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# **Human exposure to halogenated and organophosphate flame retardants through informal e-waste handling activities - a critical review**

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1 **Abstract**

2 Informal electrical and electronic waste (e-waste) handling activities constitute a potentially  
3 important source of halogenated (HFRs) and organophosphate flame retardants (OPFRs) to the  
4 environment and humans. In this review, two electronic databases (ScienceDirect and Web of  
5 Science Core Collection) were searched for papers that addressed this topic. A total of 82 relevant  
6 studies (including 72 studies selected from the two databases and 10 studies located from the  
7 references of the first 72 selected studies) were identified that reported on human external and  
8 internal exposure to HFRs and OPFRs arising as a result of informal e-waste handling activities.  
9 Compared to the general population, higher levels of external exposure (i.e., inhalation, ingestion,  
10 and dermal absorption) and internal exposure (i.e., blood serum, hair, breast milk, urine, and other  
11 human matrices) to HFRs and OPFRs were identified for e-waste recyclers and residents inhabiting  
12 e-waste dismantling and recycling zones, especially for younger adults and children. Food intake  
13 and dust ingestion were the dominant exposure pathways for the majority of brominated flame  
14 retardants (BFRs) and dechlorane plus (DP); while inhalation was identified as the most significant  
15 pathway of human exposure to OPFRs in informal e-waste sites. The majority of research to date  
16 has focused on China and thus future studies should be conducted in other regions such as Africa  
17 and South Asia. Other suggested foci of future research are: examination of exposure via dermal  
18 contact with e-waste, dietary exposure of local populations to OPFRs, confirmation of the existence  
19 of and cause(s) of the higher body burdens of females compared with males amongst populations  
20 impacted by informal e-waste handling, and characterisation of exposure of such populations to  
21 chlorinated paraffins.

22

23 **Keywords:** WEEE; Brominated flame retardants; Organophosphate esters; Chlorinated paraffins

24

25 **Main findings:** Inhalation contributes most to human exposure to OPFRs, and dietary intake  
26 contributes most to human exposure to BFRs and DPs. Children and females are more exposed.

27

28 **1. Introduction**

29 Electrical and electronic waste (e-waste), also called waste electrical and electronic equipment  
30 (WEEE), has become a global concern. In 2019, global generation of e-waste reached 53.6 million

31 tonnes (Mt), and this figure is estimated to reach 74 Mt by 2030 and 120 Mt by 2050, respectively  
32 (Forti et al., 2020; World Economic Forum, 2019). Typically, household appliances like washing  
33 machines, telecommunications, and IT equipment including computers, and consumer articles like  
34 TVs, comprise the majority of e-waste generated globally (Akram et al., 2019). Due to  
35 environmental and economic considerations, much of the e-waste generated in high-income  
36 countries has and continues to be exported for handling in low- and moderate-income countries  
37 (Mihai et al., 2019; Pathak et al., 2017). Although measures outlined in the Basel Convention were  
38 designed to prohibit the export of hazardous wastes to low- and moderate-income countries, these  
39 are unlikely to be effective as some used electrical and electronic equipment is characterised as non-  
40 hazardous waste (Khan, 2016). Moreover, it was not until June 2019 when the EU clarified their  
41 determinations on the regulation of persistent organic pollutants (POPs), that many non-hazardous  
42 WEEE items would be reclassified as hazardous and become subject to the Hazardous Waste  
43 Regulations 2005 (Official Journal of the European Union, 2019). Moreover, in low- and moderate-  
44 income countries, approximately 90 % of e-waste disposal and recycling activities are undertaken  
45 by informal sector workers (Pathak et al., 2019). In India, for instance, 97 % of total e-waste  
46 generated is handled by informal recycling yards in major cities like Delhi, Mumbai, Hyderabad,  
47 and Bangalore (Rao et al., 2017).

48

49 This is concerning as various contaminants including halogenated flame retardants (HFRs) and  
50 organophosphate flame retardants (OPFRs) can enter environmental media through different  
51 activities reported to be undertaken at informal e-waste handling facilities. Informal e-waste  
52 activities refer to unlicensed or unregulated e-waste dismantling and recycling activities that are  
53 usually practiced by individuals and families using primitive techniques. These include: open  
54 burning, acid leaching, and heating (Leung, 2019). The risks posed by such emissions are  
55 compounded by the lack of effective personal protective equipment for use by those undertaking  
56 such work, as well as by absence of other measures designed to protect the environment and human  
57 health (Lundgren, 2012). HFRs and OPFRs have been widely used as additives at concentrations of  
58 up to 10-15 % by weight in the plastic housing of various electric and electronic products to ensure  
59 that these products meet fire safety regulations (Lassen and Havelund, 2006). However, from the  
60 mid-2000s onwards, concerns about the adverse environmental and human health impacts of HFRs

61 like polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) would lead  
62 to restrictions and bans on their manufacture and use in new products in many jurisdictions,  
63 including their listing under the Stockholm Convention on POPs of the United Nations Environment  
64 Programme (UNEP) (Ge et al., 2020; Harrad, 2015; Huang et al., 2018; Ma et al., 2017b). As a  
65 result, there has been increased demand for OPFRs and other HFRs such as tetrabromobisphenol-A  
66 (TBBP-A), novel brominated flame retardants (NBFRs), dechlorane plus (DP), and chlorinated  
67 paraffins (CPs) (Chen et al., 2015; Ge et al., 2020; Gravel et al., 2020; He et al., 2017; Liu et al.,  
68 2016; Ma et al., 2017a; Zeng et al., 2018, 2020).

69  
70 Previous studies have determined high concentrations of HFRs and OPFRs in environmental media  
71 (Anh et al., 2017; Iqbal et al., 2017; Qin et al., 2019; Wang et al., 2018; Zheng et al., 2015b) and  
72 foodstuffs (Anh et al., 2017; Huang et al., 2018; Tao et al., 2016; Zheng et al., 2015a) at various  
73 informal e-waste dismantling and recycling sites such as Bui Dau (Vietnam), Guiyu and Qingyuan  
74 (China), and Karachi (Pakistan). Such findings indicate high external exposure of local populations  
75 (resident exposure) to HFRs and OPFRs through inhalation (Iqbal et al., 2017; Qin et al., 2019),  
76 ingestion (Anh et al., 2017; Huang et al., 2018), and dermal absorption (Wu et al., 2016a). Elevated  
77 internal exposure of local residents has also been demonstrated by reported concentrations of HFRs  
78 or OPFRs in human matrices like serum (Guo et al., 2018, 2020; Lv et al., 2015), hair (Liang et al.,  
79 2016; Qiao et al., 2019), milk (Li et al., 2017), and urine (Lu et al., 2017; Shi et al., 2019) in informal  
80 e-waste sites in South China (e.g., Qingyuan, Luqiao, and Wenling). Such evidence of elevated  
81 human exposure has raised concerns about potential adverse health effects on populations impacted  
82 by informal e-waste treatment (Akram et al., 2019; Asante et al., 2019; Awasthi et al., 2016, 2018;  
83 Bakhiyi et al., 2018; Orisakwe et al., 2019; Shi et al., 2018). For instance, BFRs are endocrine  
84 disruptors (Eguchi et al., 2015; Guo et al., 2018, 2019b; Zheng et al., 2017a, 2017b), and could exert  
85 adverse effects on human semen quality (Yu et al., 2018); while OPFRs have been associated with  
86 greater DNA damage (Lu et al., 2017). Moreover, health effects of HFR and OPFR exposure on  
87 fetuses and infants are of particular concern due to their substantially weaker resistance and  
88 immunity (Bai et al., 2019; Li et al., 2018; Xu et al., 2015; Zheng et al., 2017b).

