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Short Communication

Are UK E-waste recycling facilities a source of environmental contamination and occupational exposure to brominated flame retardants?

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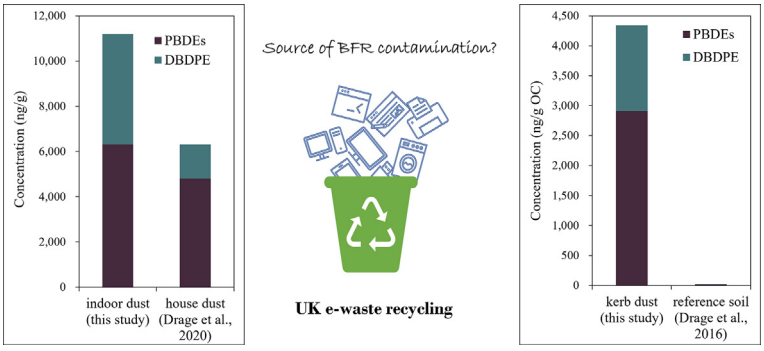
HIGHLIGHTS

- BDE-209 and DBDPE predominated BFRs in e-waste impacted dust in the UK.
- E-waste activities contributed to BFR contamination in UK urban environment.
- Industrial activities were another source of BFR contamination in the UK.
- Health burdens from BFR exposure were minimal for UK e-waste recyclers.

GRAPHICAL ABSTRACT



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ABSTRACT

Investigations into the impacts of regulated electrical and electronic waste (e-waste) recycling activities on urban environments in Europe remain rather scarce. In this study, dust samples taken both inside and outside of five UK e-waste recycling facilities were analysed for concentrations of polybrominated diphenyl ethers (PBDEs), novel brominated flame retardants (NBFRs), and hexabromocyclododecane (HBCDD). Average concentrations of ΣBFRs in dust inside and outside UK e-waste recycling facilities were 12,000 ng/g and 180 ng/g, with median concentrations of 7500 ng/g and 85 ng/g, respectively. BDE-209 and decabromodiphenyl ethane (DBDPE) were the most abundant BFRs in both indoor and kerb dust, making a combined contribution to ΣBFRs of ~90 % on average. While four out of the five studied e-waste facilities showed a lack of significant impact on BFR contamination in surrounding environment, one of the studied e-waste recycling facilities was identified as a likely source of BFR contamination to UK urban environments, with industrial activities as another potential source of NBFRs. Occupational exposure of UK e-waste recycling workers to BFRs via dust ingestion was generally lower than that estimated for e-waste recyclers from other countries, but was comparable to BFR exposure via dust ingestion of UK office workers. Our estimates suggested that health burdens posed by dust ingestion of BFRs were minimal for UK e-waste recycling workers.

1. Introduction

Proper treatment of electrical and electronic waste (e-waste), also known as waste electrical and electronic equipment (WEEE), is a global concern due to sharply increasing volumes of e-waste and the presence of a range of hazardous chemicals such as brominated flame retardants (BFRs) (Forti et al., 2020). Global phase-out of legacy BFRs including polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) has been implemented due to their persistence, bioaccumulation, and toxicity (Ma et al., 2022), e.g., these legacy BFRs bioaccumulate in humans and act as hormone disrupters (Ma et al., 2021). However, such legacy BFRs likely still present a contamination hazard due to their

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continuing presence in goods remaining in use, as well as in waste (Abbasi et al., 2019; Li and Wania, 2018). Restrictions on the use of legacy BFRs also resulted in increasing demand for novel BFRs (NBFRs) as replacements (Ma et al., 2022; Ma et al., 2021). This has generated a new challenge for public health, as some NBFRs showed similar or even stronger adverse effects on human health (Guo et al., 2019; Guo et al., 2018).

The impact of e-waste dismantling and recycling activities on BFR contamination to surrounding environments has been investigated in Asia and Africa, with China being the most studied area (Ma et al., 2022; Ma et al., 2021). Those studies revealed substantially increased concentrations of BFRs in soil or road dust in the vicinity of e-waste facilities, indicating e-waste activities, formal or informal, as an important source of BFR pollution in the studied regions (Ma et al., 2022; Ma et al., 2021). However, such investigations outside Asia and Africa are rather scarce, with only one study identified reporting considerably higher concentrations of PBDEs (median concentrations: 130–160 ng/g dw vs 21 ng/g dw) and NBFRs (median concentrations: 3.8–15 ng/g dw vs <0.02 ng/g dw) in urban soil surrounding two Australian e-waste recycling plants than in reference soil (McGrath et al., 2018). To the best of our knowledge, the impacts of regulated e-waste recycling activities on environmental contamination with BFRs have not hitherto been evaluated in Europe.

