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From e-waste to living space: Flame retardants contaminating household items add to concern about plastic recycling

Megan Liu [a,*], Sicco H. Brandsma [b], Erika Schreder [a]

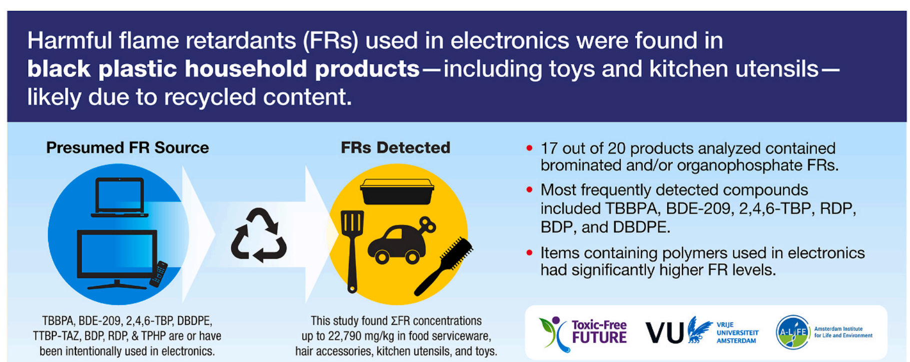
[a] Toxic-Free-Future, 4649 Sunnyside Ave N Suite 540, Seattle, WA 98103, United States

[b] Amsterdam Institute for Life and Environment, Vrije Universiteit Amsterdam, De Boelelaan 1108, 1081, HZ, Amsterdam, the Netherlands

HIGHLIGHTS

- FRs specific to electronics were detected in non-electronic household items bought in the U.S.
- 85% of products with >50 ppm Br contained toxic FRs.
- $\sum$FR concentrations ranged up to 22,800 mg/kg.
- Items containing polymers used in electronics had significantly higher FR levels.

GRAPHICAL ABSTRACT



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ABSTRACT

Brominated flame retardants (BFRs) and organophosphate flame retardants (OPFRs) are commonly used in electric and electronic products in high concentrations to prevent or retard fire. Health concerns related to flame retardants (FRs) include carcinogenicity, endocrine disruption, neurotoxicity, and reproductive and developmental toxicity. Globally, a lack of transparency related to chemicals in products and limited restrictions on use of FRs in electronics have led to widespread use and dissemination of harmful FRs. Despite the lack of transparency and restrictions, plastics from electronics are often recycled and can be incorporated in household items that do not require flame retardancy, resulting in potentially high and unnecessary exposure. This study sought to determine whether black plastic household products sold on the U.S. market contained emerging and phased-out FRs and whether polymer type was predictive of contamination. A total of 203 products were screened for bromine (Br), and products containing >50 ppm Br were analyzed for BFRs, OPFRs, and plastic polymers (e.g. acrylonitrile butadiene styrene, high impact polystyrene, polypropylene). FRs were found in 85% of analyzed products, with total FR concentrations ranging up to 22,800 mg/kg. FRs detected include the restricted compound deca-BDE, which was used widely in electronics casings, as well as its replacements decabromodiphenyl ethane (DBDPE) and 2,4,6-Tris(2,4,6-tribromophenoxy)-1,3,5-triazine (TBPP-TAZ) along with associated compound 2,4,6-tribromophenol (2,4,6-TBP), recently detected in breast milk. Plastic typically used in electronics (styrene-based) contained significantly higher levels of $\sum$FRs than plastics less typically used for electronics

* Corresponding author. Toxic-Free-Future, 4649 Sunnyside Ave N Suite 540 Seattle, WA 98103, United States.
E-mail address: mliu@toxicfreefuture.org (M. Liu).

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(polypropylene and nylon). Estimation of exposure to BDE-209 from contaminated kitchen utensils indicated users would have a median intake of 34,700 ng/day, exceeding estimates for intake from dust and diet. The detection of FRs in collected household products indicates that recycling, without the necessary transparency and restrictions to ensure safety, is resulting in unexpected exposure to toxic flame retardants in household items.