89  
90 The present review aims to: 1) summarise current research into human external exposure (i.e.,

91 inhalation, ingestion of food, dust, and soil, and dermal absorption) and internal exposure (i.e., blood  
92 and serum, hair, breast milk, urine, and other human matrices) to HFRs and OPFRs through informal  
93 e-waste dismantling and recycling activities for local residents; 2) identify potential health risks to  
94 local residents in informal e-waste sites; and 3) highlight substantial research gaps that require  
95 urgent investigation.

96

## 97 **2. Methods**

98 Between 26/09/2019 and 07/09/2020, two electronic databases (ScienceDirect and Web of Science  
99 Core Collection) were searched for research articles, reviews, book chapters, and other online  
100 resources. Terms searched were: “e-waste”, “human exposure”, and “flame retardants”, with only  
101 papers published between 2015 and 2020 selected. Using these search criteria, 1651 publications  
102 were found on ScienceDirect, with a further 131 papers located on Web of Science Core Collection.  
103 Further inspection by the authors comprising screening titles and abstracts identified relevant  
104 publications (101 from ScienceDirect and 82 from Web of Science Core Collection). After removal  
105 of duplicates (n=33), 150 publications remained for further screening.

106

107 These 150 publications were further rated for relevance by screening sampling methodology,  
108 statistical data presented, and conclusions. As a result, 78 articles were excluded (including one  
109 article not written in English), leaving 72 publications (consisting of 15 review articles, 4 book  
110 chapters, and 53 research articles). In addition, references cited in these 72 selected publications  
111 were also reviewed, adding a further 10 publications to the total included in this review. These  
112 include: 1) four research papers published before 2015 but essential to this review in terms of data  
113 interpretation (Ali et al., 2012; Labunska et al., 2014; Tue et al., 2013; Wang et al., 2010); 2) three  
114 publications published between 2015 and 2020 but could not be located under current search  
115 techniques due to their different foci (Abdallah and Harrad, 2018; Khan, 2016; Liu et al., 2016); and  
116 3) three official reports (Lassen and Havelund, 2006; Lundgren, 2012; United States Environmental  
117 Protection Agency, 2017).

118

## 119 **3. Geographical distribution of studies into HFR and OPFR contamination from e-waste** 120 **handling**

121 It is notable that all the 53 selected research articles were conducted in lower- and upper-middle  
122 income countries, according to the latest classification made by the World Bank (World Bank, 2020).  
123 These include China (n=39), Vietnam (n=7), Thailand (n=2), Pakistan (n=1), Nigeria (n=1), South  
124 Africa (n=1), Ghana (n=1), and Bangladesh (n=1) (Fig. 1).

125

#### 126 **4 Human external exposure to HFRs and OPFRs arising from informal e-waste activities**

127 Previous studies have indicated 3 major pathways of human external exposure to HFRs and OPFRs  
128 from informal e-waste activities, i.e., inhalation, ingestion of soil, dust, and food, and dermal  
129 absorption from soil, dust, and e-waste articles (Anh et al., 2017; Huang et al., 2018; Iqbal et al.,  
130 2017; Qin et al., 2019; Wu et al., 2016a; Zheng et al., 2016). Once HFRs or OPFRs enter the  
131 environment through informal e-waste activities, they can transfer between different environmental  
132 media (Akram et al., 2019; Anh et al., 2017; Wang et al., 2018; Zheng et al., 2016). Particularly  
133 important, HFRs and OPFRs can enter the food chain through bioconcentration and  
134 biomagnification, thereby entering the human body through food ingestion resulting in exposure to  
135 individuals beyond those directly undertaking e-waste handling, including young children (Anh et  
136 al., 2017; Sun et al., 2018; Wu et al., 2019; Zheng et al., 2016). Transfer of HFRs and OPFRs  
137 between different environmental media and resultant human external exposure pathways are shown  
138 in Fig.2, with recent studies (i.e. those published between 2015 and 2020) concerning human  
139 external exposure to OPFRs and HFRs (including PBDEs, NBFRs, HBCDDs, TBBP-A, DPs, and  
140 CPs) through informal e-waste handling activities summarised in Tables S1-S5 (Supplementary  
141 Material).

142

#### 143 **4.1 Human inhalation exposure to HFRs and OPFRs arising from informal e-waste activities**

144 Inhalation of air has been highlighted as an important pathway of human exposure to HFRs and  
145 OPFRs in informal e-waste dismantling and recycling sites, especially for OPFRs (Awasthi et al.,  
146 2016; Iqbal et al., 2017; Jiang et al., 2019; Luo et al., 2016). A study conducted in four e-waste  
147 recycling sites (Jacob Lines, Surjani Town, Lyari, and Shershah) in Karachi City, Pakistan identified  
148 human inhalation exposure to a range of HFRs and OPFRs. Specifically, these comprised: 8 PBDE  
149 congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209), 6 NBFRs (pentabromoethylbenzene  
150 (PBEB), hexabromobenzene (HBB), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB), 1,2-

151 bis(2,4,6-tribromophenoxy) ethane (BTBPE), bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (BEH-  
152 TBP), and decabromodiphenylethane (DBDPE)), 7 OPFRs (tris-(2-chloroethyl)-phosphate (TCEP),  
153 tris-(2,3-dichloropropyl)-phosphate (TDCIPP), ethylhexyl diphenyl phosphate (EHDPP), tris(2-  
154 ethylhexyl)phosphate (TEHP), tri-*n*-butyl phosphate (TnBP), tris-(2-chloroisopropyl)-phosphate  
155 (TCIPP), and tri-phenyl phosphate (TPHP)), as well as 2 isomers of DP (*syn*- and *anti*) (Iqbal et al.,  
156 2017). The authors reported human inhalation exposure to OPFRs to be the highest (2334 ng/kg  
157 bw/day), with the average daily dose of TPHP, TEHP, TnBP, TCEP, EHDPP, TCIPP, and TDCIPP  
158 being 1042 ng/kg bw/day, 308 ng/kg bw/day, 301 ng/kg bw/day, 230 ng/kg bw/day, 173 ng/kg  
159 bw/day, 162 ng/kg bw/day, and 118 ng/kg bw/day, respectively (Iqbal et al., 2017). This was  
160 followed by PBDEs (19.1 ng/kg bw/day),  $\Sigma$ NBFRs (13.7 ng/kg bw/day), and DP (5.42 ng/kg bw/day)  
161 (Iqbal et al., 2017). The calculated exposure doses were much lower than the reference doses  
162 suggested in previous literature for some of the target compounds (Table 1), indicating little health  
163 risk caused by inhalation exposure. However, accurate assessment of risk is difficult since no  
164 reference doses for mixtures of FRs (HFRs and OPFRs) are available, and the elevated exposures  
165 ranging between 2876 and 12087 ng  $\Sigma$ FRs/kg bw/day are notable.

166

167 In comparison, human inhalation exposure to HFRs and OPFRs in Guiyu and Qingyuan, two major  
168 e-waste recycling zones in China, was much lower (Luo et al., 2016; Wu et al., 2016a). In 2012,  
169 samples of atmospheric particles were collected in Qingyuan and analysed for 12 OPFRs (2 isomers  
170 of TnBP, TCEP, TCIPP, TDCIPP, TPHP, tris(2-butoxyethyl) phosphate (TBOEP), EHDPP, TEHP,  
171 and 3 isomers of tris(2,4,6-trimethoxyphenyl)phosphine (TMPP)) (Luo et al., 2016). Of the 12  
172 OPFRs, TEHP, and the 3 isomers of TMPP were not detected in any sample, and the average total  
173 concentration of the remaining 8 OPFRs was  $130 \pm 130$  ng/m<sup>3</sup>. This indicates a mean daily inhalation  
174 dose of  $12.1 \pm 4.1$  ng  $\Sigma$ OPFRs/kg bw/day for an average adult (TBOEP:  $6.8 \pm 4.1$  ng/kg bw/day, TnBP:  
175  $2.85 \pm 0.59$  ng/kg bw/day, TIBP:  $0.87 \pm 0.60$  ng/kg bw/day, TCIPP:  $0.64 \pm 0.28$  ng/kg bw/day, TCEP:  
176  $0.33 \pm 0.20$  ng/kg bw/day, TDCIPP:  $0.25 \pm 0.12$  ng/kg bw/day, TPHP:  $0.18 \pm 0.06$  ng/kg bw/day,  
177 EHDPP:  $0.172 \pm 0.049$  ng/kg bw/day, respectively) (Luo et al., 2016). In Guiyu, daily inhalation  
178 doses of BDE-47 and -99 were estimated to be 0.55 ng/kg bw/day and 0.33 ng/kg bw/day for adults,  
179 and 1.77 ng/kg bw/day and 1.07 ng/kg bw/day for children, respectively (Wu et al., 2016a). Given  
180 that this was much lower than the health based limit values (HBLVs) presented in previous studies



181 (Table 1), it is reasonable to conclude that human inhalation exposure to OPFRs and PBDEs in  
182 Qingyuan and Guiyu presented a low health risk.

