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

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

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

1. Introduction

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

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

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

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

2. Methods

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

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

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

3.1 Emissions of FRs to air

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

3.1.1 Impact of physicochemical properties of FRs on emissions to air

A cubic stainless-steel container (10 cm × 10 cm × 5 cm height) was placed in a constant-temperature oven (20 ℃) to allow the emission rates of tris(1,3-dichloroisopropyl) phosphate

(TDCIPP; vapour pressure: 4.1 × 10^{-4} Pa at 25 °C) and tricresyl phosphate (TCP; vapour pressure: 1.1-5.1 × 10^{-5} Pa at 25 °C) to be measured (Tokumura et al., 2019). Test results showed a substantially higher emission rate of TDCIPP (0.17 μg·m^{-2}·h^{-1}) than TCP (0.060 μg·m^{-2}·h^{-1}), despite the slightly lower concentration of TDCIPP (4,310 μg·g^{-1}) than TCP (4,840 μg·g^{-1}) in the curtains. The authors concluded that the vapour pressure of an OPE exerted an important impact on its emission rate. Similar results were also obtained for BFRs (Rauert et al., 2015; Sun et al., 2018). Specifically, significant correlations between the PBDE mass captured on a flexible polyurethane foam (PUF) cylinder (and deemed to have volatilised from curtains) and the vapour pressure (at 60°C, Table S3, Supplementary Material) of PBDE congeners were observed in a cylindrical test chamber (r^2=0.84, p=0.003) and a Micro-Chamber (r^2=0.67, p=0.024), which implied that the volatilisation emission behaviour of PBDEs was strongly influenced by their vapour pressure (Rauert et al., 2015). Sun et al. (2018) also observed higher emission rates of HBCDDs than of some NBFRs such as bis(2,4,6-tribromophenoxy) ethane (BTBPE) and bis(2-ethyl hexyl) tetrabromophthalate (BEH-TEBP or TBPH). Moreover, HBCDD had higher sensitivity to the rise of temperature (i.e., greater increase in emission rates per unit increase in temperature) than did NBFRs, which could possibly be attributed to the higher vapour pressure of HBCDD than the studied NBFRs (Table S3, Supplementary Material).

3.1.2 Impact of temperature on emissions of FRs to air

Tokumura et al. (2019) revealed a sharp (65-fold) increase in the emission rate of TDCIPP with increasing temperature, from an average emission rate of 0.17 μg·m^{-2}·h^{-1} at 20 °C to 11 μg·m^{-2}·h^{-1} at 60 °C. Similar results were reported for tris(1-chloro-2-propyl) phosphate (TCIPP) (Poppendieck et al., 2017), NBFRs (Sun et al., 2018), HBCDD (Kajiwara and Takigami, 2013; Sun et al., 2018), and PBDEs (Kajiwara and Takigami, 2013; Kemmlein et al., 2003). For instance, Kajiwara and Takigami (2013) adapted a small cylindrical stainless steel chamber (7 cm diameter × 5.5 cm height) to conduct emission tests on three BFR-treated textile samples at temperature of 20-80 °C. While the emission rates of both HBCDDs and PBDEs remained stable over a temperature range of 20-60 °C, a considerable increase in the emission rates of HBCDDs (~20 fold) and PBDEs (~6 fold) was observed at 80 °C. Notably, the relative abundance of α-HBCDD, which has the highest vapour pressure among the three HBCDD diastereomers, continued to increase as the temperature was raised, while that of γ-HBCDD (which has the lowest vapour pressure among the three HBCDD diastereomers) continued to decline (vapour pressure at 25 ℃: 1.05×10^{-8} for α-HBCDD, 5.82×10^{-9} for β-HBCDD, 8.39×10^{-11} for γ-HBCDD). Moreover, the relative abundance of $\sum_{\text{di- to octa-}}$BDEs compared to $\sum_{\text{nona- to deca-}}$BDEs was noted to increase with temperature.

