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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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Abstract

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

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

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

1. Introduction

Electrical and electronic waste (e-waste), also called waste electrical and electronic equipment (WEEE), has become a global concern. In 2019, global generation of e-waste reached 53.6 million tonnes (Mt), and this figure is estimated to reach 74 Mt by 2030 and 120 Mt by 2050, respectively (Forti et al., 2020; World Economic Forum, 2019). Typically, household appliances like washing machines, telecommunications, and IT equipment including computers, and consumer articles like TVs, comprise the majority of e-waste generated globally (Akram et al., 2019). Due to environmental and economic considerations, much of the e-waste generated in high-income countries has and continues to be exported for handling in low- and moderate-income countries (Mihai et al., 2019; Pathak et al., 2017). Although measures outlined in the Basel Convention were designed to prohibit the export of hazardous wastes to low- and moderate-income countries, these are unlikely to be effective as some used electrical and electronic equipment is characterised as non-hazardous waste (Khan, 2016). Moreover, it was not until June 2019 when the EU clarified their determinations on the regulation of persistent organic pollutants (POPs), that many non-hazardous WEEE items would be reclassified as hazardous and become subject to the Hazardous Waste Regulations 2005 (Official Journal of the European Union, 2019). Moreover, in low- and moderate-income countries, approximately 90 % of e-waste disposal and recycling activities are undertaken by informal sector workers (Pathak et al., 2019). In India, for instance, 97 % of total e-waste generated is handled by informal recycling yards in major cities like Delhi, Mumbai, Hyderabad, and Bangalore (Rao et al., 2017).

This is concerning as various contaminants including halogenated flame retardants (HFRs) and organophosphate flame retardants (OPFRs) can enter environmental media through different activities reported to be undertaken at informal e-waste handling facilities. Informal e-waste activities refer to unlicensed or unregulated e-waste dismantling and recycling activities that are usually practiced by individuals and families using primitive techniques. These include: open burning, acid leaching, and heating (Leung, 2019). The risks posed by such emissions are compounded by the lack of effective personal protective equipment for use by those undertaking such work, as well as by absence of other measures designed to protect the environment and human health (Lundgren, 2012). HFRs and OPFRs have been widely used as additives at concentrations of up to 10-15 % by weight in the plastic housing of various electric and electronic products to ensure that these products meet fire safety regulations (Lassen and Havelund, 2006). However, from the mid-2000s onwards, concerns about the adverse environmental and human health impacts of HFRs like polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) would lead to restrictions and bans on their manufacture and use in new products in many jurisdictions, including their listing under the Stockholm Convention on POPs of the United Nations Environment Programme (UNEP) (Ge et al., 2020; Harrad, 2015; Huang et al., 2018; Ma et al., 2017b). As a result, there has been increased demand for OPFRs and other HFRs such as tetrabromobisphenol-A (TBBP-A), novel brominated flame retardants (NBFRs), dechlorane plus (DP), and chlorinated paraffins (CPs) (Chen et al., 2015; Ge et al., 2020; Gravel et al., 2020; He et al., 2017; Liu et al., 2016; Ma et al., 2017a; Zeng et al., 2018, 2020).

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

The present review aims to: 1) summarise current research into human external exposure (i.e., inhalation, ingestion of food, dust, and soil, and dermal absorption) and internal exposure (i.e., blood and serum, hair, breast milk, urine, and other human matrices) to HFRs and OPFRs through informal e-waste dismantling and recycling activities for local residents; 2) identify potential health risks to local residents in informal e-waste sites; and 3) highlight substantial research gaps that require urgent investigation.