1. Introduction

Plastics can consist of a complex mixture of monomers, unreacted intermediates, and additives such as dyes, fillers, antioxidants, flame retardants (FRs), UV stabilizers, surfactants, and plasticizers (Ignatyev et al., 2014). Many additives migrate over the course of the plastic lifecycle, including production, use, and disposal, posing threats to human and environmental health. The use of FRs in particular has generated concern due to their widespread detection in the environment and associated human exposure, particularly since a large proportion of the use is additive rather than reactive, easing migration into the environment (Fromme et al., 2016; Ionas et al., 2014; Lyche et al., 2015). FRs have been detected in many environmental samples including indoor and outdoor air (Gravel et al., 2019b; Ma et al., 2012; Salamova and Hites, 2011; Venier et al., 2016), indoor dust (Cristale et al., 2016, 2018; Harrad et al., 2008; Mitro et al., 2016; Stapleton et al., 2008), sediment (Counihan et al., 2014; Guo et al., 2020; Zhu et al., 2018), water (Alvarez et al., 2014; Guo et al., 2017), and wastewater influent and effluent (Schreder and La Guardia, 2014). They have also been detected in biota, including fish (Babut et al., 2021; O'Neill et al., 2020), marine mammals (Jenssen et al., 2007), and bird eggs (Guerra et al., 2012; Klosterhaus et al., 2012; Su et al., 2015), as well as in human serum, breast milk (Schreder et al., 2023; She et al., 2007; Zhou et al., 2014), and placenta (Leonetti et al., 2016a).

Brominated flame retardants (BFRs) are a particularly concerning class of FRs due to their toxicity, persistence, and tendency to bioaccumulate. A number of BFRs are associated with human health concerns, including carcinogenicity, endocrine disruption, neurotoxicity, and reproductive and developmental toxicity (Center for International Environmental Law; Leonetti et al., 2016b; Li et al., 2014). BFRs are heavily used in electrical and electronic equipment such as plastic housings and printed circuit boards (Birnbaum and Staskal, 2004). Published product testing is limited, but available testing, as well as other sources of information, indicate that the dominant BFRs in TV and computer housings included decabromodiphenyl ether (deca-BDE) and tetrabromobisphenol A (TBBPA) (Department of Ecology and Department of Health, 2006; Gallen et al., 2014). Deca-BDE was considered the dominant FR in electronics casings until the European Union banned use in electronics in 2006 and U.S. states began to ban its use in 2007 (European Parliament and of the Council of the European Union, 2003; Washington State Legislature, 2007). After this phaseout in some jurisdictions, analysis of the plastic housings of TVs available on the U.S. market found that replacement BFRs dominated, particularly decabromodiphenyl ethane (DBDPE), 2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5-triazine (TTBP-TAZ), and 2,4,6-tribromophenol (2,4,6-TBP), along with less prevalent use of organophosphate flame retardants (OPFRs) resorcinol bis(diphenyl phosphate) (RDP) and bisphenol A bis(diphenyl phosphate) (BDP) (Schreder et al., 2017; Schreder and Uding, 2019). These halogen-free FRs are listed on the TCO Certified Accepted Substance List, used by companies committed to safer solutions (TCO Certified, 2023). Hazard assessments under GreenScreen® and the ENFIRO Project have assessed RDP and BDP as safer substitutes for the organohalogen FRs but noted concerns regarding the aquatic toxicity of these compounds (European Commission, 2013; GreenScreen for Safer Chemicals, 2023).

These BFRs and OPFRs are marketed for use in electronics or plastics associated with electronics (such as acrylonitrile butadiene styrene (ABS) and high impact polystyrene (HIPS)) by a variety of chemical manufacturers (Albemarle; ICL; Lanxess; UL Prospector; Unibrom). While this use is intentional, there are concerns that when electronic products are recycled, BFRs contained in plastic components may unintentionally be incorporated in other plastic household items, potentially in violation of local or national laws (EU Monitor, 2022; Official Journal of the European Union, 2019b). Studies in Africa, Asia, and Europe have found BFRs in black plastic hair accessories, kitchen utensils, toys, and office supplies (Fatunsin et al., 2020; Guzzonato et al., 2017; Jitka et al., 2022; Kuang et al., 2018; Petrlik et al., 2022; Puype et al., 2015), all of which are product categories without intentional FR use. The detections of BFRs associated with electronics in these household items suggests recycled content from electronics, such as the black plastic housings, as a likely source of contamination through electronic waste (e-waste) recycling. FRs have been detected in and near e-waste recycling facilities, in indoor air and dust at formal e-waste recycling facilities in Canada, China, Spain, and the U.S. (Balasch et al., 2022; Gravel et al., 2019a; Li et al., 2023; Nguyen et al., 2019; Zheng et al., 2015) and in soil samples surrounding e-waste recycling sites in China and Vietnam (Ge et al., 2020; Someya et al., 2016). These detections provide further evidence that recycling of FR-containing electronics results in human and environmental exposure.

Although some jurisdictions, such as the E.U., limit concentrations of some BFRs in waste plastics entering the recycling stream, such regulations are limited and fairly rare (Drage et al., 2022; Official Journal of the European Union, 2019b). In the U.S., some states have taken action to restrict certain FRs in new products, but state and federal policy preventing harmful chemicals from entering the recycling stream are lacking. The USEPA's recent rule banning deca-BDE specifically excludes plastics entering the recycling stream or containing recycled content from the limits in the rule (Environmental Protection Agency, 2021).