This is a potential oversight, as Europe has outperformed other regions in the world in e-waste recycling. The EU initially set a limit on the permissible concentration (1000 mg/kg) of PBDEs and HBCDD in recyclable plastics in e-waste, known as the low persistent organic pollutant (POP) content limit (LPCL) (European Union, 2019), which has been further reduced from 1000 mg/kg to 500 mg/kg in June 2022 (European Council, 2022). In 2019, around 42.5 % of e-waste generated in Europe was documented to be recycled, with this fig. 54.4 % in the UK (Forti et al., 2020). This formally collected e-waste was expected to present environmental contamination and human health risks, even when environmentally-sound techniques and personal protective equipment are implemented (Ma et al., 2022; Nguyen et al., 2019). Given this background, the current study determines BFR concentrations in dust collected from both inside and outside of five UK e-waste recycling facilities. Our aims are to: 1) evaluate potential impacts of e-waste recycling activities on BFR contamination of the surrounding environment; and 2) estimate occupational exposure to BFRs via dust ingestion for UK e-waste recycling workers, and evaluate its potential health risks.

2. Materials and methods

2.1. Sampling

Dust samples were collected from inside (n = 6; see Fig. 1) and outside (n = 30; see Fig. 2) of five e-waste recycling facilities located in the West

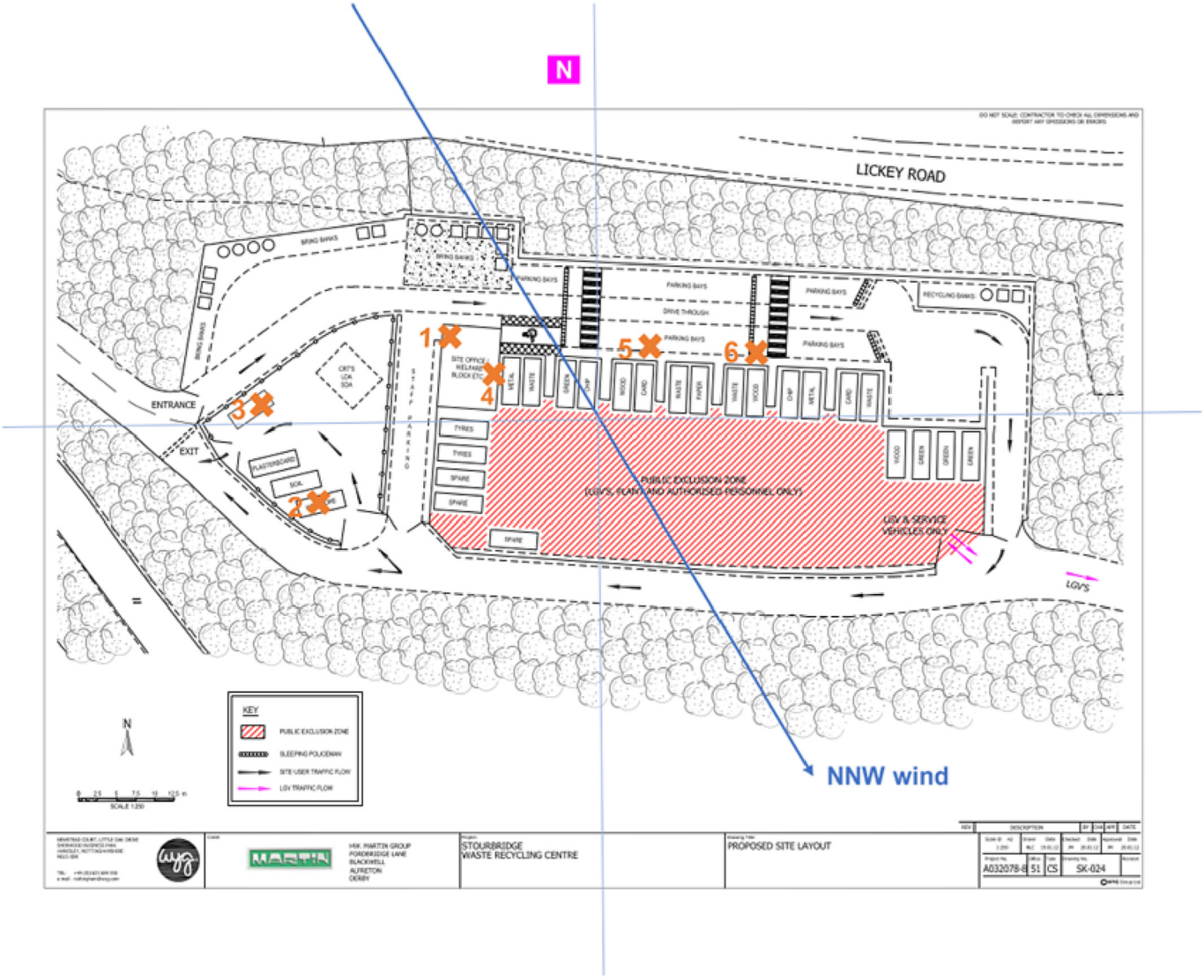

Fig. 1. Dust sampling sites inside a UK e-waste recycling facility (Facility #1).

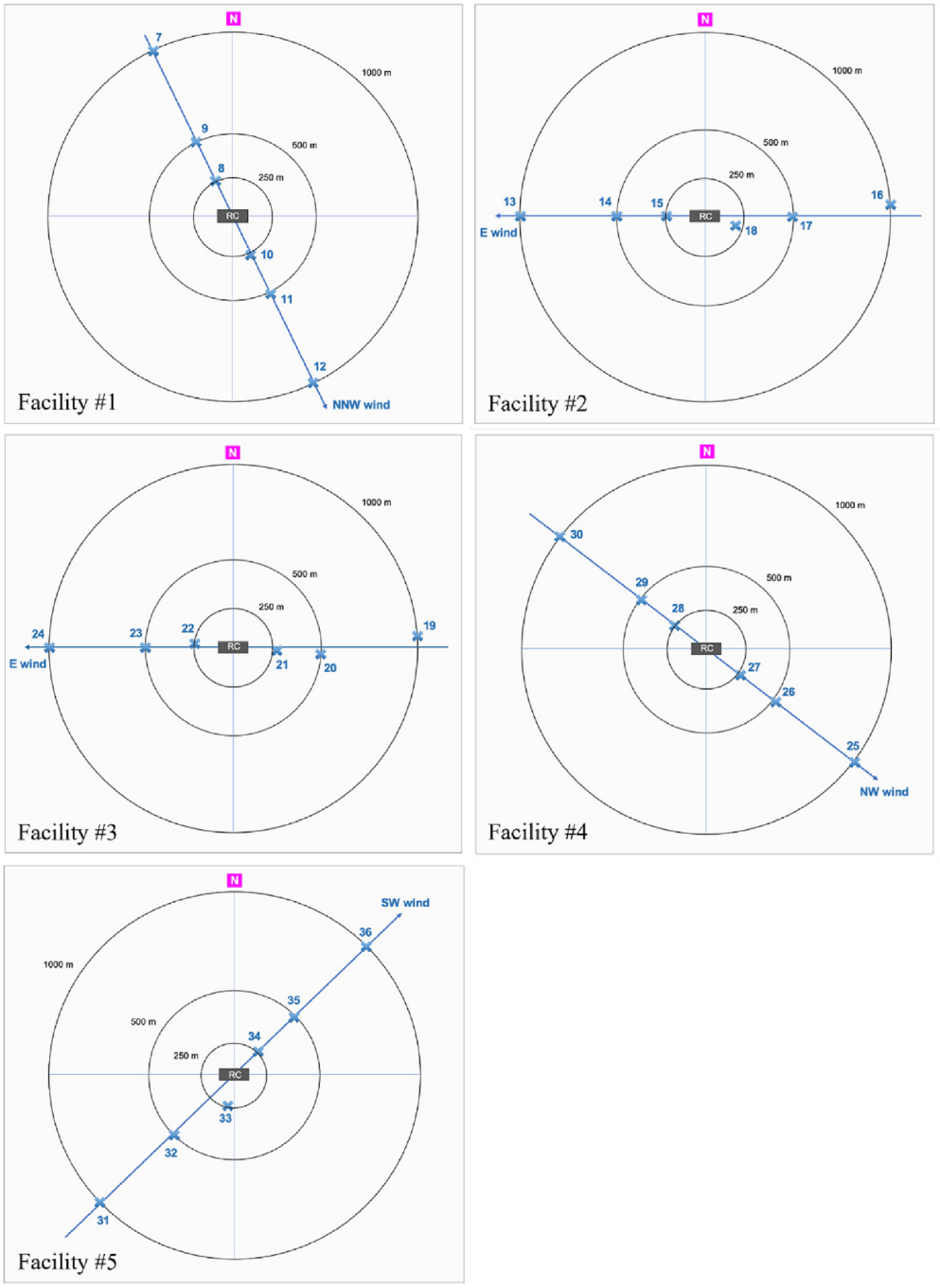

Fig. 2. Dust sampling sites outside five UK e-waste recycling facilities.

