviour of a wide range of FRs (TCIPP, HBCDD, PBDEs, etc.) 186 from four product groups (insulating materials, assembly foam, upholstery/mattresses, and 187 electronics/electrical equipment). Although some FRs (i.e., HBCDDs and deca-BDE) were barely 188 detected throughout the tests (therefore their emission rates were not discussed), emissions of other 189 FRs (i.e., TCIPP and tri- to penta-BDEs) increased considerably at relatively early stages of the tests, 190 and it is notable that equilibrium appeared to be attained for these FRs after a test period of 60-160 191 days. These findings of lower emission rates in the latter stages of emission chamber experiments 192 might provide a better reflection of "real world" emissions from formal waste treatment activities, 193 as the wastes accepted by such facilities are in most cases "old" and used products that are likely to 194 have attained product-air equilibrium. Therefore, it seems reasonable that PBDE concentrations in 195 indoor air in e-waste/plastic storage areas (1.3-230 ng·m³) were much lower than those in e-waste 196 dismantling areas (163-2,900 ng·m⁻³) in 3 formal e-waste recycling plants in China (Die et al., 2019), as e-waste dismantling activities could disturb such product-air equilibrium and release FRs into theair from inside the e-waste items.

199

200 3.1.4 Impact of concentrations of FRs in products or materials on emissions to air

201 Ni et al. (2007) observed a significant positive linear relationship ($r^2=0.935$, p<0.001) between the 202 SER of TCIPP and TCIPP content in PVC wallpaper measured at room temperature (22-28 °C). 203 Similar results were also reported by Sun et al. (2018), who observed a significant positive linear 204 relationship (r²=0.65, p=0.003) between the SERs of BFRs (hexabromobenzene (HBBz), TBBP-A, 205 HBCDD, and BEH-TEBP) and their concentrations in 6 types of materials (new carpet, computer 206 casing, sound insulation cotton, circuit board, decorative laminate, and PVC flooring). These results 207 indicate OPE and BFR emissions from commercial products are likely influenced strongly by their 208 concentrations in the materials. In addition, due to the lack of significant correlation between the 209 physicochemical properties of BFRs and their SERs, Sun et al. (2018) concluded that the 210 concentration of BFRs in a product might have a more profound effect on BFR emission behaviour 211 than either vapour pressure or octanol-air partition coefficient (K_{OA}). Interestingly, while Tokumura 212 et al. (2019) also reported higher emission rates of TDCIPP related to elevated TDCIPP 213 concentrations in curtains, the relationship might not be linear, as a 1.1-fold increase in TDCIPP 214 concentrations (from 3,900 μ g·g⁻¹ to 4,310 μ g·g⁻¹) in curtains resulted in a 4-fold increase in SERs 215 of TDCIPP (from 0.044 μ g·m⁻²·h⁻¹ to 0.17 μ g·m⁻²·h⁻¹) at a constant temperature of 20 °C. The cause(s) 216 of such disparity remain(s) unclear, and could be of potential interest in further studies.

217

218 3.1.5 Other factors influencing emissions of FRs to air from products

219 Other factors influencing HFR and OPE emission behaviour include: the rate of air flow across the 220 surface of a product, the surface area-to-volume ratio of a product, and the mode of FR incorporation 221 into the product (i.e., additive or reactive). Poppendieck et al. (2017) identified a significant increase 222 in the SER of TCIPP from PVC wallpapers with increased air flow rate in the test chamber, with the 223 SER of TCIPP almost doubling at an average flow rate of 50 mL·min⁻¹ to 200 mL·min⁻¹. In addition, 224 the strength of FR binding to products or materials might also influence the emission behaviour of 225 FRs. For instance, using a filter paper spiked with PBDE standards, Rauert et al. (2015) identified 226 detectable PBDE volatilisation in chamber experiments. However, this was not replicated when 227 PBDE-containing TV casing was studied in the same chamber configuration, suggesting minimal 228 volatilisation of PBDEs from the TV casing, even though the PBDE concentration in the TV casing 229 was almost 10 times higher than in the filter paper (Rauert and Harrad, 2015). A likely explanation 230 is that the PBDEs were bound strongly to the TV casing during the production process (melting and 231 remoulding), with the greater surface area-to-volume ratio of the filter paper another contributory 232 factor (Rauert and Harrad, 2015). Finally, the mode via which FRs are incorporated into a polymer 233 likely influences FR emission behaviour. Specifically, emission rates of FRs that are covalently 234 (reactively) bound to materials are expected to be much lower than if the FRs are incorporated by 235 physical mixing (additively) (Wolf et al., 2000). However, empirical evidence of this (especially 236 from chamber tests) remains scarce, likely due to difficulties with experimental design.

237

238 3.2 Emissions of FRs via abrasion of products

239 Abrasion of fine particles or fibres from waste materials could possibly be a contributing factor to 240 FR migration to soil. A study conducted in an e-waste dismantling site in Guiyu, China identified 241 soil microplastic pollution in 33 soil samples representing various activity zones (3 derelict e-waste 242 disassembling sites, 4 polluted farmlands, 2 fruit growing areas without dismantling activities, 1 243 dumpsite, and 1 control site under an expressway) (Chai et al., 2020). Compared to the control site 244 where microplastic was not detected and the fruit growing areas where soil contamination with 245 microplastic particles was low (mean: 36.7±24.3 particles kg⁻¹), the average density of microplastic 246 in the derelict e-waste disassembling sites, polluted farmlands, and dumpsite was 13,900±7,260 247 particles kg⁻¹, 12,300±10,500 particles kg⁻¹, and 3,570±688 particles kg⁻¹, respectively. These 248 results suggest abrasion of microplastics from waste products to soil is considerable in the vicinity 249 of e-waste dismantling sites and dumpsites. While FR concentrations were not reported in the 250 microplastics or in the soil samples, insights into the potential contribution of this "abrasion effect" 251 to FR concentrations in the surrounding environment, may be gleaned from emission chamber tests 252 where dust rather than soil particles was used in these tests (Rauert and Harrad, 2015; Rauert et al., 253 2014). Specifically, a magnetic stirrer bar was introduced into an in-house designed test chamber 254 (10 cm diameter \times 20 cm height) to mimic abrasion of fibres from a piece of HBCDD-treated curtain 255 to dust (Rauert et al., 2014). HBCDD concentrations in the dust samples increased sharply from an initial level of 110 $\text{ng}\cdot\text{g}^{-1}$ to 52,500 $\text{ng}\cdot\text{g}^{-1}$ after 48 hours of abrasion, and significant positive linear 256 257 relationships were observed between abrasion time and HBCDD concentrations in dust. Rauert et 258 al. (2014) also identified 2-10 bromine rich polymeric fragments per mg UK dust which were 259 identified as responsible for the elevated BFR concentrations in the dust samples (HBCDD: 490-260 88,600 ng·g⁻¹, BDE-209: 24,000-1,440,000 ng·g⁻¹). Similarly, a clear increase in concentrations of 261 PBDEs (BDE-153, -154, -183, and -209) in post-experiment dust was observed when a magnetic 262 stirrer bar was used to mimic the process of abrasion of a plastic TV casing (Rauert and Harrad, 263 2015). Moreover, the presence of similar PBDE congener profiles in the TV casing and in postexperiment dust, which differed from congener profiles in pre-experiment dust, further confirmed 264 265 the likely role of abrasion of fine particles from plastic TV casing as a source of PBDEs to dust 266 (Rauert and Harrad, 2015).

267

268 Hence, although dust rather than soil was used in the chamber tests of Rauert and Harrad, it is 269 reasonable to hypothesise a considerable contribution to FR concentrations in soil arising from 270 abrasion of fine particles or fibres from FR-containing products. This is likely to happen during 271 outdoor transport, storage, and landfilling of waste materials. Additionally, emissions of dust and 272 particles from indoors to outdoor environment could possibly be a contributing factor. Rauert and 273 Harrad highlighted the difficulties of extrapolating the results of their abrasion experiments to "real 274 world" scenarios, whereby a few hours simulated abrasion in their chamber might represent several 275 years abrasion in the real world (Rauert and Harrad, 2015; Rauert et al., 2014). However, abrasion 276 of plastics is likely to occur much less frequently in homes than in waste treatment plants and 277 landfills when waste products are transported, stored, or landfilled, thereby accounting for elevated 278 FR concentrations in soil in the vicinity of waste treatment facilities.