3.1.3 Impact on FR emissions to air of timescale of experiments

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

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

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

3.1.5 Other factors influencing emissions of FRs to air from products

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

3.2 Emissions of FRs via abrasion of products

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

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

3.3 Leaching of FRs to surrounding environment

Leaching of FRs occurs in landfills, dumpsites, and waste recycling and dismantling facilities, etc. when FR-treated products come in contact with aqueous fluids. For instance, the presence of HFRs and OPEs in landfill leachates has been reported in several previous studies, with concentrations of

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

3.3.1 Impact of leachate DHM content on FR leaching

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

3.3.2 Impact on FR leaching of leachate temperature

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

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

3.3.3 Impact of leachate pH on leaching of FRs

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

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

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

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

3.3.5 Other factors influencing leaching of FRs from products

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

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

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

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

Previous studies have reported high levels of HFR and OPE contamination in soil near e-waste dismantling and recycling facilities (Table 2 and Table 3), with their concentrations occasionally exceeding 100,000 $ng \cdot g^{-1}$ dry weight (dw) (Ceballos and Dong, 2016; Ge et al., 2020; McGrath et al., 2017a, 2018). McGrath et al. (2018) reported concentrations of 8 PBDE congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209) and 6 NBFRs (pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), HBBz, 2-ethyl hexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), BTBPE, and decabromodiphenyl ethane (DBDPE)) in 36 soils collected from two Australian e-waste recycling plants and 8 reference soils. Concentrations in soil of $\sum_8$PBDEs were in the range 34-5,000 $ng \cdot g^{-1}$ dw (median: 130 $ng \cdot g^{-1}$ dw) and 8.3-98,000 $ng \cdot g^{-1}$ dw (median: 160 $ng \cdot g^{-1}$ dw) in the two e-waste recycling plants, respectively. Such elevated PBDE concentrations exceeded considerably those in the reference soils (range: 0.10-44 $ng \cdot g^{-1}$ dw, median: 21 $ng \cdot g^{-1}$ dw), and concentrations of $\sum_6$NBFRs in soils from the two e-waste recycling plants (median: 3.8 and 15 $ng \cdot g^{-1}$ dw, respectively) were also significantly higher than those in reference soils (median: < 0.02 $ng \cdot g^{-1}$ dw). In addition, Ge et al. (2020) observed much greater soil contamination with FRs in an industrial park in Guiyu, China, than in surrounding areas. Median concentrations of $\sum_{20}$PBDEs (tri- to deca-BDE), $\sum_2$NBFRs (pentabromobenzene (PBBz) and HBBz), $\sum_2$DPs (dechlorane plus;

syn-DP and *anti*-DP), and $\sum_{13}$OPEs (tripropyl phosphate (TPP), tributyl phosphate (TBP), tris(2-chloroethyl) phosphate (TCEP), TCIPP, tris(2-chloropropyl) phosphate (T(2-C)PP), TDCIPP, triphenyl phosphate (TPHP), 2-ethylhexyl diphenyl phosphate (EHDPP), tris(2-ethylhexyl) phosphate (TEHP), tris(2-isopropylphenyl) phosphate (TIPPP), and 3 isomers of TCP) were 46,300 $ng \cdot g^{-1}$ dw, 294 $ng \cdot g^{-1}$ dw, 712 $ng \cdot g^{-1}$ dw, 12,000 $ng \cdot g^{-1}$ dw, respectively, in soils collected from the industrial park. These observations were 1 to 2 orders of magnitude higher than the corresponding FR concentrations in soils from surrounding areas. Similarly, higher HFR and OPE concentrations have been reported in soils from e-waste dismantling and recycling areas than in surrounding areas (e.g., urban and rural areas, farmlands, and background areas) in China (e.g., Hongkong, Tianjin, Shanghai, etc.), South Korea, and Vietnam (Li et al., 2014, 2016, 2017; Man et al., 2011; Wang et al., 2018b). Together, these observations indicate formal e-waste recycling facilities have great potential to contaminate surrounding soils, despite use of environmentally friendly recycling technologies.

Pertinently, McGrath et al. (2018) reported significant negative correlations ($p<0.05$) between PBDE and NBFR concentrations in soils and distance from the two e-waste facilities, and the significant difference between BFR concentrations in soils collected from 300-900 m away from an e-waste site and BFR concentrations in reference soils further illustrated the potential of regulated e-waste recycling to significantly elevate BFR concentrations in soils located up to 900 m from such activity. These observations were consistent with another two studies where significant negative correlations were reported between concentrations of PBDEs, NBFRs, and DPs in soil and the distance between sampling locations and an industrial park ($p<0.05$) (Ge et al., 2020) and an e-waste recycling area (p value not provided) (Hong et al., 2018). This is a clear indication that these e-waste recycling facilities acted as an emission point source, and the authors further concluded that distribution of BFRs and DPs in soil from e-waste recycling facilities matches the point source pollution pattern (Ge et al., 2020; Hong et al., 2018; McGrath et al., 2018). It is also interesting to note that significant correlations between OPE concentrations in soil and distance from the e-waste dismantling park were not found, which possibly means the e-waste dismantling park is not a dominant source of OPEs to surrounding soils compared to diffuse OPE emissions from their in-use consumer goods as both FRs and plasticisers (Ge et al., 2020). Thus, further investigation is recommended to better understand the impact of formal e-waste dismantling and recycling facilities on local OPE contamination.