Midlands of the UK between September and November 2022. Outdoor (kerb) dust was collected from the kerbs between roads and pavements in the immediate vicinity of the five investigated e-waste recycling facilities. Indoor dust was only collected from inside of Facility #1 due to limited access to other facilities. A vacuum cleaner equipped with pre-cleaned nylon mesh filters (25 μm pore size) was used for dust collection. Detailed information on dust collection protocol has previously been published (Al-Omran and Harrad, 2016b), and is given in SM.

2.2. Analytical protocols

Chemicals and reagents used in this study are listed in SM, and protocols for extraction and clean-up of dust samples have been previously reported (Al-Omran and Harrad, 2016a). Briefly, dust samples were sieved with a stainless-steel test sieve of 500 μm pore size. Approximately 200 mg of dust was accurately weighed and spiked with 15 ng of internal (surrogate) standards (see SM for detailed information) prior to extraction with hexane and acetone (3:1, v/v). The crude extracts were split into two fractions via elution through a florisil column, before further purification with acid silica and aminopropyl functionalised silica, respectively. The two fractions were then combined and reconstituted into 100 μL of toluene containing 15 ng of $^{13}C_{12}$-BDE-100 and d_{18}-γ-HBCDD as recovery determination (syringe) standards prior to GC–MS and LC-MS/MS analysis (Ma et al., 2023). Detailed information on sample preparation and instrumental analysis is provided in SM. Specifically, dried kerb dust samples were heated at 500 °C for 4 h to allow organic matter content to be determined gravimetrically.

2.3. QA/QC

A five-point calibration was conducted for all the target BFRs (Table S1). Limits of detection (LODs) and limits of quantification (LOQs) for the target BFRs were calculated from a signal to noise ratio of 3 and 10, respectively (Table S2). One method blank was conducted along with each batch of 6 samples, generating 6 analyses of method blanks. No target BFRs were detected in the blanks, so the samples were not blank-corrected. Recoveries of the internal (surrogate) standards in blanks and samples are summarised in Table S3. Ten replicate analyses of NIST SRM 2585 (organic contaminants in house dust) were conducted prior to sample analyses, with results shown in Tables S3 and S4.

2.4. Health risk assessment

Daily intake of BFRs via dust ingestion was estimated by using Eq. (1) (Abdallah et al., 2008):

$$DI = \frac{CBFR \times IR \times FT}{BW} \quad (1)$$

where *DI* is the estimated daily intake of BFRs via dust ingestion (ng/kg bw/day), *CBFR* is the concentrations of BFRs in dust (ng/g), *IR* is the daily ingestion rate of dust (mg/day), *FT* is the fraction of time e-waste recycling workers spend at work per working day, and *BW* is the average body weight of UK adults (85.1 kg for males and 71.8 kg for females, respectively (NHS Digital, 2021)). Specifically, the working time of 8 h per day was assumed for occupational exposure to BFRs under both average and upper-bound exposure scenarios.

Non-carcinogenic risks from BFR exposure were evaluated by using Eq. (2) (Babalola and Adeyi, 2018):

$$HI = \Sigma \frac{EDI}{RfD} \quad (2)$$

where *HI* is hazard index, defined as sum of the ratios of estimated daily dust ingestion of BFRs to the corresponding reference dose (RfD). An *HI* value exceeding 1 suggests likely adverse non-cancer effects on human health of dietary exposure to BFRs, while an *HI* value below 1 indicates negligible health risks.

2.5. Statistical analysis

Statistical analysis was performed with Excel (Microsoft Office 365) and IBM SPSS Statistics 29.0 (Chicago, IL, USA). The data were logarithmically transformed and normality confirmed prior to application of one-way analysis of variance (ANOVA) or *t*-test as appropriate. Only data for those BFRs with a detection frequency (DF) exceeding 50 % were included in statistical analyses, with values where BFR concentrations were below LOQ designated as 0.5 × LOQ.