279

280 3.3 Leaching of FRs to surrounding environment

281 Leaching of FRs occurs in landfills, dumpsites, and waste recycling and dismantling facilities, etc.
282 when FR-treated products come in contact with aqueous fluids. For instance, the presence of HFRs

and OPEs in landfill leachates has been reported in several previous studies, with concentrations of

BFRs and OPEs in leachate reaching up to 133 μ g·L⁻¹ and 437 μ g·L⁻¹, respectively (Kwan et al., 2013; Qi et al., 2019). In order to understand the leaching behavior of HFRs and OPEs from products or wastes under complex conditions, controlled leaching experiments have been designed to examine potential factors influencing their leaching behaviour. The most frequently examined factors are: dissolved humic matter (DHM) content, leachate temperature, leachate pH, and contact time (Aminot et al., 2020; Choi et al., 2009; Harrad et al., 2020a; Stubbings and Harrad, 2018, 2019). The following sections discuss these in turn.

291

292 3.3.1 Impact of leachate DHM content on FR leaching

A leaching test conducted by Choi et al. (2009) examined leaching of BFRs (including PBDEs, 293 294 TBBP-A, and NBFRs) from TV housing plastics at 20 °C. While most lower brominated PBDE 295 congeners and TBBP-A were not detected in distilled water, most chemicals could be detected in 296 DHM solution (1,000 mg·L⁻¹), and leaching of higher brominated PBDEs (octa-, nona-, and deca-297 BDEs) was over 10 times higher than in distilled water. Similar leaching behaviour was observed 298 for NBFRs, indicating DHM would play an important role in enhancing solubility of brominated 299 compounds. Another study conducted by Harrad et al. (2020a) reported significantly greater (p<0.05) 300 leaching of BDE-209 and HBCDD at a leachate DHM concentration of 1,000 mg·L⁻¹ than that 301 observed at DHM concentrations of 0 and 100 mg·L⁻¹. Specifically, under simulated landfill 302 conditions (pH 6.5, 20 °C, no agitation, and waste-leachate ratio 0.05), an average of 0.33% of BDE-303 209 and 0.69% of HBCDD were leached from fabrics at a DHM concentration of 1,000 mg L^{-1} , 304 while 0.18-0.23% of BDE-209 and 0.29-0.45% of HBCDD were leached at DHM concentrations 305 of 0-100 mg L^{-1} , respectively. Such enhancement of BFR solubility in leachate by DHM has been 306 reported in several studies (Danon-Schaffer et al., 2013; Kajiwara et al., 2014; Kim et al., 2006; 307 Osako et al., 2004; Stubbings and Harrad, 2019; Zhou et al., 2013), and could possibly be attributed 308 to the role of the surface-active agent (for instance, linear alkylbenzene sulfonate) and the role of 309 "facilitated transport" in the enhanced leaching behaviour of BFRs (Kim et al., 2006). Briefly, 310 increased DHM concentration helps formation of micelles combining DHM and BFRs, which then 311 allows desorption of BFRs from micelles, leading to enhanced leachability of BFRs. In the 312 meantime, DHM might also act as mobile carriers of contaminants and thus increase the mobility

313 of BFRs in leachate.

314

315 3.3.2 Impact on FR leaching of leachate temperature

316 Temperature is also an important factor when simulating leaching processes of HFRs or OPEs, as 317 leachate temperatures can reach 80-90 °C at the initial aerobic stage within a landfill (Stubbings and 318 Harrad, 2014). For instance, Stubbings et al. (2016) observed considerably increased leaching of 319 HBCDD from treated curtains with increasing temperature. An increase in temperature from 20 °C 320 to 80 °C increased concentrations of α -HBCDD in leachate by 4.3-4.8 times, while concentrations 321 of γ -HBCDD in leachate increased by 28-33 times. HBCDD leaching from building insulation 322 foams was also enhanced by increasing leaching fluid temperature (Stubbings and Harrad, 2019). 323 Furthermore, it was suggested that α -HBCDD was preferentially leached at lower temperature (e.g., 324 20 °C) compared to γ -HBCDD, while γ -HBCDD had greater sensitivity (i.e., greater increase in 325 leaching efficiency) to the rise of temperature (Aminot et al., 2020; Stubbings et al., 2016). Similar 326 enhancement of PBDE (especially BDE-99 and -209) leaching resulting from increasing 327 temperature has also been reported (Stubbings and Harrad, 2016).

328

329 Interestingly, some contrasting observations were also reported, with higher leachate temperature 330 resulting in decreased HFR leaching rates (Harrad et al., 2020a; Stubbings and Harrad, 2018). 331 Specifically, Stubbings and Harrad (2018) observed a significant increase in TCIPP concentrations 332 in distilled water, and a slight increase (though not significant) in TCIPP concentrations in DHM 333 solutions (100 mg·L⁻¹ and 1,000 mg·L⁻¹), when leachate temperature increased from 20 °C to 50 °C. 334 However, TCIPP concentrations in leachates at 80 °C were considerably lower than those at 50 °C. 335 In the meantime, Harrad et al. (2020a) reported significantly decreased concentrations in leachate 336 of both BDE-209 and HBCDD on increasing leachate temperature from 20 °C to 60 °C and 80 °C. 337 Such observations were attributed to enhanced volatilisation of these contaminants at higher 338 temperature, i.e., these compounds were more volatile at this higher temperature and could 339 potentially enter the headspace of the leaching vessel in the gas phase and be lost when the vessel 340 was subsequently opened before cooling to ambient temperature (Harrad et al., 2020a; Stubbings 341 and Harrad, 2018).

342

343 3.3.3 Impact of leachate pH on leaching of FRs

344 There is conflicting evidence over the impact of leachate pH on leaching behaviour of HFRs and 345 OPEs. Danon-Schaffer et al. (2013) studied the influence of leachate pH (4, 5, 7, and 9) on PBDE 346 leaching from e-waste by contacting e-waste with leachate collected from an urban landfill. Test 347 results indicated initially much higher leachate concentrations of lower brominated PBDE 348 congeners at a pH of 4, while the highest concentrations of Σ PBDEs in leachate were observed at a 349 pH of 5. Correlations between concentrations of individual PBDEs and pH were observed, 350 indicating a strong influence of pH. Harrad et al. (2020a) reported similar results for HBCDD, noting 351 that leaching of HBCDD from fabrics was significantly greater (p<0.05) at pH 5.8 than at either pH 352 6.5 or 8.5, but interestingly, leaching of BDE-209 was not significantly influenced by pH(p>0.05), 353 despite the slightly greater BDE-209 leaching at more acidic pH values. Such disparities were 354 further emphasised by Stubbings and Harrad (2016, 2018), who reported lower PBDE and TCIPP 355 concentrations in leachates at more acidic pH values, while studies on leaching of HBCDD from 356 building insulation foams revealed only minor effects of pH on HBCDD concentrations in leachates 357 (Stubbings and Harrad, 2019). It is notable that Harrad et al. (2020a) proposed a possible explanation 358 for such disparities, i.e., agitation introduced in experiments might lead to abrasion of small particles 359 or fibres from FR-containing materials to leachates, which might have masked any impact of pH in 360 their experiments. Therefore, such uncertainties should be eliminated in follow-up studies.

361

362 3.3.4 Impact on leaching of FRs of waste-leachate contact time

363 Choi et al. (2009) investigated the leaching characteristics of BFRs (PBDEs, TBBP-A, NBFRs etc.) 364 from TV housing plastics to distilled water and DHM solutions. Although concentrations of most 365 BFRs in DHM solutions $(1,000 \text{ mg} \cdot \text{L}^{-1})$ were over 10 times higher than those in distilled water, 366 leaching of BFRs showed similar time-dependent trends between the two different leachates, i.e., 367 high leaching rates at the initial stage followed by sharply reduced leaching rates at longer contact 368 times. Such results were consistent with those of Danon-Schaffer et al. (2013), who reported a 369 substantial increase in PBDE concentrations in leachates during the first 24 h and minimal increase 370 in the concentrations afterwards (for all 5-year intervals) when contacting e-waste with distilled 371 water and municipal landfill leachates. Furthermore, Zhou et al. (2013), together with another three 372 studies (Harrad et al., 2020a; Stubbings and Harrad, 2016; Stubbings et al., 2016), also observed 373 initially sharply increased concentrations of BFRs (PBDEs, TBBP-A, and HBCDD) in leachates 374 followed by steady or lower leachate concentrations after a certain contact period (typically a few 375 hours). It was suggested that leaching of BFRs was likely to be predominantly governed by second-376 order leaching kinetics, whereby a period of initially intense dissolution of more labile BFRs on the 377 surface of products was followed by a slower stage corresponding to external diffusion of the soluble 378 residue within products (Harrad et al., 2020a; Stubbings and Harrad, 2016; Stubbings et al., 2016).