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

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

Interestingly, Harrad et al. (2020b) reported PBDE, DBDPE, and HBCDD concentrations in landfill-related soils from Ireland and came to the opposite conclusion. Concentrations in soil of $\sum_7$PBDEs (BDE-28, -47, -99, -100, -153, -154, and -183), BDE-209, DBDPE, and $\sum_3$HBCDDs (α-, β-, γ-HBCDD) were: 0.043-7.6 ng·g^{-1} dw, 0.065-63 ng·g^{-1} dw, ND-0.54 ng·g^{-1} dw, and ND-6.2 ng·g^{-1} dw, with median concentrations of 0.14 ng·g^{-1} dw, 0.57 ng·g^{-1} dw, ND, and 0.55 ng·g^{-1} dw, respectively. These were comparable to PBDE concentrations in European background soils ($\sum_{20}$PBDEs; range: 0.065-12 ng·g^{-1} dw; median: 0.61-2.5 ng·g^{-1} dw) (Hassanin et al., 2004). Moreover, applying a non-parametric Wilcoxon signed rank test, the authors found no significant difference in BFR concentrations in soils between downwind and upwind locations ($p>0.1$), indicating no discernible impact of the landfills on concentrations of BFRs in surrounding soils. Reasons for this disparity compared to other studies are unclear, while proper management of landfills, applications of impervious polymeric liners, and sound classification of waste in Ireland could possibly be contributing factors.

In addition to PBDEs, data on soil concentrations of other FRs are scarce (Table 3). Concentrations of $\sum_{10}$OPEs (TCEP, TCIPP, TDCIPP, tri-isobutyl phosphate (TIBP), tri(n-butyl)phosphate (TNBP), tris(2-butoxyethyl) phosphate (TBOEP), TEHP, TPHP, EHDPP, and tris(methylphenyl) phosphate (TMPP)) and $\sum_4$NBFRs (DBDPE, BTBPE, EH-TBB, and BEH-TEBP) were in the range of 1.8-186 ng·g^{-1} dw and 1.1-83 ng·g^{-1} dw in soils from a waste landfill in Brazil, with mean values of 67 ng·g^{-1} dw and 19 ng·g^{-1} dw, respectively (Cristale et al., 2019). The highest soil concentrations of OPEs and NBFRs were observed in areas where e-waste and furniture PUF were disposed of, while lower concentrations were observed in soils collected from more distant locations. Concentrations of $\sum_2$DPs (*syn*- and *anti*-DP) were in the range of ND-3.97 ng·g^{-1} dw in soils collected from an official municipal waste dumpling site in Pakistan (Hafeez et al., 2016). The mean concentration of $\sum_2$DPs was highest in soils collected from dumpsites (0.48 ng·g^{-1} dw), followed by agricultural zone (0.33 ng·g^{-1} dw), roadside (0.05 ng·g^{-1} dw), and residential zone (0.04 ng·g^{-1} dw), respectively. Li et al. (2021) reported elevated concentrations of short-chain chlorinated paraffins (SCCPs) in soils (56.8-1,348 ng·g^{-1} dw) from an urban landfill and rural dumpsites in Tibet, China. Within a 5 km distance from the landfill, SCCP concentrations in soils declined rapidly with increasing distances from the landfill, while SCCP concentrations relatively leveled off outside the 5 km distance. The results suggest the potential of landfills to significantly elevate SCCP concentrations in soils located up to 5 km from such activity, and this could be attributed to the atmospheric dispersion of SCCPs (Li et al., 2021). Further studies are encouraged to facilitate better understanding of the potential of landfills to elevate FR concentrations in surrounding soils.