2. Methods

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

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

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

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

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

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

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

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

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

In comparison, human inhalation exposure to HFRs and OPFRs in Guiyu and Qingyuan, two major e-waste recycling zones in China, was much lower (Luo et al., 2016; Wu et al., 2016a). In 2012, samples of atmospheric particles were collected in Qingyuan and analysed for 12 OPFRs (2 isomers of TnBP, TCEP, TCIPP, TDCIPP, TPHP, tris(2-butoxyethyl) phosphate (TBOEP), EHDPP, TEHP, and 3 isomers of tris(2,4,6-trimethoxyphenyl)phosphine (TMPP)) (Luo et al., 2016). Of the 12 OPFRs, TEHP, and the 3 isomers of TMPP were not detected in any sample, and the average total concentration of the remaining 8 OPFRs was 130±130 ng/m^3. This indicates a mean daily inhalation dose of 12.1±4.1 ng ΣOPFRs/kg bw/day for an average adult (TBOEP: 6.8±4.1 ng/kg bw/day, TnBP: 2.85±0.59 ng/kg bw/day, TIBP: 0.87±0.60 ng/kg bw/day, TCIPP: 0.64±0.28 ng/kg bw/day, TCEP: 0.33±0.20 ng/kg bw/day, TDCIPP: 0.25±0.12 ng/kg bw/day, TPHP: 0.18±0.06 ng/kg bw/day, EHDPP: 0.172±0.049 ng/kg bw/day, respectively) (Luo et al., 2016). In Guiyu, daily inhalation 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, and 1.77 ng/kg bw/day and 1.07 ng/kg bw/day for children, respectively (Wu et al., 2016a). Given that this was much lower than the health based limit values (HBLVs) presented in previous studies

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

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

4.2.1 Dust ingestion

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

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

4.2.2 Dietary intake

In addition to dust ingestion, food intake could also be an important exposure pathway of e-waste recyclers and local residents to HFRs (Anh et al., 2017; Huang et al., 2018; Labunska et al., 2014, 2015; Tao et al., 2016; Wu et al., 2019; Zeng et al., 2016, 2018). In the same study conducted by Anh et al. (2017), the EDI of 8 PBDE congeners through fish consumption (fish samples were manually collected from ponds and canals located within the e-waste recycling area) for adults and children in Bui Dau village, Vietnam was 0.72-46.4 ng/kg bw/day and 0.89-57.0 ng/kg bw/day, respectively. In addition, the EDI of 5 NBFRs (pentabromobenzene (PBBz), HBB, BTBPE, BEH-TBP, and DBDPE), 2 DP isomers, and 3 HBCDD isomers through consumption of various foodstuffs was 36 ng/kg bw/day, 133 ng/kg bw/day, and 480 ng/kg bw/day for adults, and was 65 ng/kg bw/day, 350 ng/kg bw/day, and 1500 ng/kg bw/day for children, respectively, in Bui Dau village, Vietnam (Tao et al., 2016). It is notable that the EDI of $\sum_3$HBCDDs for adults and children was 2.4 times and 7.5 times higher than the reference dose (200 ng/kg bw/day), respectively, indicating potential health risk caused by dietary exposure to HBCDDs for local residents, especially for children. In comparison, while human dietary exposure to NBFRs in informal e-waste dismantling and recycling sites in China (Huang et al., 2018; Labunska et al., 2014, 2015) was comparable to that in Vietnam; much higher human exposure to PBDEs (1 to 2 orders of magnitude higher) and much lower human exposure to HBCDDs (1 to 2 orders of magnitude lower) was observed in China compared to Vietnam (Huang et al., 2018; Labunska et al., 2014, 2015; Wu et al., 2019; Zeng et al., 2016). Furthermore, higher EDIs via food intake were identified for various HFRs in informal e-waste dismantling and recycling sites in China between 2013 and 2016 (Tables S1, S2, S3, and S5, Supplementary Material), especially for DPs, DBDPE, and CPs (Huang et al., 2018; Zeng et al., 2018), indicating potentially greater health concerns arising from exposure to HFRs in these regions.

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

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

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

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

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

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

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

The main pathway of human exposure to OPFRs appears very different to that of BFRs and DPs, with inhalation identified as the most important exposure pathway in a recent study of an informal e-waste site in Pakistan. This study reported human exposure to OPFRs through inhalation to be 3 orders of magnitude higher than via soil ingestion (Iqbal et al., 2017). This result was in agreement with studies conducted in Qingyuan, China. Specifically, human exposure to OPFRs through inhalation was estimated to be 12.1±4.1 ng/kg bw/day (mean±SD) for adults in Qingyuan, China, which exceeded the EDI via dust ingestion reported by He et al. (2015) (median: 7.02 ng/kg bw/day), Guo et al. (2019a) (mean: 5.85 ng/kg bw/day), and Zheng et al. (2015b) (mean: 1.36-2.11 ng/kg bw/day) for adults. It is interesting that in both informal e-waste regions in Pakistan and China, non-chlorinated OPFRs comprised the majority of total OPFRs exposure via inhalation (Iqbal et al., 2017; Luo et al., 2016). This may be attributable to less extensive use of chlorinated OPFRs in electrical and electronic products. Another interesting observation is that, despite the low contribution of food intake to human exposure to OPFRs reported in informal e-waste sites to date, chlorinated OPFRs were more frequently detected in chicken eggs sourced from an informal e-waste site in China than non-chlorinated OPFRs (Zheng et al., 2016). This might be explained by the relatively longer half-life of chlorinated OPFRs (Ma et al., 2017a).