379

380 In contrast to BFRs, leaching of TCIPP from furniture PUF appears a first-order process (Stubbings 381 and Harrad, 2018). This disparity with the leaching kinetics of FRs, may be attributed to the 382 significant differences in the physicochemical properties of OPEs and BFRs, with the aqueous 383 solubility of TCIPP being 4 to 7 orders of magnitude greater than that of PBDEs (Table S3, 384 Supplementary Material). However, the properties of furniture PUF (e.g., relatively porous and 385 permeable) could also be an important factor. As current data on the leaching kinetics of OPEs are 386 severely limited, follow-up studies are strongly encouraged to elucidate the leaching characteristics 387 of OPEs and to explore whether leaching of BFRs from PUF is more facile than from hard plastic 388 materials such as EEE casing.

389

390 3.3.5 Other factors influencing leaching of FRs from products

391 Other factors influencing leaching characteristics of HFRs and OPEs may include: waste:leachate 392 ratio, agitation, initial concentration of HFRs and OPEs in treated products, and availability of 393 oxygen during leaching, etc. For example, Harrad et al. (2020a) found that leaching of both BDE-394 209 and HBCDD from fabrics was significantly greater (p < 0.05) at a waste-leachate ratio of 0.005 395 $g \cdot mL^{-1}$ than at 0.05 $g \cdot mL^{-1}$, which was likely due to the lower fabric surface area to leaching fluid 396 volume ratio at the higher waste-leachate ratio. Waste agitation was considered as a significant 397 factor enhancing leaching of both HFRs and OPEs from products (e.g., plastics, building insulation 398 foams, curtains, and furniture PUF), likely due to abrasion of fine particles or fibres from products 399 to leachates during agitation (Harrad et al., 2020a; Stubbings and Harrad, 2016, 2018, 2019;

400 Stubbings et al., 2016). Kajiwara et al. (2014) conducted a long-term landfill lysimeter experiment 401 under three different simulated landfill conditions (aerobic, semi-aerobic, and anaerobic), and found 402 that leaching of BFRs under anaerobic conditions (which resembled the conditions of an open 403 dumping site) tended to exceed that under aerobic conditions. In addition, when using bottom ash 404 from municipal solid waste incinerators as the matrix, Lin et al. (2014) indicated that higher PBDE 405 concentrations in the bottom ash led to higher PBDE leachate concentrations. This is of particular 406 concern when indoor dust is collected from workshop floors in e-waste recycling facilities and sent 407 for disposal in landfills, as this dust is generally heavily contaminated with HFRs and OPEs, and 408 could readily contaminate landfill leachate and the surrounding environment (Stubbings et al., 2019).

409

410 4. Occurrence of HFRs and OPEs in soil impacted by formal waste treatment facilities

411 4.1 Concentrations of FRs in soil in the vicinity of formal e-waste dismantling and recycling 412 facilities

413 Previous studies have reported high levels of HFR and OPE contamination in soil near e-waste 414 dismantling and recycling facilities (Table 2 and Table 3), with their concentrations occasionally exceeding 100,000 ng·g⁻¹ dry weight (dw) (Ceballos and Dong, 2016; Ge et al., 2020; McGrath et 415 416 al., 2017a, 2018). McGrath et al. (2018) reported concentrations of 8 PBDE congeners (BDE-28, -417 47, -99, -100, -153, -154, -183, and -209) and 6 NBFRs (pentabromotoluene (PBT), 418 pentabromoethylbenzene (PBEB), HBBz, 2-ethyl hexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), 419 BTBPE, and decabromodiphenyl ethane (DBDPE)) in 36 soils collected from two Australian e-420 waste recycling plants and 8 reference soils. Concentrations in soil of \sum_{8} PBDEs were in the range $34-5,000 \text{ ng} \cdot \text{g}^{-1} \text{ dw}$ (median: 130 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$) and 8.3-98,000 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$ (median: 160 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$) in 421 422 the two e-waste recycling plants, respectively. Such elevated PBDE concentrations exceeded 423 considerably those in the reference soils (range: 0.10-44 $ng \cdot g^{-1}$ dw, median: 21 $ng \cdot g^{-1}$ dw), and 424 concentrations of \sum_{6} NBFRs in soils from the two e-waste recycling plants (median: 3.8 and 15 ng·g⁻ 425 ¹ dw, respectively) were also significantly higher than those in reference soils (median: $< 0.02 \text{ ng} \cdot \text{g}^{-1}$ 426 1 dw). In addition, Ge et al. (2020) observed much greater soil contamination with FRs in an 427 industrial park in Guiyu, China, than in surrounding areas. Median concentrations of \sum_{20} PBDEs 428 (tri- to deca-BDE), Σ_2 NBFRs (pentabromobenzene (PBBz) and HBBz), Σ_2 DPs (dechlorane plus; 429 syn-DP and anti-DP), and \sum_{13} OPEs (tripropyl phosphate (TPP), tributyl phosphate (TBP), tris(2-430 chloroethyl) phosphate (TCEP), TCIPP, tris(2-chloropropyl) phosphate (T(2-C)PP), TDCIPP, 431 triphenyl phosphate (TPHP), 2-ethylhexyl diphenyl phosphate (EHDPP), tris(2-ethylhexyl) 432 phosphate (TEHP), tris(2-isopropylphenyl) phosphate (TIPPP), and 3 isomers of TCP) were 46,300 ng·g⁻¹ dw, 294 ng·g⁻¹ dw, 712 ng·g⁻¹ dw, 12,000 ng·g⁻¹ dw, respectively, in soils collected from the 433 434 industrial park. These observations were 1 to 2 orders of magnitude higher than the corresponding 435 FR concentrations in soils from surrounding areas. Similarly, higher HFR and OPE concentrations 436 have been reported in soils from e-waste dismantling and recycling areas than in surrounding areas 437 (e.g., urban and rural areas, farmlands, and background areas) in China (e.g., Hongkong, Tianjin, 438 Shanghai, etc.), South Korea, and Vietnam (Li et al., 2014, 2016, 2017; Man et al., 2011; Wang et 439 al., 2018b). Together, these observations indicate formal e-waste recycling facilities have great 440 potential to contaminate surrounding soils, despite use of environmentally friendly recycling 441 technologies.

442

443 Pertinently, McGrath et al. (2018) reported significant negative correlations (p < 0.05) between 444 PBDE and NBFR concentrations in soils and distance from the two e-waste facilities, and the 445 significant difference between BFR concentrations in soils collected from 300-900 m away from an 446 e-waste site and BFR concentrations in reference soils further illustrated the potential of regulated 447 e-waste recycling to significantly elevate BFR concentrations in soils located up to 900 m from such 448 activity. These observations were consistent with another two studies where significant negative 449 correlations were reported between concentrations of PBDEs, NBFRs, and DPs in soil and the 450 distance between sampling locations and an industrial park (p < 0.05) (Ge et al., 2020) and an e-waste 451 recycling area (p value not provided) (Hong et al., 2018). This is a clear indication that these e-452 waste recycling facilities acted as an emission point source, and the authors further concluded that 453 distribution of BFRs and DPs in soil from e-waste recycling facilities matches the point source 454 pollution pattern (Ge et al., 2020; Hong et al., 2018; McGrath et al., 2018). It is also interesting to 455 note that significant correlations between OPE concentrations in soil and distance from the e-waste 456 dismantling park were not found, which possibly means the e-waste dismantling park is not a 457 dominant source of OPEs to surrounding soils compared to diffuse OPE emissions from their in-use

458 consumer goods as both FRs and plasticisers (Ge et al., 2020). Thus, further investigation is
459 recommended to better understand the impact of formal e-waste dismantling and recycling facilities
460 on local OPE contamination.

461

462 4.2 Concentrations of FRs in soil near official waste dumpsites and landfills

463 Huang et al. (2013) reported elevated PBDE concentrations (sum of BDE-28, -47, -99, -100, -153, -154, -183, and -209; range: 69-1,122 ng g^{-1} dw; median: 234 ng g^{-1} dw) in topsoil collected from 464 465 a municipal landfill in Shanghai, China. These elevated PBDE concentrations were 1 to 3 orders of 466 magnitude higher than those reported in surface soils from multiple functional areas (e.g., 467 agricultural areas, industrial areas, commercial areas, residential areas, parks and greenbelts, 468 automobile manufacture areas, etc.) in Shanghai, China (Jiang et al., 2010, 2012; Wu et al., 2015), 469 indicating considerable PBDE emissions during the landfill disposal process. Comparably elevated 470 PBDE concentrations in soils from landfills were reported in northern Canada, South Africa, and 471 Brazil (Akortia et al., 2019; Cristale et al., 2019; Danon-Schaffer et al., 2008). For example, in 472 northern Canada, the average concentration of \sum_{60} PBDEs was 131 ng g⁻¹ dw in surface soils 473 impacted by waste dumpsites and landfills, while in background areas an average PBDE 474 concentration of 1.94 ng·g⁻¹ dw was identified (Danon-Schaffer et al., 2008). Furthermore, although 475 concentrations of PBDEs (sum of BDE-28, -47, -99, -100, -153, -154, -183, -197, -206, -207, -208, 476 and -209; range: $0.13-1.2 \text{ ng} \cdot \text{g}^{-1} \text{ dw}$) in surface soils were much lower from a municipal landfill site 477 in Tibet, China than elsewhere, the PBDE concentrations were still 3 orders of magnitude higher 478 than the background values measured in soils from the Tibetan Plateau, and elevated PBDE 479 concentrations were observed in soils collected from 9.2 km away from the landfill (Li et al., 2018b). 480 Based on principal components analysis and multiple linear regression, the authors concluded that 481 the higher PBDE concentrations in soils from the landfill could be explained by atmospheric 482 dispersion (accounting for 61% of the total concentrations) and leachate seepage (accounting for 483 39% of the total concentrations) (Li et al., 2018b). These results provide further evidence of PBDE 484 emissions arising from municipal landfill disposal activities.