3. Results and discussion

3.1. BFRs in dust inside UK e-waste facilities

Concentrations of BFRs in dust collected from inside Facility #1 are shown in Fig. 3 and Table S5. PBDEs, predominantly BDE-209, were the most abundant BFRs, contributing an average of 51 % to total BFRs. This was followed closely by NBFRs (principally decabromodiphenyl ethane (DBDPE)), which accounted for 44 % of total BFRs on average; while HBCDDs (mainly γ-HBCDD) only contributed 5 % of total BFRs. Average concentrations of PBDEs, NBFRs, and HBCDDs were 6300 ng/g, 5500 ng/g, and 600 ng/g in dust collected from Facility #1, with the corresponding median values being 5600 ng/g, 1100 ng/g, and 500 ng/g, respectively. These concentrations were generally higher than BFR concentrations reported in UK house dust in 2019 (see Table S6 for the concentrations) (Drage et al., 2020), but no significant differences were observed with an independent samples *t*-test ($p > 0.05$). This is likely because Facility #1 has only been used for storage of e-waste, while dismantling and recycling of the e-waste were done elsewhere. While recognising the limited number of indoor dust samples analysed in this study, this study provides preliminary insights into BFR contamination inside UK e-waste facilities, and further investigations are encouraged to help better understand BFR contamination inside UK e-waste facilities.

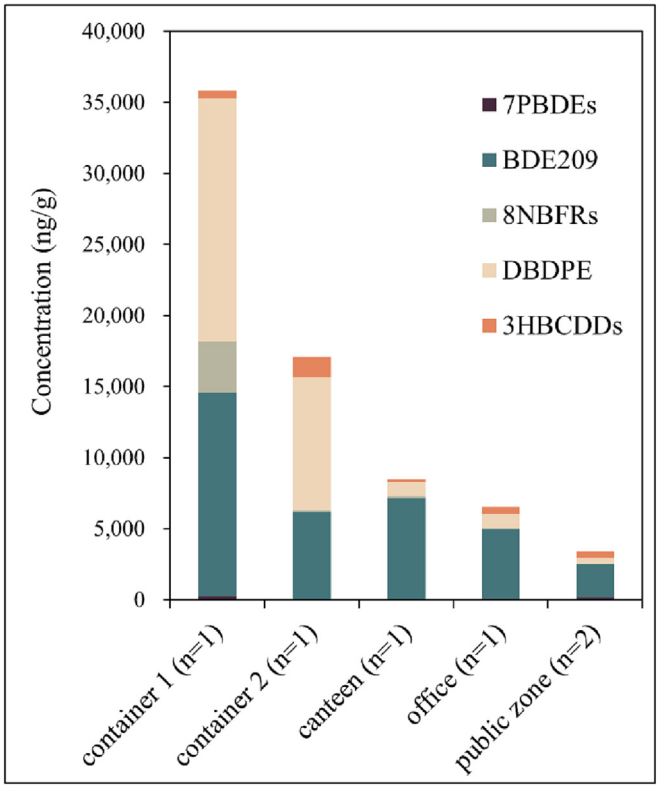

Fig. 3. Average concentrations of BFRs in dust inside UK e-waste recycling facilities; 7PBDEs are sum of BDE-28, -47, -99, -100, -153, -154, and -183; 8NBFRs are sum of PBBz, PBT, PBEB, DPTE, HBBz, EH-TBB, BTBPE, and BEH-TEBP; 3HBCDDs are sum of α-, β-, and γ-HBCDD; container 1 is used for storage of TVs & monitors; container 2 is used for storage of fridges & freezers.

The highest concentrations of BFRs were observed in dust collected from two containers used for storage of e-waste. Dust from container #1, which was used for storage of waste TVs and monitors, had higher concentrations of PBDEs (15,000 ng/g) and DBDPE (17,000 ng/g) than dust from container #2 (PBDEs: 6200 ng/g; DBDPE: 9400 ng/g), which was used to store waste fridges and freezers. Similar findings to our observations were also reported in a previous study, where considerably higher concentrations of PBDEs (DBDPE was not reported though) were found in plastics of waste displays than in plastics of waste large household appliances in Ireland (Drage et al., 2018). However, dust samples from containers #1 and #2 presented similar relative contributions of PBDEs (41 % vs 36 %) and DBDPE (48 % vs 55 %) to total BFRs. Interestingly, higher concentrations (3600 ng/g vs 98 ng/g) and relative abundance (10 % vs 0.58 %) of Σ_8NBFRs (DBDPE excluded; predominantly BTBPE), but lower concentrations (530 ng/g vs 1400 ng/g) and relative abundance (1.5 % vs 8.3 %) of HBCDD, were found in the dust from container #1 than in that from container #2. A possible explanation could be the much shorter lifespan of TVs and monitors (5–10 years) (US EPA Office of Resource Conservation and Recovery, 2011) than that of fridges and freezers (15 years) (Bawakyillenuo and Agbelie, 2016), which might allow more NBFRs, but not HBCDD, to enter the e-waste stream via waste TVs and monitors.