183

## 184 **4.2 Human ingestion exposure to HFRs and OPFRs arising from informal e-waste activities**

### 185 **4.2.1 Dust ingestion**

186 Because of their relatively high octanol-air ( $K_{OA}$ ) and octanol-water partition coefficients ( $K_{OW}$ ),  
187 many HFRs like high brominated PBDEs and NBFs are likely to accumulate in atmospheric  
188 particles and body lipids (Ji et al., 2017; Jiang et al., 2019; Luo et al., 2016; Ma et al., 2017a, 2017b;  
189 Zheng et al., 2015a). An increasing number of studies have identified high concentrations of various  
190 HFRs and OPFRs in indoor dust and foodstuffs, indicating that human exposure to HFRs and OPFRs  
191 through indoor dust and food ingestion is non-negligible (Anh et al., 2017; Huang et al., 2018; Zeng  
192 et al., 2016, 2018; Zheng et al., 2015b, 2016). For instance, Zheng et al. (2015b) determined  
193 concentrations of 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209), 4 NBFs  
194 (BEH-TBP, TBB, BTBPE, and DBDPE), 8 OPFRs (TEHP, TnBP, TCEP, TBOEP, TPHP, EHDPP,  
195 TDCIPP, and TCIPP), and 2 isomers of DP (*syn*- and *anti*-) in indoor dust samples from some of the  
196 largest e-waste dismantling and recycling sites in China (Longtang, Dali, and Guiyu), and calculated  
197 human exposure to these contaminants through indoor dust ingestion. Assuming average dust  
198 ingestion rates (20 mg/day for adults and 50 mg/day for children), and average FR concentrations  
199 in dust; estimated daily intakes (EDIs) of PBDEs, NBFs, OPFRs, and DPs for adults were: 1.11-  
200 24.1 ng/kg bw/day, 0.73-20.3 ng/kg bw/day, 1.36-23.5 ng/kg bw/day, and 0.08-1.73 ng/kg bw/day,  
201 respectively, with the corresponding values for children 16-352 ng/kg bw/day, 11-296 ng/kg bw/day,  
202 20-343 ng/kg bw/day, and 1.18-25.3 ng/kg bw/day, respectively (Zheng et al., 2015b). The highest  
203 EDI values (assuming high ingestion of dust (50 mg/day for adults and 200 mg/day for children)  
204 contaminated at the 95<sup>th</sup> percentile concentration) for PBDEs, NBFs, OPFRs, and DPs were: 168  
205 ng/kg bw/day, 165 ng/kg bw/day, 226 ng/kg bw/day, and 12.8 ng/kg bw/day for adults, and 3915  
206 ng/kg bw/day, 3844 ng/kg bw/day, 5282 ng/kg bw/day, and 298 ng/kg bw/day for children,  
207 respectively (Zheng et al., 2015b). Although the calculated exposure doses were lower than the  
208 reference doses suggested in previous studies (Table 1), the authors concluded that children had an  
209 EDI of HFRs and OPFRs, that was 1 to 2 orders of magnitude higher than adults. Similar results  
210 were reported in Vietnam (Anh et al., 2017) and Thailand (Muenhor et al., 2017, 2018).

211

212 Anh et al. (2017) measured concentrations of 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -  
213 154, -183, and -209) in samples of home dust and fish from Bui Dau village, a major e-waste  
214 recycling site in Vietnam. Total PBDE concentrations in home dust were between 250-8650 ng/g,  
215 with BDE-209 being the dominant congener (Anh et al., 2017). It is estimated that the EDI via dust  
216 ingestion for adults and children was 0.71-2.47 ng/kg bw/day and 1.04-36.0 ng/kg bw/day under a  
217 median dust ingestion scenario (20 mg/day for adults and 50 mg/day for children), and 0.18-6.19  
218 ng/kg bw/day and 4.17-144 ng/kg bw/day under a high-end dust ingestion scenario (50 mg/day for  
219 adults and 200 mg/day for children), respectively (applying an average body weight of 70 kg for  
220 adults and 12 kg for children) (Anh et al., 2017). Specifically, it is notable that total PBDE  
221 concentrations in home dust and the EDI reported by Anh et al. (2017) were similar to those reported  
222 in a previous study conducted in the same area (Tue et al., 2013), indicating that PBDE  
223 contamination in this area remained at the same level during the 7-year period (from 2008 to 2015).  
224 Furthermore, the substantial increase in PBDE and NBFs concentrations in indoor dust in Qingyuan,  
225 China from 2007 to 2013/2014 (Table 2) indicates that e-waste recycling workers and residents  
226 became increasingly exposed to BFRs through dust ingestion over this period (He et al., 2017; Wang  
227 et al., 2010; Zheng et al., 2015b). This is an interesting situation, seen from the perspective of other  
228 areas where downward trends of PBDE contaminations have been reported (Harrad, 2015; Ma et  
229 al., 2017b; Yu et al., 2016).

230

#### 231 **4.2.2 Dietary intake**

232 In addition to dust ingestion, food intake could also be an important exposure pathway of e-waste  
233 recyclers and local residents to HFRs (Anh et al., 2017; Huang et al., 2018; Labunska et al., 2014,  
234 2015; Tao et al., 2016; Wu et al., 2019; Zeng et al., 2016, 2018). In the same study conducted by  
235 Anh et al. (2017), the EDI of 8 PBDE congeners through fish consumption (fish samples were  
236 manually collected from ponds and canals located within the e-waste recycling area) for adults and  
237 children in Bui Dau village, Vietnam was 0.72-46.4 ng/kg bw/day and 0.89-57.0 ng/kg bw/day,  
238 respectively. In addition, the EDI of 5 NBFs (pentabromobenzene (PBBz), HBB, BTBPE, BEH-  
239 TBP, and DBDPE), 2 DP isomers, and 3 HBCDD isomers through consumption of various  
240 foodstuffs was 36 ng/kg bw/day, 133 ng/kg bw/day, and 480 ng/kg bw/day for adults, and was 65

241 ng/kg bw/day, 350 ng/kg bw/day, and 1500 ng/kg bw/day for children, respectively, in Bui Dau  
242 village, Vietnam (Tao et al., 2016). It is notable that the EDI of  $\sum_3$ HBCDDs for adults and children  
243 was 2.4 times and 7.5 times higher than the reference dose (200 ng/kg bw/day), respectively,  
244 indicating potential health risk caused by dietary exposure to HBCDDs for local residents,  
245 especially for children. In comparison, while human dietary exposure to NBFs in informal e-waste  
246 dismantling and recycling sites in China (Huang et al., 2018; Labunska et al., 2014, 2015) was  
247 comparable to that in Vietnam; much higher human exposure to PBDEs (1 to 2 orders of magnitude  
248 higher) and much lower human exposure to HBCDDs (1 to 2 orders of magnitude lower) was  
249 observed in China compared to Vietnam (Huang et al., 2018; Labunska et al., 2014, 2015; Wu et  
250 al., 2019; Zeng et al., 2016). Furthermore, higher EDIs via food intake were identified for various  
251 HFRs in informal e-waste dismantling and recycling sites in China between 2013 and 2016 (Tables  
252 S1, S2, S3, and S5, Supplementary Material), especially for DPs, DBDPE, and CPs (Huang et al.,  
253 2018; Zeng et al., 2018), indicating potentially greater health concerns arising from exposure to  
254 HFRs in these regions.