485

486 Interestingly, Harrad et al. (2020b) reported PBDE, DBDPE, and HBCDD concentrations in

487 landfill-related soils from Ireland and came to the opposite conclusion. Concentrations in soil of 488 Σ_{7} PBDEs (BDE-28, -47, -99, -100, -153, -154, and -183), BDE-209, DBDPE, and Σ_{3} HBCDDs (α -, 489 β -, γ -HBCDD) were: 0.043-7.6 ng·g⁻¹ dw, 0.065-63 ng·g⁻¹ dw, ND-0.54 ng·g⁻¹ dw, and ND-6.2 ng·g⁻¹ ¹ dw, with median concentrations of 0.14 $ng \cdot g^{-1}$ dw, 0.57 $ng \cdot g^{-1}$ dw, ND, and 0.55 $ng \cdot g^{-1}$ dw, 490 491 respectively. These were comparable to PBDE concentrations in European background soils $(\sum_{20} PBDEs; range: 0.065-12 \text{ ng} \cdot \text{g}^{-1} \text{ dw}; \text{ median: } 0.61-2.5 \text{ ng} \cdot \text{g}^{-1} \text{ dw})$ (Hassanin et al., 2004). 492 493 Moreover, applying a non-parametric Wilcoxon signed rank test, the authors found no significant 494 difference in BFR concentrations in soils between downwind and upwind locations (p>0.1), 495 indicating no discernible impact of the landfills on concentrations of BFRs in surrounding soils. 496 Reasons for this disparity compared to other studies are unclear, while proper management of 497 landfills, applications of impervious polymeric liners, and sound classification of waste in Ireland 498 could possibly be contributing factors.

499

500 In addition to PBDEs, data on soil concentrations of other FRs are scarce (Table 3). Concentrations 501 of \sum_{10} OPEs (TCEP, TCIPP, TDCIPP, tri-isobutyl phosphate (TIBP), tri(n-butyl)phosphate (TNBP), 502 tris(2-butoxyethyl) phosphate (TBOEP), TEHP, TPHP, EHDPP, and tris(methylphenyl) phosphate 503 (TMPP)) and Σ_4 NBFRs (DBDPE, BTBPE, EH-TBB, and BEH-TEBP) were in the range of 1.8-504 186 ng·g⁻¹ dw and 1.1-83 ng·g⁻¹ dw in soils from a waste landfill in Brazil, with mean values of 67 $ng \cdot g^{-1}$ dw and 19 $ng \cdot g^{-1}$ dw, respectively (Cristale et al., 2019). The highest soil concentrations of 505 506 OPEs and NBFRs were observed in areas where e-waste and furniture PUF were disposed of, while 507 lower concentrations were observed in soils collected from more distant locations. Concentrations 508 of Σ_2 DPs (syn- and anti-DP) were in the range of ND-3.97 ng·g⁻¹ dw in soils collected from an 509 official municipal waste dumpling site in Pakistan (Hafeez et al., 2016). The mean concentration of 510 Σ_2 DPs was highest in soils collected from dumpsites (0.48 ng \cdot g⁻¹ dw), followed by agricultural zone 511 (0.33 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$), roadside (0.05 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$), and residential zone (0.04 $\text{ng} \cdot \text{g}^{-1} \text{ dw}$), respectively. Li 512 et al. (2021) reported elevated concentrations of short-chain chlorinated paraffins (SCCPs) in soils 513 (56.8-1,348 ng·g⁻¹ dw) from an urban landfill and rural dumpsites in Tibet, China. Within a 5 km 514 distance from the landfill, SCCP concentrations in soils declined rapidly with increasing distances 515 from the landfill, while SCCP concentrations relatively leveled off outside the 5 km distance. The

results suggest the potential of landfills to significantly elevate SCCP concentrations in soils located
up to 5 km from such activity, and this could be attributed to the atmospheric dispersion of SCCPs
(Li et al., 2021). Further studies are encouraged to facilitate better understanding of the potential of
landfills to elevate FR concentrations in surrounding soils.

520

521 4.3 FR concentrations in soil near waste incineration plants

522 Contamination with HFRs and OPEs of soil surrounding waste incinerators has rarely been 523 investigated previously (Table 2 and Table 3). Zhang et al. (2013) determined PBDE concentrations 524 in 14 soil samples collected from the vicinity of a solid waste incineration plant in Beijing, China. Concentrations of \sum_{42} PBDEs (mono- to deca-BDEs) were in the range 0.29-120 ng g⁻¹ dw, with a 525 median concentration of 1.4 ng·g⁻¹ dw. A declining trend of concentrations of \sum_{42} PBDEs was 526 527 identified as the distance from the investigated incinerator increased, potentially indicating that the 528 solid waste incineration plant was a potential pollution source of PBDEs. However, it is noteworthy 529 that PBDE concentrations in soils from the incineration facility were not significantly different (p=0.098) to those of \sum_{40} PBDEs in agricultural soils (range: 0.50-3.3 ng·g⁻¹ dw; median: 1.3 ng·g⁻¹ 530 531 dw) in Beijing, China (Sun et al., 2009). Although the lack of statistical significance could be 532 attributed to the small sample size in the two studies, this might also indicate a relatively small 533 influence of the solid waste incineration plant on PBDE contamination of surrounding soil. Another 534 study reported PBDE (BDE-28, -47, -99, -100, -153, -154 -183 and -209) concentrations in urban 535 soils in Melbourne, Australia (McGrath et al., 2016). Two soil samples were collected from a waste 536 incinerator facility, and concentrations of \sum_{8} PBDEs were 13.6 ng·g⁻¹ dw and 80.8 ng·g⁻¹ dw, 537 respectively. These concentrations were much lower than those in soils collected from an e-waste recycling area (1,080 ng \cdot g⁻¹ dw and 13,200 ng \cdot g⁻¹ dw; n=2) and a domestic dumpsite (24.6 ng \cdot g⁻¹ 538 539 dw and 776 ng·g⁻¹ dw; n=2), and only slightly exceeded those detected in soils from residential 540 areas, parkland, and urban background locations (range: 0.12-43.8 ng·g⁻¹ dw; n=6). Furthermore, 541 out of the 6 targeted NBFRs (PBT, PBEB, HBBz, EH-TBB, DBDPE, and BTBPE), only HBBz was 542 quantifiable in one of the two soil samples $(0.34 \text{ ng} \cdot \text{g}^{-1} \text{ dw})$ (McGrath et al., 2017b). These results 543 suggest a minimal impact of the studied waste incinerator on BFR emissions to soil. In the meantime, 544 it is important to acknowledge that our current understanding of the contribution of incinerators to

FR (especially alternative FRs such as NBFRs and OPEs which are very important in the current
FR market) concentrations in soil is very limited due to the lack of data, thus further studies are
encouraged to fill the research gap.