As shown in Table S6, indoor dust concentrations of PBDEs observed in this study were considerably lower than those reported inside four e-waste storage facilities in Thailand, while concentrations of DBDPE in indoor dust

observed in this study were comparable to those observed in the Thai e-waste storage facilities (Muenhor et al., 2010). Further, our observed PBDE and bis (2-ethyl hexyl) tetrabromophthalate (BEH-TEBP) concentrations were one to two orders of magnitude lower than those found in floor dust (no e-waste recycling or dismantling activities were conducted in the areas where floor dust was collected) of a Canadian e-waste facility (Nguyen et al., 2019). When comparing with other facilities where e-waste was dismantled or recycled, indoor dust concentrations of PBDEs observed in this study were also considerably lower than those reported inside both regulated and unregulated e-waste recycling facilities in China (Xu et al., 2015; Zheng et al., 2015; Zhou et al., 2022), Vietnam (Wannomai et al., 2020), and Canada (Stubbings et al., 2019). Moreover, concentrations of 1,2-bis (2,4,6-tribromophenoxy) ethane (BTBPE), BEH-TEBP, and DBDPE in indoor dust inside Facility #1 were at the lower end of those observed in regulated e-waste recycling facilities in Canada (Stubbings et al., 2019) and in unregulated e-waste recycling facilities in China (Xu et al., 2015; Zheng et al., 2015) and Vietnam (Wannomai et al., 2020).

3.2. BFRs in kerb dust outside UK e-waste recycling facilities

Average concentrations of PBDEs, NBFRs, and HBCDDs were 100 ng/g (2,900 ng/g OC, normalised to the organic carbon content of each dust sample), 66 ng/g (1,700 ng/g OC), and 11 ng/g (250 ng/g OC) in dust collected outside of UK e-waste facilities, with the corresponding median concentrations of 22 ng/g (430 ng/g OC), 47 ng/g (950 ng/g OC), and 6.3 ng/g (140 ng/g OC), respectively (Table S5). As this is the first study reporting BFRs in kerb dust in the UK, comparisons between our observations and BFR concentrations in non-e-waste impacted kerb dust in the UK are unlikely. However, urban soil samples collected from Birmingham, UK could be a reasonable substitute when OC normalised concentrations were used for comparison. Specifically, these concentrations were generally 2 to 3 orders of magnitude higher than OC normalised concentrations of BFRs in urban soils collected from sites with no direct input sources in Birmingham, UK (Table S6) (Drage et al., 2016). An independent samples *t*-test revealed significantly lower concentrations of BFRs in dust collected from kerbs outside Facility #1 than in dust collected inside the same facility (p = 0.001–0.029). Despite the significant differences in absolute concentrations of BFRs, similar profiles of BFRs were observed in dust sampled outside vs inside Facility #1 (Fig. S1). Overall, these results seem to suggest that e-waste recycling activities may be a substantial source of BFR contamination to the surrounding environment.

To verify this, BFRs in kerb dust samples were compared both upwind and downwind of each facility at distances from the sampling sites to the corresponding facility of 250 m, 500 m, and 1000 m, respectively (Fig. 2). A paired samples *t*-test revealed no significant difference between the upwind concentrations and the downwind concentrations of BFRs in dust near the UK e-waste recycling facilities (p = 0.134–0.976; see Table S7 for the concentrations). No significant correlations were observed with Spearman's rho ($p > 0.05$) between BFR concentrations in kerb dust and distances from Facilities #1, #2, #3, and #4, either suggesting a lack of significant impact of these e-waste facilities on BFR contamination of surrounding environment, or indicating the number of samples analysed in this study too small to allow any significant impacts to be observed. However, negative correlations between BFR concentrations and distance from facility #5 were observed for all the target BFRs (Spearman's rho; r = −0.239 to −0.837). These negative correlations were statistically significant for BDE-47, −99, and −100 ($p < 0.05$). Such observations indicated significant impacts of e-waste recycling activities in Facility #5 on BFR contamination in kerb dust. Unlike Facility #1, which is only used for e-waste storage, Facility #5 receives a wide range of e-waste for recycling, including small devices, large domestic appliances, etc. It remains unclear how much e-waste is recycled in Facility #5 per day, but it is reasonable to presume that Facility #5 receives a much larger volume of e-waste compared to Facilities #1, #2, #3, and #4, as Facility #5 is located in a city with a much larger population (>1000,000 vs <200,000). Overall, our observations could be strong evidence of e-waste recycling facilities as a potential source of BFR contamination to the urban environment in the UK.