4.3 FR concentrations in soil near waste incineration plants

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

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

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

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

Hong et al. (2018) reported concentrations of a broad range of HFRs in the atmosphere in Ziya Circular Economy Area (Tianjin, China), including 13 PBDEs (BDE-17, -28, -47, -49, -66, -85, -99, -100, -138, -153, -154, 183, and -209), 17 NBFRs (BTBPE, DBDPE, allyl 2,4,6-tribromophenyl ether (ATE), 2,3,5,6-tetrabromo-p-xylene (p-TBX), α- and β-tetrabromoethylcyclohexane (TBECH), PBBz, 2-bromoallyl 2,4,6-tribromophenyl ether (BATE), PBT, α- and β-1,2,5,6-tetrabromocyclooctane (TBCO), PBEB, HBBz, 2,3-dibromopropyl 2,4,6-tribromophenyl ether (DPTE), pentabromobenzyl acrylate (PBBA), hexachlorocyclopentenyl-dibromocyclooctane (HCDBCO), and octabromotrimethylphenylindane (OBTMPI)), and 2 DP isomers (*syn*- and *anti*-DP). All the targeted HFRs were detected except for OBTMPI, with concentrations of $\sum_{13}$PBDEs, $\sum_{16}$NBFRs (OBTMPI excluded), and $\sum_{8}$DPs being 10,600 pg·m^{-3}, 1,330 pg·m^{-3}, and 109 pg·m^{-3}, respectively. These concentrations detected in the Ziya Circular Economy Area were comparable to those detected in outdoor air in informal e-waste sites in South China (PBDEs + NBFRs: 120-19,000 pg·m^{-3}, DPs: 13-1,800 pg·m^{-3}), but exceeded substantially those at reference sites (PBDEs + NBFRs: 55-1,700 pg·m^{-3}, DPs: 0.47-36 pg·m^{-3}) (Chen et al., 2011; Tian et al., 2011). Cahill et al. (2007) reported a similar impact of formal e-waste recycling activities on elevated PBDE concentrations in outdoor air in California, US. Specifically, concentrations of $\sum_{30}$PBDE congeners (tri- to deca-BDEs; mean: 93 pg·m^{-3}) in the ambient atmosphere were 1 to 2 orders of magnitude lower than those recorded outside an e-waste recycling facility (mean: 340-8,600 pg·m^{-3}) and an automotive shredding and metal recycling facility (mean: 390-810 pg·m^{-3}). Furthermore, in air sampled on the e-waste site, samples collected near the waste loading dock had mean PBDE levels that were an order of magnitude higher than those in outdoor air collected from the opposite side of the building and downwind of the facility. Moreover, atmospheric PBDE concentrations doubled when normal operation activities were performed in the automotive shredding and metal recycling facility compared to when no activity was performed. Elevated atmospheric concentrations of PBDEs were also observed in formal e-waste recycling facilities in Norway (Morin et al., 2017) as well as at official e-waste storage facilities in Thailand (Muenhor et al., 2010). These results imply significant impacts of formal e-waste activities on HFR contamination in outdoor air.

Similar results were also observed for OPEs in a gridded field study conducted in Tianjin, China. Liang et al. (2020) found higher concentrations of TDCIPP, TCIPP, EHDPP, TEHP, and $\sum_3$TCPs (*o*-TCP, *m*-TCP, and *p*-TCP) in an e-waste dismantling area than at other sampling sites (urban, suburban, and rural areas), but the specific concentrations were not presented. Furthermore, Wang et al. (2018a) reported high concentrations of 12 OPEs (triethyl phosphate (TEP), TNBP, TBOEP, TEHP, TCEP, TCIPP, TDCIPP, EHDPP, TPHP, TPP, TIPPP, and TMPP) in fine particulate matter ($PM_{2.5}$) at four e-waste recycling sites in a rural region in Guangzhou, China. OPE concentrations ranged from 780 pg·m^{-3} to 14,000 pg·m^{-3} with a median concentration of 3,300 pg·m^{-3} in the four e-waste recycling facilities. These concentrations were 1 to 2 orders of magnitude higher than those in airborne particles over the South China Sea (47-160 pg·m^{-3}) (Lai et al., 2015), which possibly indicates that e-waste recycling facilities could be a potential source of OPEs to the surrounding atmosphere. However, it is notable that OPE concentrations in $PM_{2.5}$ at the e-waste recycling sites were comparable to those at 20 industrial sites in an urban region (range: 520-63,000 pg·m^{-3}; median: 2,800 pg·m^{-3}) (Wang et al., 2018a), which implies the e-waste recycling facilities are not a dominant source of OPEs to the surrounding atmosphere compared to OPE emissions from certain industries. The authors suggested high OPE concentrations in $PM_{2.5}$ were associated with: electrical and electronics manufacturing, plastics manufacture, waste recycling, and certain other chemical industries, while lower concentrations were related to: machinery, paper, clothing, and furnishing industries (Wang et al., 2018a). Therefore, despite the limited data reported hitherto, these studies provide evidence of a non-negligible (although not dominant) impact of e-waste recycling on OPE emissions to ambient air.