255

### 256 **4.3 Human dermal uptake of HFRs and OPFRs arising from informal e-waste activities**

257 There is some debate about the contribution of dermal absorption to human external exposure to  
258 HFRs and OPFRs. While some researchers state exposure risk through dermal contact is almost  
259 negligible (Wu et al., 2016b), Wu et al. (2016a) suggest that the contribution of dermal absorption  
260 to human external exposure to HFRs and OPFRs has been underestimated because previous  
261 assessments of dermal exposure have addressed only inadvertent contact with contaminated dust or  
262 soil and overlooked dermal absorption of both particulate and gaseous contaminants through air-to-  
263 skin transfer, as well as from direct skin contact with e-waste articles. Most studies to date have  
264 focused on human exposure to HFRs and OPFRs through inhalation and ingestion (Anh et al., 2017;  
265 Huang et al., 2018; Iqbal et al., 2017; Luo et al., 2016; Zeng et al., 2016; Zheng et al., 2015b), with  
266 some consideration of exposure via dermal absorption limited. Afafe and Martincigh (2015)  
267 estimated dermal absorption from dust of 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -154,  
268 -183, and -209) for e-waste dismantling and recycling workers from Durban, South Africa. Mean  
269 and high-end exposure estimates were 0.87 ng/kg bw/day and 3.40 ng/kg bw/day, respectively, when  
270 an equation adapted from the US Environmental Protection Agency was applied (Afafe and

271 Martincigh, 2015). The contribution of dermal absorption, defined as dermal absorption from dust  
272 / (dust ingestion + dermal absorption from dust), was 40% under a mean dust ingestion scenario (20  
273 mg/day for adults), and 20% under a high-end dust ingestion scenario (50 mg/day for adults).  
274 However, this was likely underestimated since other dermal pathways such as air-to-skin transport,  
275 especially those of lower brominated congeners considered more easily absorbed by human skin  
276 due to their lower  $K_{OW}$  (Abdallah et al., 2015; Wu et al., 2016a), was not considered. In Guiyu, for  
277 example, the daily dermal intake of gaseous BDE-47 and BDE-99 through air-mediated transfer by  
278 adults was estimated to be 0.65 ng/kg bw/day and 0.61 ng/kg bw/day, respectively, exceeding  
279 exposure via inhalation of both gaseous and particle-bound BDE-47 (0.55 ng/kg bw/day) and BDE-  
280 99 (0.33 ng/kg bw/day) (Wu et al., 2016a). Similar results were also reported by Shen et al. (2019),  
281 who identified higher EDIs of NFRs, TBBP-A, and HBCDDs through dermal absorption rather  
282 than dust ingestion for recyclers, local adults, and local children in Qingyuan, China (the same  
283 equation adopted by Afafe and Martincigh (2015) was used in this study). Moreover, there appears  
284 to date to have been no consideration of exposure via direct dermal contact with e-waste articles,  
285 which may be a significant omission given recent data demonstrating the importance of dermal  
286 exposure via direct contact with FR-treated fabrics (Abdallah and Harrad, 2018). Overall, dermal  
287 absorption appears to be a potentially underestimated pathway of human external exposure to HFRs  
288 (and perhaps, OPFRs) for e-waste dismantling and recycling workers or residents inhabiting e-waste  
289 dismantling and recycling zones.

290

#### 291 **4.4 Variation in relative significance of different exposure pathways for HFRs and OPFRs**

292 PBDEs are the most frequently reported HFRs globally in terms of human external exposure through  
293 informal e-waste activities, especially through food and dust ingestion (Table S1, Supplementary  
294 Material). Specifically, the EDI of PBDEs through food intake has been suggested to exceed other  
295 exposure pathways such as inhalation (Wu et al., 2016a) and dust ingestion (Labunska et al., 2014).  
296 In Wenling and Luqiao, China, for instance, the median EDI of PBDEs through food intake was 5  
297 times and 2 times higher than that through dust ingestion for adults and children, respectively, with  
298 high-end exposure via food intake approximately 10 and 30 times higher than that through dust  
299 ingestion for adults and children, respectively (Labunska et al., 2014). Higher contribution to human  
300 exposure to PBDEs via food consumption than dust ingestion was also identified for adults in two

301 Vietnamese informal e-waste sites when a medium dust ingestion rate (20 mg/day for adults and 50  
302 mg/day for children) was applied (Anh et al., 2017). Moreover, for children, exposure through food  
303 consumption was found roughly equivalent to that through dust ingestion (Anh et al., 2017).  
304 However, the contribution of food intake to PBDE exposure was very likely underestimated by Anh  
305 et al. (2017), since only fish consumption was included in that study. Furthermore, it is notable that  
306 the relative contribution of different pathways varies for different PBDE congeners. In a study  
307 conducted by Labunska et al. (2014), the relative contribution of dust ingestion to PBDE exposure  
308 compared to that of food consumption (defined as  $EDI_{dust} / (EDI_{food} + EDI_{dust})$ , using median  
309 exposure scenarios) was 1.0%, 2.1%, 0.2%, 0.2%, and 30.5% for BDE-47, -99, -153, -154, and -  
310 209, respectively, for adults, and 2.3%, 4.3%, 0.5%, 0.5%, and 49% for BDE-47, -99, -153, -154,  
311 and -209, respectively, for children. Compared to lower brominated PBDE congeners, the  
312 contribution of dust ingestion to overall exposure to BDE-209 is much higher.

313

314 Food intake has also been reported as the dominant pathway of human exposure to NBFRs (DBDPE,  
315 BTBPE, etc), DPs, and HBCDD. In Bui Dau, Vietnam, the EDI for DBDPE, BTBPE, and 2 isomers  
316 of DP through food intake was 5 ng/kg bw/day, 31 ng/kg bw/day, and 133 ng/kg bw/day for adults,  
317 and 2.7 ng/kg bw/day, 61 ng/kg bw/day, and 350 ng/kg bw/day for children, respectively (Tao et al.,  
318 2016). This figure was 2 to 4 orders of magnitude higher than exposure via dermal absorption of  
319 soil and soil ingestion of DBDPE, BTBPE, and DPs in the same area (Someya et al., 2016). Similar  
320 results were also reported in Qingyuan, China, where the estimated dietary intake of DBDPE, DPs,  
321 as well as  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD were about 1 to 2 orders of magnitude higher than dust ingestion  
322 for both adults and children (He et al., 2017; Huang et al., 2018; Shen et al., 2019).

323

324 The main pathway of human exposure to OPFRs appears very different to that of BFRs and DPs,  
325 with inhalation identified as the most important exposure pathway in a recent study of an informal  
326 e-waste site in Pakistan. This study reported human exposure to OPFRs through inhalation to be 3  
327 orders of magnitude higher than via soil ingestion (Iqbal et al., 2017). This result was in agreement  
328 with studies conducted in Qingyuan, China. Specifically, human exposure to OPFRs through  
329 inhalation was estimated to be  $12.1 \pm 4.1$  ng/kg bw/day (mean  $\pm$  SD) for adults in Qingyuan, China,  
330 which exceeded the EDI via dust ingestion reported by He et al. (2015) (median: 7.02 ng/kg bw/day),

331 Guo et al. (2019a) (mean: 5.85 ng/kg bw/day), and Zheng et al. (2015b) (mean: 1.36-2.11 ng/kg  
332 bw/day) for adults. It is interesting that in both informal e-waste regions in Pakistan and China, non-  
333 chlorinated OPFRs comprised the majority of total OPFRs exposure via inhalation (Iqbal et al.,  
334 2017; Luo et al., 2016). This may be attributable to less extensive use of chlorinated OPFRs in  
335 electrical and electronic products. Another interesting observation is that, despite the low  
336 contribution of food intake to human exposure to OPFRs reported in informal e-waste sites to date,  
337 chlorinated OPFRs were more frequently detected in chicken eggs sourced from an informal e-waste  
338 site in China than non-chlorinated OPFRs (Zheng et al., 2016). This might be explained by the  
339 relatively longer half-life of chlorinated OPFRs (Ma et al., 2017a).