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

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

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

5.1 Human blood and serum

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

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

5.2 Human hair

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

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

5.3 Human milk

Concentrations of HFRs in human milk can not only reflect internal exposure of female adults but also dietary exposure of nursing infants (Shi et al., 2018; Tang and Zhai, 2017). Concentrations of HFRs in human milk and the associated implications for human exposure, especially for infants, have previously been reviewed for China and Africa (Shi et al., 2018; Orisakwe et al., 2019). Specifically, a recent study conducted in an e-waste handling area in Wenling, China, collected 25 human milk samples from mothers who had lived there for over 20 years (defined as the R_{20} group) and 21 human milk samples from mothers who had resided there for no more than 3 years (defined as the R_3 group), and determined concentrations of 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209) in these samples (Li et al., 2017). It found that ΣPBDE concentrations in the R_{20} group (mean: 25.7±20.0 ng/g lipid; range: 7.89-90.6 ng/g lipid) exceeded significantly those in the R_3 group (mean: 6.68±5.61 ng/g lipid; range: 1.87-22.0 ng/g lipid) (Li et al., 2017). The two groups had similar congener profiles of PBDEs, with BDE-209 and -153 being the most abundant congeners in both groups (Li et al., 2017). Furthermore, the EDI of PBDEs for infants in the R_{20} group was in the range 15.8-243 ng/kg bw/day (median: 45.3 ng/kg bw/day), while in the R_3 group the range was 5.43-43.0 ng/kg bw/day (median: 11.4 ng/kg bw/day) (Li et al., 2017). Although this was much lower than the reference doses of the United States Environmental Protection Agency (2017), the maximum EDI of BDE-47 (76.9 ng/kg bw/day) and BDE-153 (98.9 ng/kg bw/day) (Li et al., 2017) for nursing infants in the R_{20} group approached the corresponding reference doses (BDE-47: 100 ng/kg bw/day; BDE-153: 200 ng/kg bw/day).

5.4 Human urine

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

5.5 Other human matrices

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

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

Health risk assessments of human exposure to HFRs and OPFRs have previously been reviewed in Africa (Asante et al., 2019; Orisakwe et al., 2019), India (Awasthi et al., 2016), China (Awasthi et al., 2018; Shi et al., 2018), and other regions on a global scale (Akram et al., 2019; Bakhiyi et al., 2018). Specifically, in informal e-waste sites, human exposure to TCIPP, TCEP, TNBP, and TPHP were correlated with elevated DNA oxidative stress in e-waste sites, as the urinary concentrations of BCIPP, BCEP, DBP, and DPHP were significantly increased as the concentration of 8-hydroxy-2′-deoxyguanosine (8-OHdG), a marker of DNA oxidative stress, increased (Lu et al., 2017). Moreover, PBDEs and NBFRs are reported thyroid hormone (TH) disruptors (Eguchi et al., 2015; Guo et al., 2019b; Zheng et al., 2017a, 2017b). They have strong binding affinity to thyroid-stimulating hormone (TSH), thyroglobulin, thyroxine-binding globulin (TBG), TH receptor α (TRα), and iodothyronine deiodinase I (ID1), and therefore could disrupt TH-regulated proteins and gene expression (Guo et al., 2019b). It is also suggested that PBDEs, NBFRs (e.g., DBDPE, BTBPE, BEH-TBP, etc.), and DP could exert similar disrupting effects on female follicle-stimulating hormone (FSH) and male testosterone, with NBFRs showing stronger disrupting effects on human sex hormones than do PBDEs (Guo et al., 2018). Furthermore, PBDEs were found to have adverse effects on human semen quality measured by sperm concentration and count, sperm progressive motility, and sperm viability (Yu et al., 2018).