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

A study conducted in Ireland reported atmospheric concentrations of 8 PBDEs (BDE-28, -47, -99, -100, -153, -154, -183, and -209), 3 HBCDDs (α-, β-, and γ-HBCDD), and DBDPE in the vicinity of 10 municipal solid waste landfills (Harrad et al., 2020b). BDE-47 and -209 were the only PBDE congeners detected, with median concentrations of 0.23 pg·m^{-3} and 4.3 pg·m^{-3}, respectively. $\sum_3$HBCDDs were not detected in most samples, and the highest concentration was 6.1 pg·m^{-3}, while DBDPE was only detected in one of the 20 air samples (2.0 pg·m^{-3}). Similar results were also reported by Weinberg et al. (2011), Morin et al. (2017), and St-Amand et al. (2008), who observed comparable PBDE concentrations (generally lower than 20 pg·m^{-3}) in outdoor air collected from landfills in Germany, Norway, and Canada, respectively. The atmospheric BFR concentrations observed in the four studies were generally at the same levels with those observed in urban and rural areas across Europe (Jaward et al., 2004a, 2004b; Law et al., 2008), indicating minimal impact of landfills on HFR concentrations in outdoor air.

Compared to those observations in European countries, HFR concentrations in ambient air in landfills were higher in Pakistan and South Africa (Hafeez et al., 2016; Katima et al., 2018). Concentrations of $\sum_8$PBDEs (BDE-28, -35, -47, -99, -100, -153, -154, and -183) and $\sum_2$DPs (*syn-* and *anti*-DP) ranged 53.8-454 pg·m^{-3} and 0.02-1.56 pg·m^{-3}, respectively, in 6 ambient air samples collected from three major zones (main dumping site and Lahore compost zone, adjacent agricultural zone, and residential zone) in the vicinity of Mahmood Booti, the only official municipal waste dumping site in Lahore, Pakistan (Hafeez et al., 2016). Interestingly, mean concentrations of $\sum_8$PBDEs (212 pg·m^{-3}) and $\sum_2$DPs (0.58 pg·m^{-3}) were higher in the main dumpsite than in other zones ($\sum_8$PBDEs mean: 79.4-175 pg·m^{-3}; and $\sum_2$DPs mean: 0.41-0.55 pg·m^{-3}). Although the significance of this difference was not available due to the limited number of samples, the relatively higher HFR concentrations near the dumpsite could be explained by the failure to fulfill sanitary landfill requirements and the lack of pollution control facilities and leachate treatment in Mahmood Booti. Similarly, atmospheric concentrations of $\sum_9$PBDEs (BDE-17, -28, -47, -99, -100, -153, -154, -183, and -209), $\sum_3$HBCDDs (α-, β-, and γ-HBCDD), EH-TBB, BEH-TEBP, and BTBPE in landfill sites were also relatively higher than those observed in other zones (including industrial, urban, semi-urban, and rural areas) in Gauteng Province, South Africa, which could probably be attributed to the improper management of wastes disposed in the landfills (Katima et al., 2018). To the best of our knowledge, no data exist on atmospheric concentrations of OPEs in the vicinity of regulated waste dumpsites or landfills, thus further studies should be undertaken to fill this knowledge gap.