548

549 5. Occurrence of HFRs and OPEs in outdoor air impacted by formal waste treatment550 facilities

551 5.1 Concentrations of FRs in air near formal e-waste dismantling and recycling facilities

552 Hong et al. (2018) reported concentrations of a broad range of HFRs in the atmosphere in Ziya Circular Economy Area (Tianjin, China), including 13 PBDEs (BDE-17, -28, -47, -49, -66, -85, -553 554 99, -100, -138, -153, -154, 183, and -209), 17 NBFRs (BTBPE, DBDPE, allyl 2,4,6-tribromophenyl 555 ether (ATE), 2,3,5,6-tetrabromo-p-xylene (p-TBX), α - and β -tetrabromoethylcyclohexane 556 (TBECH), PBBz, 2-bromoallyl 2,4,6-tribromophenyl ether (BATE), PBT, α- and β-1,2,5,6-557 tetrabromocyclooctane (TBCO), PBEB, HBBz, 2,3-dibromopropyl 2,4,6-tribromophenyl ether 558 (DPTE), pentabromobenzyl acrylate (PBBA), hexachlorocyclopentenyl-dibromocyclooctane 559 (HCDBCO), and octabromotrimethylphenylindane (OBTMPI)), and 2 DP isomers (syn- and anti-560 DP). All the targeted HFRs were detected except for OBTMPI, with concentrations of Σ_{13} PBDEs, 561 \sum_{16} NBFRs (OBTMPI excluded), and \sum_{8} DPs being 10,600 pg·m⁻³, 1,330 pg·m⁻³, and 109 pg·m⁻³, 562 respectively. These concentrations detected in the Ziya Circular Economy Area were comparable to 563 those detected in outdoor air in informal e-waste sites in South China (PBDEs + NBFRs: 120-19,000 564 $pg \cdot m^{-3}$, DPs: 13-1,800 $pg \cdot m^{-3}$), but exceeded substantially those at reference sites (PBDEs + NBFRs: 565 55-1,700 pg·m⁻³, DPs: 0.47-36 pg·m⁻³) (Chen et al., 2011; Tian et al., 2011). Cahill et al. (2007) 566 reported a similar impact of formal e-waste recycling activities on elevated PBDE concentrations in 567 outdoor air in California, US. Specifically, concentrations of \sum_{30} PBDE congeners (tri- to deca-BDEs; 568 mean: 93 pg·m⁻³) in the ambient atmosphere were 1 to 2 orders of magnitude lower than those 569 recorded outside an e-waste recycling facility (mean: 340-8,600 pg·m⁻³) and an automotive 570 shredding and metal recycling facility (mean: 390-810 pg·m⁻³). Furthermore, in air sampled on the 571 e-waste site, samples collected near the waste loading dock had mean PBDE levels that were an 572 order of magnitude higher than those in outdoor air collected from the opposite side of the building 573 and downwind of the facility. Moreover, atmospheric PBDE concentrations doubled when normal

574 operation activities were performed in the automotive shredding and metal recycling facility 575 compared to when no activity was performed. Elevated atmospheric concentrations of PBDEs were 576 also observed in formal e-waste recycling facilities in Norway (Morin et al., 2017) as well as at 577 official e-waste storage facilities in Thailand (Muenhor et al., 2010). These results imply significant 578 impacts of formal e-waste activities on HFR contamination in outdoor air.

579

580 Similar results were also observed for OPEs in a gridded field study conducted in Tianjin, China. 581 Liang et al. (2020) found higher concentrations of TDCIPP, TCIPP, EHDPP, TEHP, and \sum_{3} TCPs (o-582 TCP, *m*-TCP, and *p*-TCP) in an e-waste dismantling area than at other sampling sites (urban, 583 suburban, and rural areas), but the specific concentrations were not presented. Furthermore, Wang 584 et al. (2018a) reported high concentrations of 12 OPEs (triethyl phosphate (TEP), TNBP, TBOEP, 585 TEHP, TCEP, TCIPP, TDCIPP, EHDPP, TPHP, TPP, TIPPP, and TMPP) in fine particulate matter 586 (PM_{2.5}) at four e-waste recycling sites in a rural region in Guangzhou, China. OPE concentrations ranged from 780 pg·m⁻³ to 14,000 pg·m⁻³ with a median concentration of 3,300 pg·m⁻³ in the four 587 588 e-waste recycling facilities. These concentrations were 1 to 2 orders of magnitude higher than those 589 in airborne particles over the South China Sea (47-160 $pg \cdot m^{-3}$) (Lai et al., 2015), which possibly 590 indicates that e-waste recycling facilities could be a potential source of OPEs to the surrounding 591 atmosphere. However, it is notable that OPE concentrations in PM2.5 at the e-waste recycling sites 592 were comparable to those at 20 industrial sites in an urban region (range: $520-63,000 \text{ pg} \cdot \text{m}^{-3}$; median: 593 2,800 pg·m⁻³) (Wang et al., 2018a), which implies the e-waste recycling facilities are not a dominant 594 source of OPEs to the surrounding atmosphere compared to OPE emissions from certain industries. 595 The authors suggested high OPE concentrations in $PM_{2,5}$ were associated with: electrical and 596 electronics manufacturing, plastics manufacture, waste recycling, and certain other chemical 597 industries, while lower concentrations were related to: machinery, paper, clothing, and furnishing 598 industries (Wang et al., 2018a). Therefore, despite the limited data reported hitherto, these studies 599 provide evidence of a non-negligible (although not dominant) impact of e-waste recycling on OPE 600 emissions to ambient air.

601

602 5.2 Concentrations of FRs in air near official waste dumpsites and landfills

603 A study conducted in Ireland reported atmospheric concentrations of 8 PBDEs (BDE-28, -47, -99, 604 -100, -153, -154, -183, and -209), 3 HBCDDs (α -, β -, and γ -HBCDD), and DBDPE in the vicinity 605 of 10 municipal solid waste landfills (Harrad et al., 2020b). BDE-47 and -209 were the only PBDE 606 congeners detected, with median concentrations of 0.23 $pg \cdot m^{-3}$ and 4.3 $pg \cdot m^{-3}$, respectively. 607 Σ_3 HBCDDs were not detected in most samples, and the highest concentration was 6.1 pg m⁻³, while DBDPE was only detected in one of the 20 air samples (2.0 pg·m⁻³). Similar results were also 608 609 reported by Weinberg et al. (2011), Morin et al. (2017), and St-Amand et al. (2008), who observed 610 comparable PBDE concentrations (generally lower than 20 pg·m⁻³) in outdoor air collected from 611 landfills in Germany, Norway, and Canada, respectively. The atmospheric BFR concentrations observed in the four studies were generally at the same levels with those observed in urban and rural 612 613 areas across Europe (Jaward et al., 2004a, 2004b; Law et al., 2008), indicating minimal impact of 614 landfills on HFR concentrations in outdoor air.

615

616 Compared to those observations in European countries, HFR concentrations in ambient air in 617 landfills were higher in Pakistan and South Africa (Hafeez et al., 2016; Katima et al., 2018). 618 Concentrations of Σ_8 PBDEs (BDE-28, -35, -47, -99, -100, -153, -154, and -183) and Σ_2 DPs (syn-619 and anti-DP) ranged 53.8-454 pg·m⁻³ and 0.02-1.56 pg·m⁻³, respectively, in 6 ambient air samples 620 collected from three major zones (main dumping site and Lahore compost zone, adjacent 621 agricultural zone, and residential zone) in the vicinity of Mahmood Booti, the only official municipal 622 waste dumping site in Lahore, Pakistan (Hafeez et al., 2016). Interestingly, mean concentrations of 623 \sum_{8} PBDEs (212 pg·m⁻³) and \sum_{2} DPs (0.58 pg·m⁻³) were higher in the main dumpsite than in other 624 zones (Σ_{8} PBDEs mean: 79.4-175 pg·m⁻³; and Σ_{2} DPs mean: 0.41-0.55 pg·m⁻³). Although the 625 significance of this difference was not available due to the limited number of samples, the relatively 626 higher HFR concentrations near the dumpsite could be explained by the failure to fulfill sanitary 627 landfill requirements and the lack of pollution control facilities and leachate treatment in Mahmood 628 Booti. Similarly, atmospheric concentrations of \sum 9PBDEs (BDE-17, -28, -47, -99, -100, -153, -154, 629 -183, and -209), \sum_{3} HBCDDs (α -, β -, and γ -HBCDD), EH-TBB, BEH-TEBP, and BTBPE in landfill 630 sites were also relatively higher than those observed in other zones (including industrial, urban, 631 semi-urban, and rural areas) in Gauteng Province, South Africa, which could probably be attributed

to the improper management of wastes disposed in the landfills (Katima et al., 2018). To the best of

633 our knowledge, no data exist on atmospheric concentrations of OPEs in the vicinity of regulated

- 634 waste dumpsites or landfills, thus further studies should be undertaken to fill this knowledge gap.
- 635