340

341 Very limited data on human exposure to CPs through informal e-waste activities are available. To  
342 the best of our knowledge, only 3 publications have reported human dietary exposure to CPs in  
343 informal e-waste sites in China (Yuan et al., 2017; Zeng et al., 2016, 2018), with an increasing trend  
344 of human dietary exposure identified in Longtang, China (Table S5, Supplementary Material).  
345 Specifically, the EDI of short-chain chlorinated paraffins (SCCPs, C<sub>10</sub>-C<sub>13</sub>) through chicken egg  
346 consumption increased by 4 times for adults and children between 2013 and 2016, and the EDI of  
347 median-chain chlorinated paraffins (MCCPs, C<sub>14</sub>-C<sub>17</sub>) also increased by nearly 30% (Zeng et al.,  
348 2018). No data were found about human exposure to CPs through inhalation, dermal contact, or dust  
349 ingestion.

350

## 351 **5 Human internal exposure to HFRs and OPFRs at informal e-waste handling sites**

352 In addition to assessments of external exposure, internal human exposure to HFRs and OPFRs has  
353 also been frequently examined in various informal e-waste dismantling and recycling areas, with  
354 human blood and serum (Chen et al., 2015; Eguchi et al., 2015; Guo et al., 2020; Lv et al., 2015;  
355 Schecter et al., 2018), human hair (Chen et al., 2015; Liang et al., 2016; Qiao et al., 2019), human  
356 milk (Awasthi et al., 2016; Li et al., 2017; Shi et al., 2018), and human urine (Bai et al., 2019; Lu et  
357 al., 2017; Shi et al., 2019; Yan et al., 2018) being the most commonly used biomarkers. An overview  
358 of recent studies (i.e. those published 2015-2020) of internal human exposure to HFRs and OPFRs  
359 through informal e-waste handling activities is provided as Tables S6 and S7 (Supplementary  
360 Material).

361

## 362 **5.1 Human blood and serum**

363 Human blood and serum are frequently used indicators of human internal exposure to HFRs, since  
364 HFRs are likely transported into multiple human organs and tissues through blood circulation (Chen  
365 et al., 2015; Eguchi et al., 2015; Kuo et al., 2019; Lv et al., 2015; Schechter et al., 2018). In a study  
366 conducted in Baoai, an e-waste treatment site in northern Vietnam, serum samples from 40 female  
367 e-waste recyclers were collected to assess their internal exposure to various contaminants including  
368 7 PBDE congeners (BDE-47, -99, -100, -153, -154, -183, and -209) (Schechter et al., 2018). Among  
369 the analyzed PBDE congeners, BDE-209 (median: 73.3 ng/g lipid; 95% confidence interval (CI):  
370 32.4-138.2 ng/g lipid) was dominant, followed by BDE-153 (median: 13.0 ng/g lipid; 95% CI: 10.2–  
371 18.8 ng/g lipid) and BDE-183 (median: 7.3 ng/g lipid; 95% CI: 6.1-10.0 ng/g lipid) (Schechter et al.,  
372 2018). PBDE concentrations in serum from e-waste recyclers were 1 to 2 orders of magnitude higher  
373 than those in non-recyclers whose PBDE concentrations in serum were frequently below limits of  
374 detection (Schechter et al., 2018), indicating high occupational exposure of e-waste recyclers to  
375 PBDEs. Similar conclusions have been reached in other studies (Eguchi et al., 2015; Guo et al.,  
376 2019b; Liang et al., 2016). For instance, in an e-waste recycling site in Wenling, China and an urban  
377 area where no e-waste recycling activities were undertaken, 14 PBDE congeners (BDE-17, -28, -  
378 47, -66, -99, -100, -153, -154, -183, -203, -206, -207, -208, and -209) and DBDPE were determined  
379 in serum samples taken from e-waste recyclers, non-occupationally-exposed residents of the e-waste  
380 site, and urban residents (Liang et al., 2016). Mean concentrations of total PBDEs and DBDPE in  
381 serum from e-waste recyclers were 656 ng/g lipid (range: 167-2530 ng/g lipid) and 125 ng/g lipid  
382 (range: 26.7-439 ng/g lipid), respectively. These concentrations exceeded significantly those  
383 detected in serum of non-occupationally-exposed residents (PBDEs: 123 ng/g lipid, range: 45.9-243  
384 ng/g lipid; DBDPE: 56.1 ng/g lipid, range: 4.20-127 ng/g lipid), and urban residents (PBDEs: 24.6  
385 ng/g lipid, range: 10.1-48.2 ng/g lipid; DBDPE: 13.8 ng/g lipid, range: nd-33.2 ng/g lipid) (Liang  
386 et al., 2016). Moreover, Zheng et al. (2017b) identified higher PBDE concentrations in serum taken  
387 from donors who lived in an e-waste site for over 20 years than those who lived there for less than  
388 3 years (Table S6, Supplementary Material), indicating that PBDE concentrations were likely to  
389 increase with increasing duration of residence in informal e-waste sites.

390

391 It is interesting to note that females have been reported to display greater HFR contamination in  
392 serum than males in e-waste-impacted areas. Zheng et al. (2017a) reported that total PBDE  
393 concentrations in female serum samples (mean: 2309 ng/g lipid; range: 206-35902 ng/g lipid) were  
394 significantly higher ( $p < 0.05$ ) than those in male serum samples (mean: 690 ng/g lipid; range: 105-  
395 1806 ng/g lipid) in Qingyuan, China. Specifically, females were found to have significantly higher  
396 ( $p < 0.05$ ) serum concentrations of congeners associated with the Deca-BDE commercial  
397 formulation (sum of BDE-196, -197, -202, -203, -206, -207, -208, and -209; mean: 1896 ng/g lipid;  
398 range: 100-34482 ng/g lipid) than did males (mean: 509 ng/g lipid; range: 64.5-1494 ng/g lipid).  
399 Moreover, females displayed higher serum concentrations of congeners associated with the Penta-  
400 BDE formulation (sum of BDE-28, -47, -66, -85, -99, and -100; mean: 125 ng/g lipid; range: 44.6-  
401 853 ng/g lipid) and Octa-BDEs (sum of BDE-153, -154, and -183; mean: 287 ng/g lipid; range:  
402 4.58-2667 ng/g lipid) than males (Penta-BDEs: mean: 104 ng/g lipid, range: 17.1-242 ng/g lipid;  
403 Octa-BDEs: mean: 77.3 ng/g lipid, range: 8.02-283 ng/g lipid), but the difference was not significant  
404 in this instance ( $p > 0.05$ ) (Zheng et al., 2017a). Similar findings were also reported by (Chen et al.,  
405 2015), who found that concentrations of DPs in female serum (median: 230 ng/g lipid, range: 37-  
406 1400 ng/g lipid) were slightly higher than that in male serum (median: 180 ng/g lipid, range: 22-  
407 510 ng/g lipid). Unfortunately, no explanation for this difference was provided in either publication,  
408 and the cause of this gender disparity is unclear.