This study explored the presence of FRs in plastic household items on the U.S. market, particularly in items with high exposure potential, hypothesizing that some black plastic products on the U.S. market would be contaminated with emerging, phased-out, and commonly-used FRs and that products containing electronics-associated polymers would have higher levels of contamination. The potential presence of BFRs in food-contact materials is particularly concerning, since FRs may leach during use (Kuang et al., 2018). BFRs in toys also pose a great concern, with migration tests indicating FRs can leach into children's saliva through mouthing of toys (Brandsma et al., 2022). To investigate whether U.S. consumer products that do not require flame retardancy are contaminated with novel or phased-out FRs, a selection of food-contact items, hair accessories, kitchen utensils, and toys was analyzed for the presence of 20 FRs. This study does not solely include regulated FRs but also focuses on a wide range of FRs that substituted the phased-out commercial BDE mixture. Products were purchased from both large and small retailers to investigate whether different supply chains would result in differential levels of contamination.

2. Methods

2.1. Sample selection

Analytes were chosen based on reports of FRs detected in TVs and other electronics and a review of manufacturer websites for FRs marketed for use in electronics (Table S1). Product types were selected based on prevalence of black plastic, since most electronics enclosures are made of black plastic, and previous findings of FR contamination (Jitka et al., 2022; Petrlik et al., 2022; Turner, 2018).

A total of 203 black plastic products (food serviceware, n = 28; hair accessories, n = 30; kitchen utensils, n = 109; toys, n = 36) were purchased from online retailers and local stores in and around Seattle, USA from 2020 to 2022 (Table 1). These included 105 products purchased in-person from local, non-chain retailers and 98 products purchased in-person or online from large chain retailers. Products were purchased from both small, local shops as well as national, chain retailers to obtain a sample composed of products from different supply chains. We hypothesized larger retailers may have more stringent controls preventing unintentional contamination and aimed for a sample representative of products used by people from a range of demographics, including immigrant communities. Product purchasing began in 2020 but paused due to the COVID-19 pandemic and resumed in 2022.

Products purchased were either composed entirely of black plastic or had black plastic components (e.g. toy cars were often made with different colors of plastic but with black plastic undersides). Product information was catalogued including brand, retailer, country of manufacture, and if available, polymer type.

2.2. XRF screening and selection of samples for FR analysis

All 203 products were screened for Br using an Olympus Innov-X Alpha 6500 x-ray fluorescence spectrometer (XRF). Antimony (Sb) was also screened for by XRF as Sb is often used as a synergist with BFRs, thus presence of Sb can provide further indication of BFR presence. Heavy metals (cadmium, chromium, and lead) in ABS resin standards at both high and low concentrations were routinely tested with the XRF prior to product screening to monitor accuracy. Each product was screened using a 60 s test in RoHS/WEEE mode in at least 3 different spots. Items with Br detections were screened again in the same spot for confirmation. The 20 items with the highest Br concentrations all contained >50 ppm Br and were selected for FR analysis (we made an a priori decision to limit FR analysis to 20 items).

2.3. Sample preparation for FR analysis

Before extraction, a dissolving experiment was performed using toluene and dichloromethane (DCM) (Table S2). Plastics that dissolved in toluene were treated as follows: approximately 50 mg of the plastic sample was cut into small pieces (<5 mm), accurately weighed, and transferred into a 10 mL glass tube. After adding 5 mL of toluene, the samples were vortex mixed for 1 min, followed by ultrasonication for 10 min. For the plastics that did not dissolve in toluene, the samples were also cut into small pieces (<5 mm) and extracted overnight (12 h) in 3 mL of DCM on an orbital shaker at 250 revolutions per minute (RPM). After extraction, the DCM was transferred into a new glass tube and evaporated to a volume of 0.1 mL at 30 °C under nitrogen. The plastic sample was then re-extracted with 5 mL of toluene, as described above, and quantitatively combined with the 0.1 mL of DCM extract.

The above-prepared extracts were further diluted depending on the Br content (XRF values) in acetonitrile (ACN). Finally, 60 μL of labeled standards solution (1000 ng/mL; $^{13}C_{12}$-HBCDD, $^{13}C_{12}$-TBBPA, $^{13}C_{12}$-BDE209, $^{13}C_{12}$-BDBPE, $^{13}C_{12}$-BTBPE, $^{13}C_{18}$-TTBP-TAZ, $^{13}C_{6}$-246-TBP and TPHP-$_{d15}$) was added to the final dilution and evaporated until 10 μL of toluene, and reconstructed in 600 μL ACN for quantitative analysis (see Table 3 for full chemical names and acronyms, and Table S5 for CAS numbers of labeled standards).

Table 2
Br levels detected by XRF.