636 5.3 Concentrations of FRs in air near waste incineration plants

637 Data on atmospheric concentrations of FRs in incineration plants is very limited, with only 3 638 publications located and included in this review (Table 4). Atmospheric \sum_{30} PBDE (di- to deca-BDE) 639 concentrations ranged from 24.9-139 pg·m⁻³ and 32.1-71.6 pg·m⁻³ in two municipal solid waste incinerators in Taiwan, China, with median values of 51.9 and 48.9 pg·m⁻³, respectively (Tu et al., 640 641 2012). Similarly, Wang et al. (2010b) also observed comparable \sum_{30} PBDE concentrations (25.7-100 642 pg·m⁻³) and significantly lower Σ PBB concentrations (congeners not reported; range: 0.149-0.556 643 $pg \cdot m^{-3}$) in ambient air collected from one municipal solid waste incinerator in Taiwan, China. The 644 contribution of the two municipal solid waste incinerators to total PBDE concentration in ambient 645 air was only 0.026%, indicating minimal impact of incinerators on PBDE emissions to ambient air 646 (Tu et al., 2012). This was further supported by the comparable atmospheric concentrations of 647 PBDEs in incineration plants to those detected in ambient air collected in urban areas in Taiwan, 648 China (\sum_{30} PBDE median: 34.7 pg·m⁻³) (Wang et al., 2011). PBDE concentrations in ambient air 649 were also studied in the vicinity of a solid waste incinerator in Sweden (Agrell et al., 2004). 650 Specifically, *S*₈PBDEs (BDE-28, -47, -66, -100, -153, -154, -183, and -209) were determined to fall 651 in the range 2.2-123.5 pg·m⁻³, with a median value of 19.2 pg·m⁻³. These were generally at the same 652 level to \sum_{B} PBDE concentrations (range: ND-192.8 pg·m⁻³; median: 15.1 pg·m⁻³) detected in outdoor 653 air at urban reference sites. A significant difference (paired t-test, p<0.01) was observed between 654 atmospheric concentrations of BDE-47 and Σ_7 PBDEs (excluding BDE-209) between incinerator 655 sites and urban reference sites, but not for BDE-209 and \sum_{s} PBDEs (paired t-test, p>0.1). Given that 656 the commercial deca-BDE mixture was still in widespread use in Sweden during this study (2001-657 2002), the authors suggested that the comparable BDE-209 concentrations at incinerator-impacted 658 and urban reference sites were a reflection of BDE-209 emissions from in-use consumer products; 659 but that for those "older" PBDEs (i.e., commercial penta-BDEs) which had at the time of the study 660 already been restricted in the country, waste incineration might contribute meaningfully to their

occurrence in outdoor air (Agrell et al., 2004). To the best of our knowledge, no data is available on
atmospheric concentrations of OPEs in the vicinity of incineration plants, and further studies are
recommended to fill this knowledge gap.

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6. Conclusions and research gaps

666 This study reviews three pathways of HFR and OPE transfer from FR-containing wastes to the 667 outdoor environment, i.e., volatilisation, abrasion, and leaching. Several factors are likely to 668 contribute greatly to FR emissions to air, including: higher FR vapour pressure, increased 669 temperature, and elevated concentrations of FRs in products; while the highest emission rates occur 670 at the beginning of chamber tests. Abrasion of fine particles or fibres from products to soil is likely, 671 and current evidence (although very limited, and further investigations are required) suggests that 672 such abrasion processes likely contribute significantly to FR transfer from products to soil. 673 Furthermore, higher DHM concentrations in leachate, increased leachate temperature, lower 674 waste:leachate ratios, agitation, and higher concentrations of HFRs and OPEs in treated products all 675 contribute to elevated leaching rates, while the impact of leachate pH on the leaching behaviour of 676 HFRs and OPEs remains unclear. Leaching of BFRs is likely to be a second-order process, i.e., high 677 initial leaching rates that diminish considerably at longer contact times. However, despite the first-678 order leaching of TCIPP from furniture PUF observed in one study (Stubbings and Harrad, 2018), 679 leaching characteristics of OPEs are poorly understood due to very limited data, and follow-up 680 studies are strongly recommended.

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682 We also reviewed the occurrence of HFRs and OPEs in outdoor air and soil in the vicinity of formal 683 waste treatment facilities, including formal e-waste dismantling and recycling facilities, official 684 waste dumpsites and landfills, and waste incineration plants. Despite the environmentally friendly 685 technologies adopted in formal e-waste facilities, such activities likely contribute significantly to 686 ambient air and soil contamination, evidenced by the elevated concentrations of HFRs and OPEs 687 frequently reported in atmosphere and soil close to formal e-waste treatment facilities. Waste 688 dumpsites and landfills may constitute a further source of HFRs and OPEs in surrounding soil. 689 However, the contribution of waste dumpsites and landfills to atmospheric HFR concentrations 690 appears much smaller than that of formal e-waste facilities, even though improper management of 691 waste disposal might also contribute to HFR contamination in ambient air. By contrast, despite the 692 limited data, current evidence suggests the impact of waste incineration plants on BFR 693 contamination of outdoor air and soil is minimal. Based on our findings in this work as well as in a 694 previous publication (Ma et al., 2021), we believe government regulation and proper management 695 of waste disposal are important. On the one hand, unregulated waste treatment needs to be stopped, 696 and formal waste activities should be conducted with environmentally-sound techniques and 697 personal protective equipment fully implemented. On the other hand, more attention should be paid 698 to e-waste recycling and dismantling (both regulated and unregulated), as well as unregulated 699 landfilling, as these activities are more likely to increase environmental burdens of HFRs and OPEs. 700

701 This review highlights that our current understanding of HFR and OPE contamination in outdoor 702 air and soil resulting from formal waste treatment activities is still limited. Current data on OPEs 703 are rather scarce, especially as this review could not identify any data on the occurrence of OPEs 704 either in ambient air near landfills or in ambient air and soil near incinerators. The relationship 705 between FR contamination and landfill/incinerator size remains unclear. Moreover, only a small 706 number of studies exist that address the occurrence of HFRs in outdoor air and soil in the vicinity 707 of waste incineration plants, making it difficult to evaluate the impact of waste incineration 708 processes (e.g., incinerator size, waste incineration technologies, and treatment of flue gas, etc.) on 709 HFR and OPE contamination in the surrounding environment. As we move progressively deeper 710 into the end-of-life phase of the life cycle of many FRs, far greater priority needs to be assigned to 711 research that will fill the considerable gaps in our understanding of the environmental and human 712 health impacts of the handling of FR-containing waste (particularly NBFR- and OPE-containing 713 waste).

714

715 **Declarations of interest**

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- 717
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- 723 Supplementary data to this article can be found online at

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FRs	Product	Temperature (°C)	SER (µg·m ⁻² ·h ⁻¹)	Reference
BDE-28	TV housing	23	0.0002	(Kemmlein et al., 2003)
BDE-47	TV housing	23	0.0066	(Kemmlein et al., 2003)
BDE-66	TV housing	23	0.0005	(Kemmlein et al., 2003)
BDE-99	TV housing	23	0.0017	(Kemmlein et al., 2003)
BDE-100	TV housing	23	0.0005	(Kemmlein et al., 2003)
BDE-153	TV housing	23	0.001	(Kemmlein et al., 2003)
BDE-154	TV housing	23	0.0002	(Kemmlein et al., 2003)
hepta-BDE	TV housing	23	0.0045	(Kemmlein et al., 2003)
octa-BDE	TV housing	23	0.0015	(Kemmlein et al., 2003)
nona-BDE	TV housing	23	0.0008	(Kemmlein et al., 2003)
deca-BDE	TV housing	23	0.0003	(Kemmlein et al., 2003)
∑PBDEs	polyester textile	20	0.0022	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	40	0.0062	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	60	0.0048	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	80	0.029	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	20	0.065-0.098	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	40	0.082-0.61	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	60	0.22-0.27	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	80	4.2-5.1	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	20	0.025-0.044	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	40	0029-0.29	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	60	0.027-0.042	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	80	0.66-0.88	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	20	0.068-0.11	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	40	0.080-0.66	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	60	0.049-0.087	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	80	0.88-1.5	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	20	0.16-0.25	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	40	0.19-1.6	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	60	0.30-0.40	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	80	5.7-7.5	(Kajiwara and Takigami, 2013)
∑HBCDDs	insulating board	23	0.0001-0.029	(Kemmlein et al., 2003)
TDCIPP	polyester curtain	20	0.044-0.17	(Tokumura et al., 2019)
TDCIPP	polyester curtain	60	11	(Tokumura et al., 2019)
ТСР	polyester curtain	20	0.06	(Tokumura et al., 2019)
TCIPP	PVC wallpaper	25	645	(Ni et al., 2007)
TCIPP	PVC wallpaper	40	1136	(Ni et al., 2007)
TCIPP	PVC wallpaper	60	2841	(Ni et al., 2007)
TCIPP	insulating board	23	0.21-0.70	(Kemmlein et al., 2003)
TCIPP	assembly foam	23	50-140	(Kemmlein et al., 2003)
TCIPP	upholstery stool	23	28-77	(Kemmlein et al., 2003)
TCIPP	mattress	23	0.012	(Kemmlein et al., 2003)