5.3 Concentrations of FRs in air near waste incineration plants

Data on atmospheric concentrations of FRs in incineration plants is very limited, with only 3 publications located and included in this review (Table 4). Atmospheric $\sum_{30}$PBDE (di- to deca-BDE) concentrations ranged from 24.9-139 pg·m^{-3} and 32.1-71.6 pg·m^{-3} in two municipal solid waste incinerators in Taiwan, China, with median values of 51.9 and 48.9 pg·m^{-3}, respectively (Tu et al., 2012). Similarly, Wang et al. (2010b) also observed comparable $\sum_{30}$PBDE concentrations (25.7-100 pg·m^{-3}) and significantly lower $\sum$PBB concentrations (congeners not reported; range: 0.149-0.556 pg·m^{-3}) in ambient air collected from one municipal solid waste incinerator in Taiwan, China. The contribution of the two municipal solid waste incinerators to total PBDE concentration in ambient air was only 0.026%, indicating minimal impact of incinerators on PBDE emissions to ambient air (Tu et al., 2012). This was further supported by the comparable atmospheric concentrations of PBDEs in incineration plants to those detected in ambient air collected in urban areas in Taiwan, China ($\sum_{30}$PBDE median: 34.7 pg·m^{-3}) (Wang et al., 2011). PBDE concentrations in ambient air were also studied in the vicinity of a solid waste incinerator in Sweden (Agrell et al., 2004). Specifically, $\sum_{8}$PBDEs (BDE-28, -47, -66, -100, -153, -154, -183, and -209) were determined to fall in the range 2.2-123.5 pg·m^{-3}, with a median value of 19.2 pg·m^{-3}. These were generally at the same level to $\sum_{8}$PBDE concentrations (range: ND-192.8 pg·m^{-3}; median: 15.1 pg·m^{-3}) detected in outdoor air at urban reference sites. A significant difference (paired t-test, $p<0.01$) was observed between atmospheric concentrations of BDE-47 and $\sum_{7}$PBDEs (excluding BDE-209) between incinerator sites and urban reference sites, but not for BDE-209 and $\sum_{8}$PBDEs (paired t-test, $p>0.1$). Given that the commercial deca-BDE mixture was still in widespread use in Sweden during this study (2001-2002), the authors suggested that the comparable BDE-209 concentrations at incinerator-impacted and urban reference sites were a reflection of BDE-209 emissions from in-use consumer products; but that for those "older" PBDEs (i.e., commercial penta-BDEs) which had at the time of the study already been restricted in the country, waste incineration might contribute meaningfully to their occurrence in outdoor air (Agrell et al., 2004). To the best of our knowledge, no data is available on atmospheric concentrations of OPEs in the vicinity of incineration plants, and further studies are recommended to fill this knowledge gap.

6. Conclusions and research gaps

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

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

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

Declarations of interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at

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

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

Table 2. Occurrence of PBDEs in soil in the vicinity of formal waste treatment facilities

Waste treatment method	Country	Sampling period	PBDE congeners	Concentration ($ng \cdot g^{-1}$ dw)	Reference
e-waste recycling	China	2018	$\sum_{20}$PBDEs	46300 (reference soil: 575) [a]	(Ge et al., 2020)
e-waste recycling	China	2015	$\sum_{13}$PBDEs	13 [b]	(Wu et al., 2019)
e-waste recycling	China	2015	BDE-209	90 [b]	(Wu et al., 2019)
e-waste recycling	China	2011	$\sum_{13}$PBDEs	250 [b]	(Hong et al., 2018)
e-waste recycling	China	2012	$\sum_{23}$PBDEs	3900 (background soil: 0.77) [b]	(Li et al., 2016)
e-waste recycling	China	2009	$\sum_{18}$PBDEs	34-1069 [c]	(Li et al., 2014)
e-waste recycling	China	2009	BDE-209	110-5850 [c]	(Li et al., 2014)
e-waste storage	China	-	$\sum_{22}$PBDEs	50.5 (agricultural soil: 27.5) [b]	(Man et al., 2011)
e-waste dismantling	China	-	$\sum_{22}$PBDEs	6875 (agricultural soil: 27.5) [b]	(Man et al., 2011)
e-waste recycling	Australia	2017	$\sum_{8}$PBDEs	130-160 (urban soil: 21) [a]	(McGrath et al., 2018)
e-waste recycling	South Korea	2012	$\sum_{23}$PBDEs	9.0 (background soil: 1.4) [b]	(Li et al., 2016)
e-waste recycling	Vietnam	2012	$\sum_{23}$PBDEs	68 (background soil: 0.23) [b]	(Li et al., 2016)
landfill	China	2011-2012	$\sum_{8}$PBDEs	234 [a]	(Huang et al., 2013)
landfill	China	2017	$\sum_{12}$PBDEs	0.13-1.2 [c]	(Li et al., 2018b)
landfill	Ireland	2018-2019	$\sum_{7}$PBDEs	0.14 [a]	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	BDE-209	0.57 [a]	(Harrad et al., 2020b)
landfill	Brazil	2015	$\sum_{8}$PBDEs	276 [b]	(Cristale et al., 2019)
landfill	South Africa	-	$\sum_{7}$PBDEs	7.43 [a]	(Akortia et al., 2019)
landfill	Canada	2004-2006	$\sum_{60}$PBDEs	131 (background soil: 1.94) [b]	(Danon-Schaffer et al., 2008)
dumpsite	Pakistan	-	$\sum_{8}$PBDEs	1.11 [b]	(Hafeez et al., 2016)
incinerator	Australia	2014	$\sum_{8}$PBDEs	13.6-80.8 (reference soil: 0.12-43.8) [c]	(McGrath et al., 2016)
incinerator	China	-	$\sum_{42}$PBDEs	1.4 [a]	(Zhang et al., 2013)