409

## 410 **5.2 Human hair**

411 As a non-invasive sampling matrix, human hair has been frequently used to measure human internal  
412 exposure to HFRs and OPFRs (Chen et al., 2015; Liang et al., 2016; Qiao et al., 2019). A recent  
413 study recruited 31 female e-waste dismantling workers from an e-waste recycling site in South  
414 China, and measured concentrations of a wide variety of FRs (i.e., 8 PBDE congeners (BDE-28, -  
415 47, -99, -100, -153, -154, -183, and -209), 2 NBFRs (DBDPE and BTBPE), *syn*- and *anti*-DP, and  
416 13 OPFRs (TnBP, TCEP, TDCIPP, TBOEP, TEHP, TPHP, EHDPP, triisopropyl phosphate (TiPrP),  
417 tri-*n*-propyl phosphate (TPrP), 3 isomers of tricresyl phosphate (TCP), and TCIPP)) in hair samples  
418 (Qiao et al., 2019). The mean concentration of  $\Sigma$ OPFRs (the most abundant FRs) was 431 ng/g  
419 (range: 189-1558 ng/g), with TBOEP, TCIPP, TEHP, and TPHP the dominant congeners, accounting  
420 for 24.2%, 18.7%, 15.9%, and 11.1% of  $\Sigma$ OPFR concentrations, respectively (Qiao et al., 2019).



421 For  $\Sigma$ PBDEs, the mean concentration was 271 ng/g (range: 49.8-2104 ng/g), with BDE-209  
422 dominant (accounting for 92.8% of  $\Sigma$ PBDEs) (Qiao et al., 2019). For  $\Sigma$ DPs and  $\Sigma$ NBFRs, mean  
423 concentrations were 61.3 ng/g and 211 ng/g, with ranges of 1.64-360 ng/g and 16.4-991 ng/g,  
424 respectively (Qiao et al., 2019). Notably, the study also identified an increasing trend in  
425 concentrations of PBDEs, DPs, and NBFRs from 2009 to 2015 (Qiao et al., 2019). Another study  
426 conducted by Liang et al. (2016) also indicated high concentrations of PBDEs and DBDPE in human  
427 hair, noting that exposure of e-waste recyclers (mean value: 292.9 ng/g for PBDEs and 82.5 ng/g  
428 for DBDPE) was significantly higher than that of non-occupationally exposed residents (mean value:  
429 55.8 ng/g for PBDEs and 29.4 ng/g for DBDPE) and urban residents (mean value: 12.9 ng/g for  
430 PBDEs and 10.9 ng/g for DBDPE). Significant correlations were reported between concentrations  
431 of PBDEs and DBDPE in serum and hair, thereby indicating hair to be a useful matrix for  
432 biomonitoring PBDEs and DBDPE exposure in humans (Liang et al., 2016). Similar conclusions  
433 were also drawn regarding DPs in human hair, and as highlighted above for serum, it is notable that  
434 female hair (median: 200 ng/g; range: 17-1100 ng/g) was more contaminated with DPs than male  
435 hair (median: 19 ng/g; range: 6.3-150 ng/g). However, in this instance the gender difference was  
436 attributed to the longer external exposure time of female hair (Chen et al., 2015).

437

### 438 **5.3 Human milk**

439 Concentrations of HFRs in human milk can not only reflect internal exposure of female adults but  
440 also dietary exposure of nursing infants (Shi et al., 2018; Tang and Zhai, 2017). Concentrations of  
441 HFRs in human milk and the associated implications for human exposure, especially for infants,  
442 have previously been reviewed for China and Africa (Shi et al., 2018; Orisakwe et al., 2019).  
443 Specifically, a recent study conducted in an e-waste handling area in Wenling, China, collected 25  
444 human milk samples from mothers who had lived there for over 20 years (defined as the R<sub>20</sub> group)  
445 and 21 human milk samples from mothers who had resided there for no more than 3 years (defined  
446 as the R<sub>3</sub> group), and determined concentrations of 8 PBDE congeners (BDE-28, -47, -99, -100, -  
447 153, -154, -183, and -209) in these samples (Li et al., 2017). It found that  $\Sigma$ PBDE concentrations in  
448 the R<sub>20</sub> group (mean: 25.7±20.0 ng/g lipid; range: 7.89-90.6 ng/g lipid) exceeded significantly those  
449 in the R<sub>3</sub> group (mean: 6.68±5.61 ng/g lipid; range: 1.87-22.0 ng/g lipid) (Li et al., 2017). The two  
450 groups had similar congener profiles of PBDEs, with BDE-209 and -153 being the most abundant

451 congeners in both groups (Li et al., 2017). Furthermore, the EDI of PBDEs for infants in the R<sub>20</sub>  
452 group was in the range 15.8-243 ng/kg bw/day (median: 45.3 ng/kg bw/day), while in the R<sub>3</sub> group  
453 the range was 5.43-43.0 ng/kg bw/day (median: 11.4 ng/kg bw/day) (Li et al., 2017). Although this  
454 was much lower than the reference doses of the United States Environmental Protection Agency  
455 (2017), the maximum EDI of BDE-47 (76.9 ng/kg bw/day ) and BDE-153 (98.9 ng/kg bw/day) (Li  
456 et al., 2017) for nursing infants in the R<sub>20</sub> group approached the corresponding reference doses  
457 (BDE-47: 100 ng/kg bw/day; BDE-153: 200 ng/kg bw/day).

458

#### 459 **5.4 Human urine**

460 OPFR metabolites have been measured in human urine samples from e-waste recyclers, including  
461 those of both chlorinated and non-chlorinated OPFRs (Bai et al., 2019; Lu et al., 2017; Shi et al.,  
462 2019; Yan et al., 2018). For instance, Lu et al. (2017) found urinary concentrations of chlorinated  
463 and non-chlorinated OPFR metabolites in an e-waste impacted area in Qingyuan, China (mean: 4.0  
464 ng/mL for chlorinated OPFR metabolites and 2.3 ng/mL for non-chlorinated OPFR metabolites)  
465 exceeded significantly those in subjects from a rural area (mean: 2.1 ng/mL for chlorinated OPFR  
466 metabolites and 0.74 ng/mL for non-chlorinated OPFR metabolites), suggesting substantial human  
467 exposure to OPFRs through e-waste handling activities. Negative correlations were determined  
468 between age and urinary concentrations of each OPFR metabolite, and for bis(2-chloroethyl)  
469 phosphate (BCEP), bis(1-chloro-2-propyl) phosphate (BCIPP), bis(1,3-dichloro-2-propyl)  
470 phosphate (BDCIPP), and diphenyl phosphate (DPHP), the negative correlations were significant  
471 (Lu et al., 2017). These are similar to results reported by Yan et al. (2018) of significantly higher  
472 concentrations of BCEP in urine samples from the 21-30 age group than the older age groups. This  
473 might indicate higher exposure of younger people. However, a survey conducted by Shi et al. (2019)  
474 generated different results, specifically that concentrations of OPFR metabolites in urine samples  
475 from children were significantly lower than for adults who have been participating in e-waste  
476 treatment for years. This discrepancy could be explained by the different sampling methodologies  
477 adopted. In particular, only residents (and no e-waste workers) were sampled by Lu et al. (2017),  
478 while only e-waste recyclers were sampled by Yan et al. (2018), which means the difference between  
479 occupational and non-occupational exposure was not considered. However, the results reported by  
480 Shi et al. (2019) could be explained by high occupational exposure of adults since the children were

481 not involved in e-waste recycling activities. No data was found about concentrations of HFRs (or  
482 metabolites) other than chlorinated OPFRs in urine samples from e-waste dismantling and recycling  
483 areas.