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

6 Conclusions

The evidence reviewed in this study indicates 3 main pathways of human external exposure to HFRs and OPFRs in informal e-waste handling sites, i.e., inhalation, ingestion of dust and food, and dermal absorption. Current evidence suggests EDIs of OPFRs via inhalation exceed those via food intake and dust ingestion, and thus inhalation could be the dominant pathway of human exposure to OPFRs in informal e-waste sites; while for PBDEs, NBFRs, and DPs, food consumption and indoor dust ingestion are likely more important contributors. An important factor emerging from our review is that human exposure to HFRs and OPFRs through dermal absorption is insufficiently well-understood and may well be underestimated as pathways such as dermal absorption of both particulate and gaseous contaminants through air-to-skin transfer, as well as from direct skin contact with e-waste articles, have been largely overlooked. Children, infants, and fetuses, as well as e-waste recycling workers were found to experience higher HFR and OPFR exposure than did non-occupationally-exposed adults inhabiting e-waste handling zones, or those living in non-e-waste-impacted locations. Gender differences in human internal exposure to HFRs in informal e-waste locations were reported in some studies with higher HFR concentrations in serum and hair observed in females compared to males. The cause of these higher HFR concentrations in females is unclear, especially in serum.

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

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

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

Declarations of interest

The authors declare no conflict of interest.

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Tables and figures

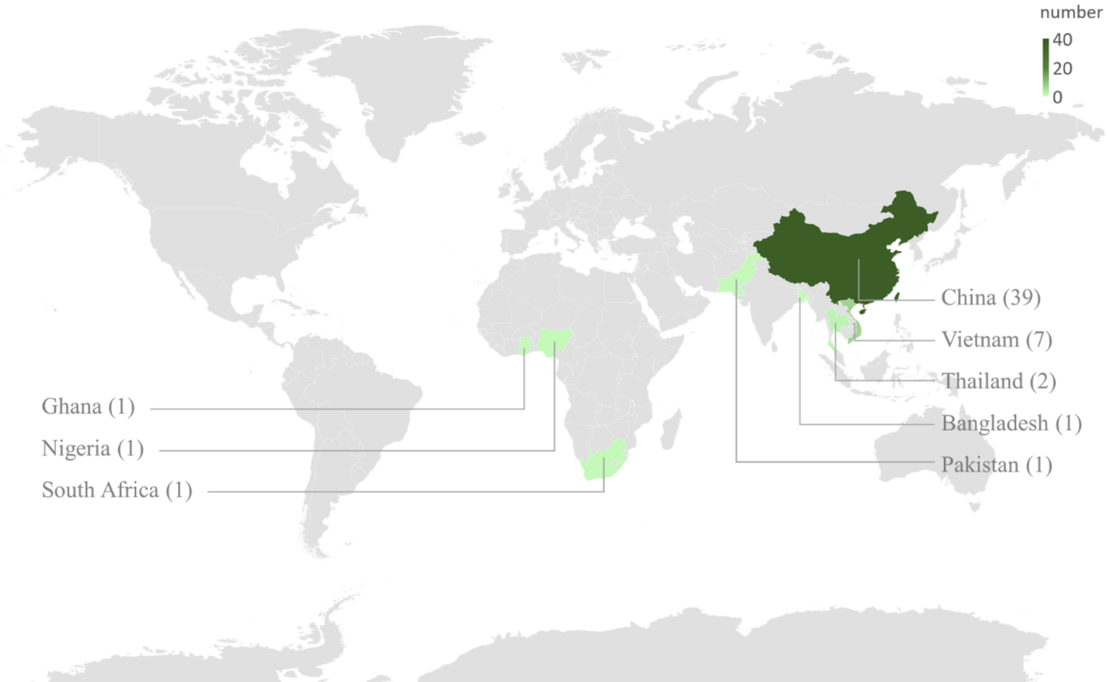

Fig. 1: Global distribution of recent research studies reporting human exposure to HFRs and OPFRs through informal e-waste handling activities (2015-2020)