1059 Table 1. Reported area specific emission rates (SER) of some HFRs and OPEs

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Table 2. Occurrence of Pl	BDEs in soil in the	vicinity of formal wa	aste treatment faciliti	S	
Waste treatment	Country	Sampling period	PBDE congeners	Concentration (ng·g ⁻¹ dw)	Reference
method					
e-waste recycling	China	2018	\sum_{20} PBDEs	46300 (reference soil: 575) ^a	(Ge et al., 2020)
e-waste recycling	China	2015	\sum_{13} PBDEs	13 ^b	(Wu et al., 2019)
e-waste recycling	China	2015	BDE-209	90 p	(Wu et al., 2019)
e-waste recycling	China	2011	\sum_{13} PBDEs	250 b	(Hong et al., 2018)
e-waste recycling	China	2012	\sum_{23} PBDEs	3900 (background soil: 0.77) ^b	(Li et al., 2016)
e-waste recycling	China	2009	\sum_{18} PBDEs	34-1069 °	(Li et al., 2014)
e-waste recycling	China	2009	BDE-209	110-5850 °	(Li et al., 2014)
e-waste storage	China		\sum_{22} PBDEs	50.5 (agricultural soil: 27.5) ^b	(Man et al., 2011)
e-waste dismantling	China		\sum_{22} PBDEs	6875 (agricultural soil: 27.5) ^b	(Man et al., 2011)
e-waste recycling	Australia	2017	Σ_8 PBDEs	130-160 (urban soil: 21) ^a	(McGrath et al., 2018)
e-waste recycling	South Korea	2012	\sum_{23} PBDEs	9.0 (background soil: 1.4) ^b	(Li et al., 2016)
e-waste recycling	Vietnam	2012	\sum_{23} PBDEs	68 (background soil: 0.23) ^b	(Li et al., 2016)
landfill	China	2011-2012	Σ_8 PBDEs	234 a	(Huang et al., 2013)
landfill	China	2017	\sum_{12} PBDEs	0.13-1.2 °	(Li et al., 2018b)
landfill	Ireland	2018-2019	$\Sigma_7 PBDEs$	0.14 ^a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	BDE-209	0.57 a	(Harrad et al., 2020b)
landfill	Brazil	2015	Σ_8 PBDEs	276 ^b	(Cristale et al., 2019)
landfill	South Africa		∑7PBDEs	7.43 a	(Akortia et al., 2019)
landfill	Canada	2004-2006	Σ_{60} PBDEs	131 (background soil: 1.94) ^b	(Danon-Schaffer et al., 2008)
dumpsite	Pakistan		Σ_8 PBDEs	1.11 ^b	(Hafeez et al., 2016)
incinerator	Australia	2014	Σ_8 PBDEs	13.6-80.8 (reference soil: 0.12-43.8) °	(McGrath et al., 2016)
incinerator	China		\sum_{42} PBDEs	1.4 a	(Zhang et al., 2013)
Note: a: median concentration;	b: mean concentration	; c: range			

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Table 5. Occurrence of first	NS (F DDES EXCIU	ited) and OF ES III SOILT	и спе менну от тог	mai waste treatment facilities	
Waste treatment method	Country	Sampling period	FRs	Concentration (ng·g ⁻¹ dw)	References
e-waste recycling	China	2018	PBBz	47.9 (reference soil: 0.58) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	HBBz	249 (reference soil: 0.57) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_2 DP_s$	712 (reference soil: 11.3) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	Σ_{13} OPEs	12000 (reference soil: 256) ^a	(Ge et al., 2020)
e-waste recycling	China	2011	\sum_{14} NBFRs	128 ^b	(Hong et al., 2018)
e-waste recycling	China	2011	$\Sigma_2 DP_s$	34.7 b	(Hong et al., 2018)
e-waste recycling	China	2012	\sum_{19} NBFRs	800 (reference soil: 12) ^b	(Li et al., 2017)
e-waste recycling	South Korea	2012	\sum_{19} NBFRs	18 (reference soil: 13) ^b	(Li et al., 2017)
e-waste recycling	Vietnam	2012	\sum_{19} NBFRs	21 (reference soil: 0.68) ^b	(Li et al., 2017)
e-waste recycling	Australia	2017	$\Sigma_6 \text{NBFRs}$	3.8-15 (urban soil: ND) ^a	(McGrath et al., 2018)
multi-waste recycling	China		\sum_{12} OPEs	116 (farmland soil) 56.3 ^a	(Wang et al., 2018b)
landfill	Ireland	2018-2019	Σ_3 HBCDDs	0.55 a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	DBDPE	ND a	(Harrad et al., 2020b)
landfill	Brazil	2015	\sum_{10} OPEs	67 b	(Cristale et al., 2019)
landfill	Brazil	2015	$\Sigma_4 NBFRs$	19 ^b	(Cristale et al., 2019)
landfill	China	2017	SCCPs	56.8-1348 °	(Li et al., 2021)
dumpsite	Pakistan		$\Sigma_2 DPs$	0.48 ^b	(Hafeez et al., 2016)
incinerator	Australia	2014	HBBz	ND-0.34 (reference soil: ND) °	(McGrath et al., 2017b)
Note: a: median concentration; b:	mean concentration;	c: range.			

Table 3. Occurrence of HFRs (PBDEs excluded) and OPEs in soil in the vicinity of formal wa P t atment facilities

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Table 4. Occurrence of HFRs and OPEs in outdoor air in the vicinity of formal waste treatment facilities

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(Wang et al., 2010b)	0.149-0.556 °	$\Sigma PBBs$	I	China	incinerator
(Wang et al., 2010b)	25.7-100 °	\sum_{30} PBDEs	I	China	incinerator
(Tu et al., 2012)	24.9-139 °	\sum_{30} PBDEs	2009	China	incinerator

L LOW 5 ш, о. auon, c. range.

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FRs	Product	Temperature (°C)	SER (µg·m ⁻² ·h ⁻¹)	Reference
BDE-28	TV housing	23	0.0002	(Kemmlein et al., 2003)
BDE-47	TV housing	23	0.0066	(Kemmlein et al., 2003)
BDE-66	TV housing	23	0.0005	(Kemmlein et al., 2003)
BDE-99	TV housing	23	0.0017	(Kemmlein et al., 2003)
BDE-100	TV housing	23	0.0005	(Kemmlein et al., 2003)
BDE-153	TV housing	23	0.001	(Kemmlein et al., 2003)
BDE-154	TV housing	23	0.0002	(Kemmlein et al., 2003)
hepta-BDE	TV housing	23	0.0045	(Kemmlein et al., 2003)
octa-BDE	TV housing	23	0.0015	(Kemmlein et al., 2003)
nona-BDE	TV housing	23	0.0008	(Kemmlein et al., 2003)
deca-BDE	TV housing	23	0.0003	(Kemmlein et al., 2003)
∑PBDEs	polyester textile	20	0.0022	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	40	0.0062	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	60	0.0048	(Kajiwara and Takigami, 2013)
∑PBDEs	polyester textile	80	0.029	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	20	0.065-0.098	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	40	0.082-0.61	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	60	0.22-0.27	(Kajiwara and Takigami, 2013)
α-HBCDD	polyester textile	80	4.2-5.1	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	20	0.025-0.044	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	40	0029-0.29	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	60	0.027-0.042	(Kajiwara and Takigami, 2013)
β-HBCDD	polyester textile	80	0.66-0.88	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	20	0.068-0.11	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	40	0.080-0.66	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	60	0.049-0.087	(Kajiwara and Takigami, 2013)
γ-HBCDD	polyester textile	80	0.88-1.5	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	20	0.16-0.25	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	40	0.19-1.6	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	60	0.30-0.40	(Kajiwara and Takigami, 2013)
∑HBCDDs	polyester textile	80	5.7-7.5	(Kajiwara and Takigami, 2013)
∑HBCDDs	insulating board	23	0.0001-0.029	(Kemmlein et al., 2003)
TDCIPP	polyester curtain	20	0.044-0.17	(Tokumura et al., 2019)
TDCIPP	polyester curtain	60	11	(Tokumura et al., 2019)
ТСР	polyester curtain	20	0.06	(Tokumura et al., 2019)
TCIPP	PVC wallpaper	25	645	(Ni et al., 2007)
TCIPP	PVC wallpaper	40	1136	(Ni et al., 2007)
TCIPP	PVC wallpaper	60	2841	(Ni et al., 2007)
TCIPP	insulating board	23	0.21-0.70	(Kemmlein et al., 2003)
TCIPP	assembly foam	23	50-140	(Kemmlein et al., 2003)
TCIPP	upholstery stool	23	28-77	(Kemmlein et al., 2003)
TCIPP	mattress	23	0.012	(Kemmlein et al., 2003)