484

### 485 **5.5 Other human matrices**

486 Xu et al. (2015) compared 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209)  
487 in human placental tissue samples from an e-waste recycling site (Guiyu, China) and a reference  
488 area (Haojiang, China). The study found that PBDE concentrations were much higher in samples  
489 from the e-waste recycling site (mean: 61.39±85.42 ng/g/lipid; range: 0.89-516.97 ng/g lipid) than  
490 those from the reference area (mean: 13.03±195.46 ng/g/lipid; range: 0.66-195.46 ng/g/lipid). This  
491 could indicate higher exposure not only for mothers but also for fetuses in the e-waste recycling site  
492 since partitioning of PBDEs from mothers to fetuses is considered as an important pathway of  
493 exposure of fetuses to PBDEs (Xu et al., 2015; Zheng et al., 2017b). High concentrations of PBDEs  
494 were also detected in human nails, abdominal subcutaneous adipose tissue, and umbilical cord tissue  
495 from residents inhabiting informal e-waste sites in China (Li et al., 2018; Lv et al., 2015; Meng et  
496 al., 2020). Apart from PBDEs, some OPFR metabolites (i.e., dibutyl phosphate (DBP) and DPHP)  
497 were also frequently detected in amniotic fluid samples (Bai et al., 2019), indicating fetal exposure  
498 to OPFRs.

499

### 500 **5.6 Potential health risks originated from human internal exposure to HFRs and OPFRs**

501 Health risk assessments of human exposure to HFRs and OPFRs have previously been reviewed in  
502 Africa (Asante et al., 2019; Orisakwe et al., 2019), India (Awasthi et al., 2016), China (Awasthi et  
503 al., 2018; Shi et al., 2018), and other regions on a global scale (Akram et al., 2019; Bakhiyi et al.,  
504 2018). Specifically, in informal e-waste sites, human exposure to TCIPP, TCEP, TNBP, and TPHP  
505 were correlated with elevated DNA oxidative stress in e-waste sites, as the urinary concentrations  
506 of BCIPP, BCEP, DBP, and DPHP were significantly increased as the concentration of 8-hydroxy-  
507 2'-deoxyguanosine (8-OHdG), a marker of DNA oxidative stress, increased (Lu et al., 2017).  
508 Moreover, PBDEs and NBRs are reported thyroid hormone (TH) disruptors (Eguchi et al., 2015;  
509 Guo et al., 2019b; Zheng et al., 2017a, 2017b). They have strong binding affinity to thyroid-  
510 stimulating hormone (TSH), thyroglobulin, thyroxine-binding globulin (TBG), TH receptor  $\alpha$  (TR $\alpha$ ),

511 and iodothyronine deiodinase I (ID1), and therefore could disrupt TH-regulated proteins and gene  
512 expression (Guo et al., 2019b). It is also suggested that PBDEs, NBFRs (e.g., DBDPE, BTBPE,  
513 BEH-TBP, etc.), and DP could exert similar disrupting effects on female follicle-stimulating  
514 hormone (FSH) and male testosterone, with NBFRs showing stronger disrupting effects on human  
515 sex hormones than do PBDEs (Guo et al., 2018). Furthermore, PBDEs were found to have adverse  
516 effects on human semen quality measured by sperm concentration and count, sperm progressive  
517 motility, and sperm viability (Yu et al., 2018).

518

519 Specifically, health effects of HFR exposure on fetuses and infants are of particular concern due to  
520 their substantially weaker resistance and immunity (Bai et al., 2019; Li et al., 2018; Xu et al., 2015;  
521 Zheng et al., 2017b). Potential health risks for pregnant women and fetuses arising from OPFR  
522 (especially TPHP and TnBP) exposure in an e-waste site (Qingyuan, China) were implied by Bai et  
523 al. (2019), who determined high concentrations of OPFRs in paired amniotic fluid as well as in  
524 maternal urine. Meanwhile, Xu et al. (2015), along with Li et al. (2018) reported that high prenatal  
525 exposure to PBDEs in e-waste recycling areas may lead to adverse physiological development in  
526 fetuses, in terms of reduced body-mass index, Apgar 1 score, and head circumference.

527

## 528 **6 Conclusions**

529 The evidence reviewed in this study indicates 3 main pathways of human external exposure to HFRs  
530 and OPFRs in informal e-waste handling sites, i.e., inhalation, ingestion of dust and food, and  
531 dermal absorption. Current evidence suggests EDIs of OPFRs via inhalation exceed those via food  
532 intake and dust ingestion, and thus inhalation could be the dominant pathway of human exposure to  
533 OPFRs in informal e-waste sites; while for PBDEs, NBFRs, and DPs, food consumption and indoor  
534 dust ingestion are likely more important contributors. An important factor emerging from our review  
535 is that human exposure to HFRs and OPFRs through dermal absorption is insufficiently well-  
536 understood and may well be underestimated as pathways such as dermal absorption of both  
537 particulate and gaseous contaminants through air-to-skin transfer, as well as from direct skin contact  
538 with e-waste articles, have been largely overlooked. Children, infants, and fetuses, as well as e-  
539 waste recycling workers were found to experience higher HFR and OPFR exposure than did non-  
540 occupationally-exposed adults inhabiting e-waste handling zones, or those living in non-e-waste-

541 impacted locations. Gender differences in human internal exposure to HFRs in informal e-waste  
542 locations were reported in some studies with higher HFR concentrations in serum and hair observed  
543 in females compared to males. The cause of these higher HFR concentrations in females is unclear,  
544 especially in serum.

545

546 Temporal changes in HFR concentrations in environmental media and humans have also been  
547 identified from previous studies. The evidence reviewed in this study shows increasing levels of  
548 HFRs in indoor dust, foodstuffs, and human hair with increasing duration of e-waste handling  
549 activity at a given site. Furthermore, PBDE concentrations in human serum and breast milk were  
550 likely to increase with increasing duration of residence in informal e-waste sites.

551

552 Most studies about human exposure to HFRs and OPFRs through informal e-waste handling  
553 activities were conducted in China, while studies in other countries or regions were limited. As the  
554 import of wastes from foreign countries was banned in China at the end of 2018, other low- and  
555 moderate-income countries are likely to receive more waste, and greater environmental and health  
556 effects could be caused by the improper treatment of this waste. More attention should therefore be  
557 paid to problems associated with informal e-waste treatment in low- and moderate-income countries  
558 in the future.

559

560 Based on the present review, we recommend that the following research gaps should be addressed  
561 urgently. Firstly, very little is known about HFR and OPFR contamination of the environment and  
562 humans arising from informal e-waste activities outside China, especially in Africa where such  
563 activities appear to be growing substantially. Secondly, more detailed consideration of dermal  
564 absorption as a pathway of human exposure to HFRs and OPFRs to those working in and inhabiting  
565 e-waste handling areas is required. Thirdly, data on human dietary intake of OPFRs of residents of  
566 informal e-waste sites is a priority for investigation. Moreover, the higher body burdens of HFRs in  
567 females associated with informal e-waste recycling compared to males that have been reported  
568 require further detailed study, both to verify such findings and to elucidate their cause(s). Finally,  
569 very little is understood about human exposure to CPs via all potential pathways as a result of  
570 informal e-waste handling, and research to better understand the magnitude of such exposure and

571 the pathways via which it occurs is recommended.