Facility #5, located in an industrial city in the UK, also had significantly higher BFR concentrations in kerb dust than did other facilities (ANOVA, p = 0.0028). This might indicate industrial activities as another potential source of BFR contamination to the urban environment in the UK. To confirm this, BFR concentrations in kerb dust from different areas (categorized as: industrial, housing, supermarket, park, and agricultural) were compared (Fig. 4). Higher concentrations of BFRs (average: 450 ng/g vs 40–110 ng/g) were found in dust from industrial areas compared to non-industrial areas. An independent samples *t*-test revealed a significant difference in concentrations of NBFRs (p = 0.044), but not PBDEs (p = 0.253) and HBCDDs (p = 0.983), between industrial areas and non-industrial areas. This could reflect the phase-out of PBDEs and HBCDDs and the continuous use of NBFRs in industrial activities, and likely indicated industrial activities as a potential source of NBFR contamination. Given the limited sample size of this study, further investigations are encouraged to confirm this.

Concentrations of PBDEs and NBFRs in kerb dust observed here were generally lower than those observed in surface soil surrounding two regulated e-waste recycling facilities in Australia (McGrath et al., 2018), or in kerb dust and surface soil in the vicinity of informal e-waste facilities in China (Ling et al., 2022; Tang et al., 2016; Xu et al., 2015) and Vietnam (Matsukami et al., 2017). However, our observations of PBDE and NBFR concentrations in kerb dust were comparable to or slightly higher than the concentrations determined in urban road dust in China (Cao et al., 2017) and Vietnam (Anh et al., 2018).

3.3. Occupational exposure to BFRs via dust ingestion

Occupational exposure of UK e-waste recycling workers to BFRs via dust ingestion was estimated by using Eq. (1). Average BFR concentrations in

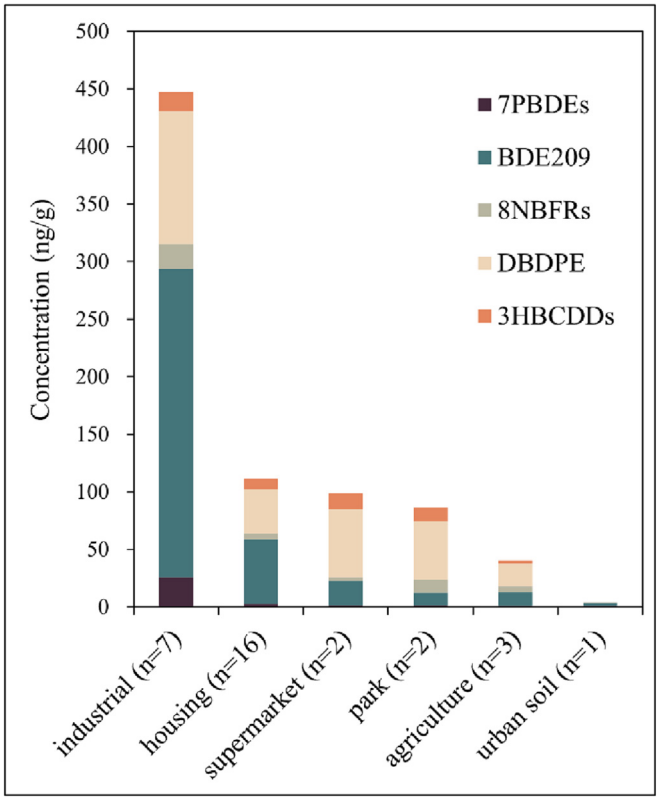

Fig. 4. Average concentrations of BFRs in kerb dust from different location categories in the vicinity of UK e-waste recycling facilities, and comparison with PBDE and DBDPE concentrations in an urban soil in the UK (Drage et al., 2016); 7PBDEs are sum of BDE-28, -47, -99, -100, -153, -154, and -183; 8NBFRs are sum of PBBz, PBT, PBEB, DPTE, HBBz, EH-TBB, BTBPE, and BEH-TEBP; 3HBCDDs are sum of α-, β-, and γ-HBCDD.

indoor dust (or kerb dust) and the recommended central tendency of dust ingestion rate (20 mg/day for indoor dust and 10 mg/day for kerb dust) (US EPA, 2017) were assumed for the average exposure scenario; while the 95th percentile for BFR concentrations in indoor dust and the recommended upper percentile dust ingestion rate (60 mg/day for indoor dust and 50 mg/day for kerb dust) (US EPA, 2017) were assumed to derive the upper-bound estimates of BFR exposure.