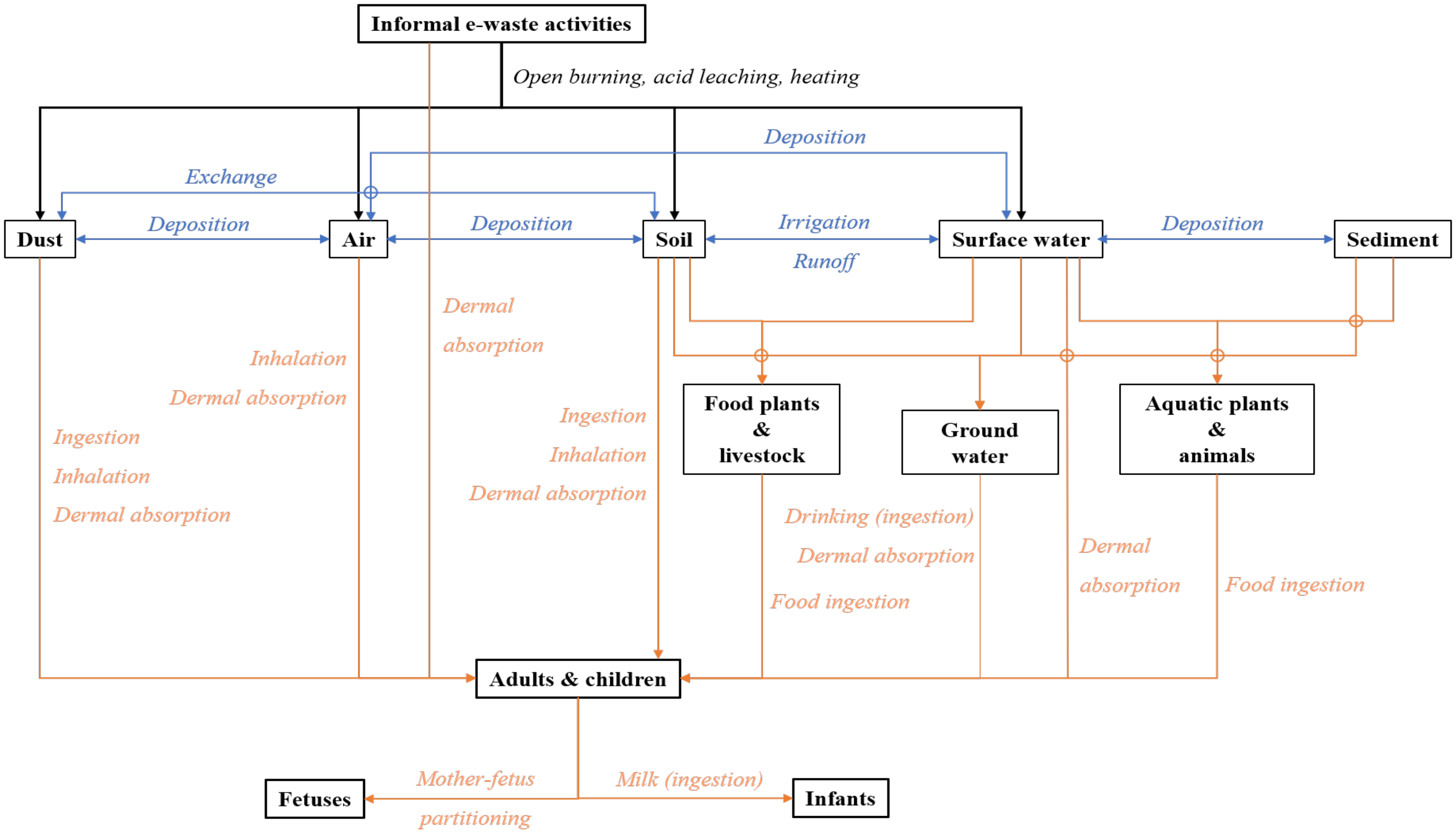

Fig. 2: Transfer of HFRs and OPFRs between various environmental media and associated human exposure pathways (Leung, 2019)

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

Compound	RfD	Compound	RfD
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(1)
BTBPE	243000	DP	2000000(2)
TBB	20000	DP	10000(3)
BEH-TBP	20000	∑HBCDDs	200
DBDPE	333333	TBBP-A	600000

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

Source: Ali et al. (2012), except United States Environmental Protection Agency (2017) for PBDEs, Wang et al. (2013) for DP, and Besis et al. (2017) for HBCDD and TBBP-A.

Table 2. Concentrations of BDE-209, DBDPE, and BTBPE in indoor dust collected from 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.(1)-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.(2)	n.a.	(He et al., 2017)

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

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