Note: a: median concentration; b: mean concentration; c: range

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

Waste treatment method	Country	Sampling period	FRs	Concentration ($ng \cdot g^{-1}$ dw)	References
e-waste recycling	China	2018	PBBz	47.9 (reference soil: 0.58) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	HBBz	249 (reference soil: 0.57) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_{2}$DPs	712 (reference soil: 11.3) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_{13}$OPEs	12000 (reference soil: 256) [a]	(Ge et al., 2020)
e-waste recycling	China	2011	$\sum_{14}$NBFRs	128 [b]	(Hong et al., 2018)
e-waste recycling	China	2011	$\sum_{2}$DPs	34.7 [b]	(Hong et al., 2018)
e-waste recycling	China	2012	$\sum_{19}$NBFRs	800 (reference soil: 12) [b]	(Li et al., 2017)
e-waste recycling	South Korea	2012	$\sum_{19}$NBFRs	18 (reference soil: 13) [b]	(Li et al., 2017)
e-waste recycling	Vietnam	2012	$\sum_{19}$NBFRs	21 (reference soil: 0.68) [b]	(Li et al., 2017)
e-waste recycling	Australia	2017	$\sum_{6}$NBFRs	3.8-15 (urban soil: ND) [a]	(McGrath et al., 2018)
multi-waste recycling	China	-	$\sum_{12}$OPEs	116 (farmland soil) 56.3 [a]	(Wang et al., 2018b)
landfill	Ireland	2018-2019	$\sum_{3}$HBCDDs	0.55 [a]	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	DBDPE	ND [a]	(Harrad et al., 2020b)
landfill	Brazil	2015	$\sum_{10}$OPEs	67 [b]	(Cristale et al., 2019)
landfill	Brazil	2015	$\sum_{4}$NBFRs	19 [b]	(Cristale et al., 2019)
landfill	China	2017	SCCPs	56.8-1348 [c]	(Li et al., 2021)
dumpsite	Pakistan	-	$\sum_{2}$DPs	0.48 [b]	(Hafeez et al., 2016)
incinerator	Australia	2014	HBBz	ND-0.34 (reference soil: ND) [c]	(McGrath et al., 2017b)

Note: a: median concentration; b: mean concentration; c: range.

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

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

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

Note: a: median concentration; b: mean concentration; c: range.

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

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

Table 2. Occurrence of PBDEs in soil in the vicinity of formal waste treatment facilities