Under the average exposure scenario, daily intake of BFRs via indoor dust ingestion was estimated to be 0.98 ng/kg bw/day for male workers and 1.2 ng/kg bw/day for female workers, with the upper-bound estimates of daily intake of BFRs of 7.5 ng/kg bw/day for males and 8.8 ng/kg bw/day for females, respectively. By comparison, daily intake of BFRs via kerb dust ingestion contributed <5 % to dust ingestion of BFRs (sum of indoor dust ingestion and kerb dust ingestion) of UK e-waste workers (Table 1). BDE-209 and DBDPE predominated dust ingestion of BFRs for UK e-waste recycling workers, together contributing an average of 88 % to daily intake of total BFRs.

As shown in Table S8, our estimates of dust ingestion of PBDEs for UK e-waste recycling workers were generally one order of magnitude lower than those for e-waste recyclers working in formal and informal e-waste recycling facilities in Canada (Nguyen et al., 2019), China (Xu et al., 2015; Zheng et al., 2015; Zhou et al., 2022), and Vietnam (Wannomai et al., 2020); but were comparable to dust ingestion of PBDEs estimated for UK office workers (Tao et al., 2016). In terms of dust ingestion of DBDPE, our estimates for UK e-waste recyclers were at the lower bound of dust ingestion of DBDPE estimated for informal e-waste recycling workers in China (Xu et al., 2015; Zheng et al., 2015), and were considerably lower than the estimates for informal e-waste recycling workers in Vietnam (Wannomai et al., 2020). Our estimates of dust ingestion of DBDPE for UK e-waste recyclers were also similar to the estimations generated for office workers in China (Sun et al., 2018) and the UK (Tao et al., 2016).

Table S9 shows the reference doses (RfDs) for BFRs of interest. Our estimates of BFR exposure doses were considerably lower than the RfDs (HI < 0.001, see Table 1), suggesting minimal health burdens posed by occupational exposure to BFRs to UK e-waste recycling workers. However, it is notable that dust ingestion of BFRs during working hours is likely to contribute only a small proportion to total BFR exposure for UK e-waste recyclers, especially given comparable or even higher concentrations of BFRs than those in this study have been reported in house dust in the UK (Drage et al., 2020; Tao et al., 2016). Also, we realise the limitations of this study, e.g., small sample size and lack of estimates of BFR exposure via inhalation and dermal uptake, could result in underestimation of occupational exposure to BFRs for UK e-waste recycling workers. Further investigations are encouraged to fill the gap.

Table 1
Occupational exposure of UK e-waste recycling workers to BFRs via dust ingestion (ng/kg bw/day).

Scenarios	Σ_7PBDEs[a]	BDE-209	Σ_8NBFRs[b]	DBDPE	Σ_3HBCDDs[c]
Average estimates					
Male (indoor dust ingestion)	0.010	0.49	0.052	0.38	0.047
Male (kerb dust ingestion)	0.00091	0.011	0.0011	0.0067	0.0012
Male (HI)	<0.001	<0.001	<0.001	<0.001	<0.001
Female (indoor dust ingestion)	0.012	0.58	0.061	0.45	0.055
Female (kerb dust ingestion)	0.0011	0.013	0.0013	0.0079	0.0015
Female (HI)	<0.001	<0.001	<0.001	<0.001	<0.001
Upper-bound estimates					
Male (indoor dust ingestion)	0.080	2.9	0.66	3.6	0.30
Male (kerb dust ingestion)	0.0075	0.16	0.010	0.074	0.020
Male (HI)	<0.001	<0.001	<0.001	<0.001	<0.001
Female (indoor dust ingestion)	0.094	3.5	0.78	4.2	0.36
Female (kerb dust ingestion)	0.0089	0.19	0.012	0.088	0.024
Female (HI)	<0.001	<0.001	<0.001	<0.001	<0.001

[a] Sum of BDE-28, -47, -99, -100, -153, -154, and -183.
[b] Sum of PBBz, PBT, PBEB, DPTE, HBBz, EH-TBB, BTBPE, and BEH-TEBP.
[c] Sum of α-, β-, and γ-HBCDD.

CRediT authorship contribution statement

Yulong Ma: Methodology, Validation, Formal analysis, Investigation, Visualization, Writing – original draft. **Ester Konecna:** Methodology, Validation, Formal analysis, Investigation, Visualization. **Reginald Cline-Cole:** Supervision, Writing – review & editing. **Stuart Harrad:** Conceptualization, Supervision, Writing – review & editing, Project administration, Funding acquisition. **Mohamed Abou-Elwafa Abdallah:** Conceptualization, Supervision, Writing – review & editing, Project administration, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.165403.

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