1 Table 1. Reported area specific emission rates (SER) of some HFRs and OPEs

Table 2. Occurrence of P	BDEs in soil in the	vicinity of formal wa	iste treatment facilitie	28	
Waste treatment	Country	Sampling period	PBDE congeners	Concentration (ng·g ⁻¹ dw)	Reference
method					
e-waste recycling	China	2018	\sum_{20} PBDEs	46300 (reference soil: 575) ^a	(Ge et al., 2020)
e-waste recycling	China	2015	\sum_{13} PBDEs	13 ^b	(Wu et al., 2019)
e-waste recycling	China	2015	BDE-209	90 p	(Wu et al., 2019)
e-waste recycling	China	2011	\sum_{13} PBDEs	250 ^b	(Hong et al., 2018)
e-waste recycling	China	2012	\sum_{23} PBDEs	3900 (background soil: 0.77) ^b	(Li et al., 2016)
e-waste recycling	China	2009	\sum_{18} PBDEs	34-1069 °	(Li et al., 2014)
e-waste recycling	China	2009	BDE-209	110-5850 °	(Li et al., 2014)
e-waste storage	China		\sum_{22} PBDEs	50.5 (agricultural soil: 27.5) ^b	(Man et al., 2011)
e-waste dismantling	China		\sum_{22} PBDEs	6875 (agricultural soil: 27.5) ^b	(Man et al., 2011)
e-waste recycling	Australia	2017	\sum_{8} PBDEs	130-160 (urban soil: 21) ^a	(McGrath et al., 2018)
e-waste recycling	South Korea	2012	\sum_{23} PBDEs	9.0 (background soil: 1.4) ^b	(Li et al., 2016)
e-waste recycling	Vietnam	2012	\sum_{23} PBDEs	68 (background soil: 0.23) ^b	(Li et al., 2016)
landfill	China	2011-2012	\sum_{8} PBDEs	234 a	(Huang et al., 2013)
landfill	China	2017	\sum_{12} PBDEs	0.13-1.2 °	(Li et al., 2018b)
landfill	Ireland	2018-2019	$\Sigma_7 PBDEs$	0.14 ^a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	BDE-209	0.57 a	(Harrad et al., 2020b)
landfill	Brazil	2015	\sum_{8} PBDEs	276 ^b	(Cristale et al., 2019)
landfill	South Africa		$\Sigma_7 PBDEs$	7.43 a	(Akortia et al., 2019)
landfill	Canada	2004-2006	\sum_{60} PBDEs	131 (background soil: 1.94) ^b	(Danon-Schaffer et al., 2008)
dumpsite	Pakistan		\sum_{8} PBDEs	1.11 ^b	(Hafeez et al., 2016)
incinerator	Australia	2014	\sum_{8} PBDEs	13.6-80.8 (reference soil: 0.12-43.8) °	(McGrath et al., 2016)
incinerator	China		\sum_{42} PBDEs	1.4 ^a	(Zhang et al., 2013)
Note: a: median concentration;	b: mean concentration	; c: range			

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Table 5. Occurrence of mrn	vs (r diffes exclud	ieu) allu Of Es III sull II	r the vicinity of for	mai waste treatment facilities	
Waste treatment method	Country	Sampling period	FRs	Concentration (ng·g ⁻¹ dw)	References
e-waste recycling	China	2018	PBBz	47.9 (reference soil: 0.58) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	HBBz	249 (reference soil: 0.57) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_2 DP_S$	712 (reference soil: 11.3) ^a	(Ge et al., 2020)
e-waste recycling	China	2018	\sum_{13} OPEs	12000 (reference soil: 256) ^a	(Ge et al., 2020)
e-waste recycling	China	2011	\sum_{14} NBFRs	128 ^b	(Hong et al., 2018)
e-waste recycling	China	2011	$\sum_2 DP_S$	34.7 ^b	(Hong et al., 2018)
e-waste recycling	China	2012	\sum_{19} NBFRs	800 (reference soil: 12) ^b	(Li et al., 2017)
e-waste recycling	South Korea	2012	\sum_{19} NBFRs	18 (reference soil: 13) ^b	(Li et al., 2017)
e-waste recycling	Vietnam	2012	\sum_{19} NBFRs	21 (reference soil: 0.68) ^b	(Li et al., 2017)
e-waste recycling	Australia	2017	$\Sigma_6 NBFRs$	3.8-15 (urban soil: ND) ^a	(McGrath et al., 2018)
multi-waste recycling	China		\sum_{12} OPEs	116 (farmland soil) 56.3 ^a	(Wang et al., 2018b)
landfill	Ireland	2018-2019	Σ_3 HBCDDs	0.55 ^a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	DBDPE	ND a	(Harrad et al., 2020b)
landfill	Brazil	2015	\sum_{10} OPEs	67 ^b	(Cristale et al., 2019)
landfill	Brazil	2015	\sum_{4} NBFRs	19 ^b	(Cristale et al., 2019)
landfill	China	2017	SCCPs	56.8-1348 °	(Li et al., 2021)
dumpsite	Pakistan		$\sum_2 DPs$	0.48 ^b	(Hafeez et al., 2016)
incinerator	Australia	2014	HBBz	ND-0.34 (reference soil: ND) °	(McGrath et al., 2017b)
Note: a: median concentration; b: 1	mean concentration;	c: range.			

Table 3. Occurrence of HFRs (PBDEs excluded) and OPEs in soil in the vicinity of formal waste treatment facilities

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Waste treatment method	Country	Sampling period	FRs	Concentration (pg·m ⁻³)	References
e-waste recycling	China	2011	\sum_{13} PBDEs	10600	(Hong et al., 2018)
e-waste recycling	China	2011	\sum_{16} NBFRs	1330	(Hong et al., 2018)
e-waste recycling	China	2011	$\sum_2 DP_S$	109	(Hong et al., 2018)
e-waste recycling	China	2015-2016	\sum_{12} OPEs	3300 (urban area: 2800) ^a	(Wang et al., 2018a)
e-waste recycling	SN	2004	\sum_{30} PBDEs	340-8600 (control site: 93) b	(Cahill et al., 2007)
e-waste recycling	South Korea	2012	\sum_{37} PBDEs	321-5550 (reference site: 77.5) °	(Park et al., 2014)
e-waste storage	Thailand	2007-2008	\sum_{10} PBDEs	8-150 °	(Muenhor et al., 2010)
vehicle dismantling	China	2012-2013	\sum_{30} PBDEs	200-494 ^b	(Gou et al., 2016)
metal recycling	SN	2004	\sum_{30} PBDEs	390-810 (control site: 93) ^b	(Cahill et al., 2007)
landfill	Ireland	2018-2019	BDE-47	0.23 ^a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	BDE-209	4.3 ^a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	Σ_3 HBCDDs	ND a	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	DBDPE	ND ^a	(Harrad et al., 2020b)
landfill	South Africa	2016-2017	\sum_{9} PBDEs	954-2820 (rural site: 100-284) °	(Katima et al., 2018)
landfill	South Africa	2016-2017	\sum_{3} HBCDDs	50.3-117 (rural site: ND-100) °	(Katima et al., 2018)
landfill	South Africa	2016-2017	EH-TBB	ND-2070 (rural site: ND-69.5) °	(Katima et al., 2018)
landfill	South Africa	2016-2017	BEH-TEBP	ND-1200 (rural site: ND) °	(Katima et al., 2018)
landfill	South Africa	2016-2017	BTBPE	ND-1400 (rural site: ND-46.5) °	(Katima et al., 2018)
landfill	Canada	2004-2005	\sum_{16} PBDEs	0.72-145 °	(St-Amand et al., 2008)
landfill	Germany	2009	∑7PBDEs	0.4-10.7 (reference site: ND-33.5) °	(Weinberg et al., 2011)
dumpsite	Pakistan		\sum_{8} PBDEs	212 ^b	(Hafeez et al., 2016)
dumpsite	Pakistan		$\sum_{2} DPs$	0.58 ^b	(Hafeez et al., 2016)
incinerator	Sweden	2001-2002	\sum_{8} PBDEs	19.2 (urban area: 15.1) ^a	(Agrell et al., 2004)

Table 4. Occurrence of HFRs and OPEs in outdoor air in the vicinity of formal waste treatment facilities

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incinerator	China	2009	\sum_{30} PBDEs	24.9-139 °	(Tu et al., 2012)
incinerator	China		\sum_{30} PBDEs	25.7-100 °	(Wang et al., 2010b)
incinerator	China		ΣPBBs	0.149-0.556 °	(Wang et al., 2010b)
Note: a: median concentration	1: h. mean concentrat	ion. C. range			

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