Waste treatment method	Country	Sampling period	PBDE congeners	Concentration ($ng \cdot g^{-1}$ dw)	Reference
e-waste recycling	China	2018	$\sum_{20}$PBDEs	46300 (reference soil: 575) [a]	(Ge et al., 2020)
e-waste recycling	China	2015	$\sum_{13}$PBDEs	13 [b]	(Wu et al., 2019)
e-waste recycling	China	2015	BDE-209	90 [b]	(Wu et al., 2019)
e-waste recycling	China	2011	$\sum_{13}$PBDEs	250 [b]	(Hong et al., 2018)
e-waste recycling	China	2012	$\sum_{23}$PBDEs	3900 (background soil: 0.77) [b]	(Li et al., 2016)
e-waste recycling	China	2009	$\sum_{18}$PBDEs	34-1069 [c]	(Li et al., 2014)
e-waste recycling	China	2009	BDE-209	110-5850 [c]	(Li et al., 2014)
e-waste storage	China	-	$\sum_{22}$PBDEs	50.5 (agricultural soil: 27.5) [b]	(Man et al., 2011)
e-waste dismantling	China	-	$\sum_{22}$PBDEs	6875 (agricultural soil: 27.5) [b]	(Man et al., 2011)
e-waste recycling	Australia	2017	$\sum_{8}$PBDEs	130-160 (urban soil: 21) [a]	(McGrath et al., 2018)
e-waste recycling	South Korea	2012	$\sum_{23}$PBDEs	9.0 (background soil: 1.4) [b]	(Li et al., 2016)
e-waste recycling	Vietnam	2012	$\sum_{23}$PBDEs	68 (background soil: 0.23) [b]	(Li et al., 2016)
landfill	China	2011-2012	$\sum_{8}$PBDEs	234 [a]	(Huang et al., 2013)
landfill	China	2017	$\sum_{12}$PBDEs	0.13-1.2 [c]	(Li et al., 2018b)
landfill	Ireland	2018-2019	$\sum_{7}$PBDEs	0.14 [a]	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	BDE-209	0.57 [a]	(Harrad et al., 2020b)
landfill	Brazil	2015	$\sum_{8}$PBDEs	276 [b]	(Cristale et al., 2019)
landfill	South Africa	-	$\sum_{7}$PBDEs	7.43 [a]	(Akortia et al., 2019)
landfill	Canada	2004-2006	$\sum_{60}$PBDEs	131 (background soil: 1.94) [b]	(Danon-Schaffer et al., 2008)
dumpsite	Pakistan	-	$\sum_{8}$PBDEs	1.11 [b]	(Hafeez et al., 2016)
incinerator	Australia	2014	$\sum_{8}$PBDEs	13.6-80.8 (reference soil: 0.12-43.8) [c]	(McGrath et al., 2016)
incinerator	China	-	$\sum_{42}$PBDEs	1.4 [a]	(Zhang et al., 2013)

Note: a: median concentration; b: mean concentration; c: range

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

Waste treatment method	Country	Sampling period	FRs	Concentration ($ng \cdot g^{-1}$ dw)	References
e-waste recycling	China	2018	PBBz	47.9 (reference soil: 0.58) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	HBBz	249 (reference soil: 0.57) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_{2}$DPs	712 (reference soil: 11.3) [a]	(Ge et al., 2020)
e-waste recycling	China	2018	$\sum_{13}$OPEs	12000 (reference soil: 256) [a]	(Ge et al., 2020)
e-waste recycling	China	2011	$\sum_{14}$NBFRs	128 [b]	(Hong et al., 2018)
e-waste recycling	China	2011	$\sum_{2}$DPs	34.7 [b]	(Hong et al., 2018)
e-waste recycling	China	2012	$\sum_{19}$NBFRs	800 (reference soil: 12) [b]	(Li et al., 2017)
e-waste recycling	South Korea	2012	$\sum_{19}$NBFRs	18 (reference soil: 13) [b]	(Li et al., 2017)
e-waste recycling	Vietnam	2012	$\sum_{19}$NBFRs	21 (reference soil: 0.68) [b]	(Li et al., 2017)
e-waste recycling	Australia	2017	$\sum_{6}$NBFRs	3.8-15 (urban soil: ND) [a]	(McGrath et al., 2018)
multi-waste recycling	China	-	$\sum_{12}$OPEs	116 (farmland soil) 56.3 [a]	(Wang et al., 2018b)
landfill	Ireland	2018-2019	$\sum_{3}$HBCDDs	0.55 [a]	(Harrad et al., 2020b)
landfill	Ireland	2018-2019	DBDPE	ND [a]	(Harrad et al., 2020b)
landfill	Brazil	2015	$\sum_{10}$OPEs	67 [b]	(Cristale et al., 2019)
landfill	Brazil	2015	$\sum_{4}$NBFRs	19 [b]	(Cristale et al., 2019)
landfill	China	2017	SCCPs	56.8-1348 [c]	(Li et al., 2021)
dumpsite	Pakistan	-	$\sum_{2}$DPs	0.48 [b]	(Hafeez et al., 2016)
incinerator	Australia	2014	HBBz	ND-0.34 (reference soil: ND) [c]	(McGrath et al., 2017b)

Note: a: median concentration; b: mean concentration; c: range.

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

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

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

8 Note: a: median concentration; b: mean concentration; c: range.

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