572

573 **Declarations of interest**

574 The authors declare no conflict of interest.

575

576 **Acknowledgments**

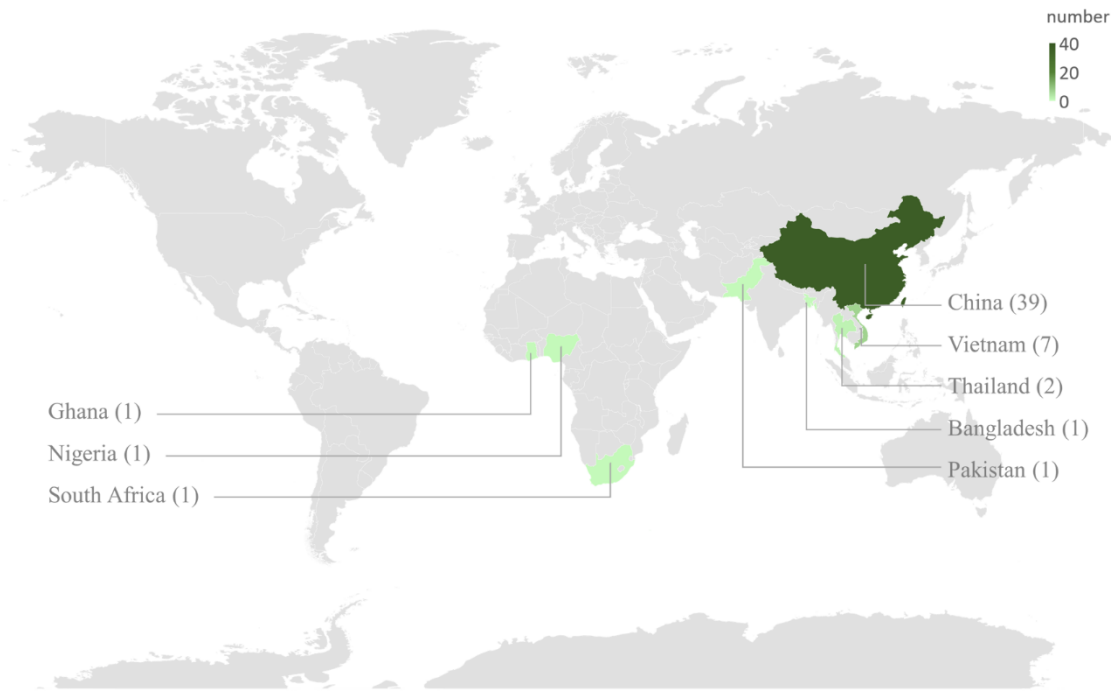
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579

580 **Tables and figures**

581

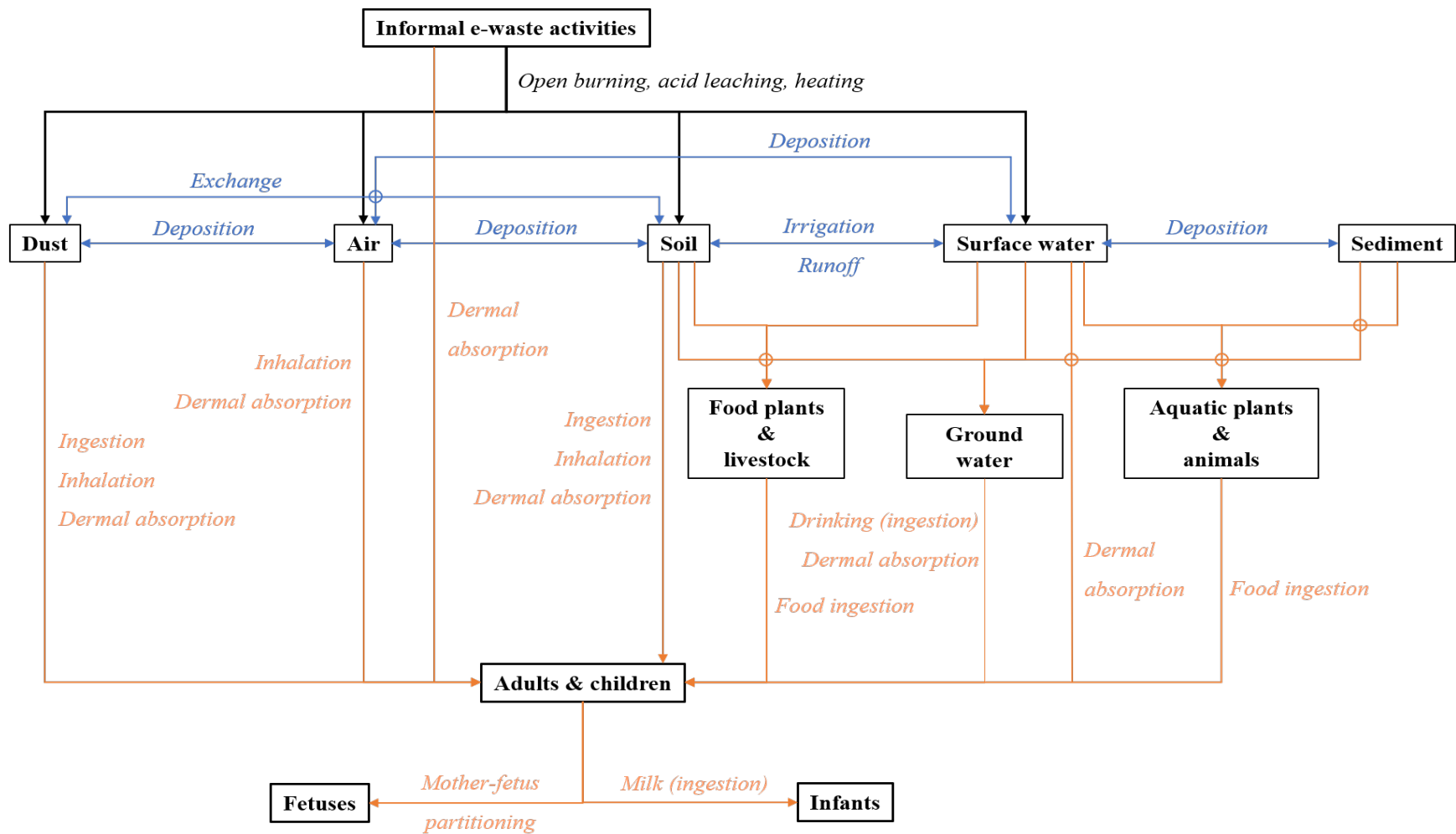


582

583 **Fig. 1: Global distribution of recent research studies reporting human exposure to HFRs and**

584

**OPFRs through informal e-waste handling activities (2015-2020)**



585

586

Fig. 2: Transfer of HFRs and OPFRs between various environmental media and associated human exposure pathways (Leung, 2019)



587

**Table 1. Reference doses (RfD) values (ng/kg bw/day) for some HFRs and OPFRs**

<b>Compound</b>	<b>RfD</b>	<b>Compound</b>	<b>RfD</b>
TnBP	24000	BDE-209	7000
TCEP	22000	Octa-BDE	3000
TCIPP	80000	Penta-BDE	2000
TBOEP	15000	BDE-47	100
TPHP	70000	BDE-99	100
TDCIPP	15000	BDE-153	200
TCP	13000	DP	5000000 <sup>(1)</sup>
BTBPE	243000	DP	2000000 <sup>(2)</sup>
TBB	20000	DP	10000 <sup>(3)</sup>
BEH-TBP	20000	$\Sigma$ HBCDDs	200
DBDPE	333333	TBBP-A	600000

588 Notes: (1) chronic oral RfD; (2) dermal RfD; (3) inhalation RfDs

589 Source: Ali et al. (2012), except United States Environmental Protection Agency (2017) for PBDEs, Wang et al.

590 (2013) for DP, and Besis et al. (2017) for HBCDD and TBBP-A.

591

592

593 **Table 2. Concentrations of BDE-209, DBDPE, and BTBPE in indoor dust collected from**  
 594 **Qingyuan, China (ng/g)**

sampling period	BDE-209		DBDPE		BTBPE		references
	median	range	median	range	median	range	
2007	988	105-140000	63.1	13.5-1144	20	n.d. <sup>(1)</sup> -998	(Wang et al., 2010)
2013	644-22500	146-195000	1160-26300	n.d.-181000	28-148	2.8-12700	(Zheng et al., 2015)
2013-2014	23800	8530-152000	2720	669-15000	n.a. <sup>(2)</sup>	n.a.	(He et al., 2017)

595 Notes: (1) n.d. = not detected; (2) n.a. = not available

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