	Product Name	Product Category	Br (ppm)	Sb (ppm)[a]
1	Sushi Tray	Food Serviceware	18,600	3230
2	Toy Car	Toys	5410	ND
3	Peeler	Kitchen Utensils	4480	1140
4	Travel Checkers Set	Toys	4350	993
5	Pirate Coin Medallion Beads	Toys	3600	ND
6	Peeler	Kitchen Utensils	3280	ND
7	Slotted Spoon	Kitchen Utensils	1100	ND
8	Slotted Turner	Kitchen Utensils	1010	ND
9	Basting Spoon	Kitchen Utensils	993	ND
10	Pasta Server	Kitchen Utensils	957	ND
11	Laid Back Party Beads- Charm	Toys	682	ND
12	Slotted Turner	Kitchen Utensils	594	ND
13	Slotted Turner	Kitchen Utensils	528	ND
14	Slotted Spoon	Kitchen Utensils	144	ND
15	Laid Back Party Beads- Beads	Toys	136	ND
16	Mini Tabletop Pool	Toys	133	ND
17	Pirate Hook	Toys	124	ND
18	Hair Brush	Hair Accessories	123	ND
19	Toy Car	Toys	69	ND
20	Fast Food Tray	Food Serviceware	51	ND

[a] ND = not detected.

2.4. FR analysis

BFRs and OPFRs were analyzed using a high-resolution quadrupole time-of-flight (qTOF) mass spectrometer (MS) (Compact, Bruker, Bremen, Germany) connected to an LC 1260 HPLC (Agilent, Amstelveen, the Netherlands). Measurement of OPFRs was performed in positive atmospheric pressure chemical ionization (APCI) mode and the BFRs in negative APCI mode. The mobile phase consisted of HPLC water and methanol using gradient conditions. The *m/z* values of the native and labeled FRs used for the quantification are given in Tables S3 and S4. Further detailed settings can be found in Brandsma et al. (2022).

2.5. QA/QC

Blanks were included and randomly analyzed with the samples. In the blanks, none of the target compounds were detected at levels exceeding the limit of detection (LOD). Consequently, the LOD and limit of quantification (LOQ) were calculated using a signal-to-noise ratio (S/N) of 3:1 and 10:1, respectively. The LOQ ranged from 5 to 20 ng absolute for the BFRs and OPFRs. The LOQs for each target compound within each sample in mg/kg are given in Table S5 and adjusted for the dilution of the samples according to their Br content. There is a lack of certified reference materials (CRM) for the targeted compounds included in this study. However, for the PBDEs, CRMs were available and ERM-EC591 was incorporated for this purpose. BDE-209, present in this CRM, served as a good representative for the BFRs, given its general difficulty in extraction. The measured value for BDE-209 in the ERM-EC591 was 707 mg/kg which is within the uncertainty of the assigned value (740 ± 80 mg/kg). Recovery of the labeled standards ($^{13}C_{12}$-

Table 1
Sample summary.

Product Category	# of Products Purchased from a Local Retailer	# of Products Purchased from a Chain Retailer	% of Products >50 mg/kg Br
Food Serviceware	11	17	7%
Hair Accessories	17	13	3%
Kitchen Utensils	55	54	8%
Toys	22	14	22%

Table 3
Summary of FRs detected.

FRs	Full Chemical Name	Detection Frequency (n = 20)	Median (mg/kg)	Mean (mg/kg)	Range (mg/kg)
TBBPA	Tetrabromobisphenol A	75%	145	260	<0.8–1370
BDE-209	Decabromodiphenyl ether	70%	103	1100	<2–11,900
2,4,6-TBP	2,4,6-Tribromophenol	70%	9.0	35	<0.4–310
RDP	Resorcinol bis(diphenylphosphate)	60%	158	2110	<8.0–15,300
BDP	Bisphenol-A bis(diphenyl phosphate)	60%	35	679	<2–4240
DBDPE	Decabromodiphenyl ethane	60%	33	366	<2–2220
TPHP	Triphenyl phosphate	55%	32	181	<2–1100
TTBP-TAZ	2,4,6-Tris(2,4,6-tribromophenoxy)-1,3,5-triazine	50%	7.0	56	<0.4–730
TBBPA-BDBPE	Tetrabromobisphenol A bis (2,3-dibromopropyl ether)	30%	<4.0	49	<4–850
BTBPE	1,1'-[ethane-1,2-diylbisoxy]bis[2,4,6-tribromobenzene]	5%	<4.0	20	<4–390
HBCD	α-Hexabromocyclododecane	5%	<2.0	14.5	<2–290

HBCD, $^{13}C_{12}$-TBBPA, $^{13}C_{12}$-BDE209, $^{13}C_{12}$-BDBPE, $^{13}C_{12}$-BTBPE, $^{13}C_{18}$-TTBP-TAZ, $^{13}C_6$-2,4,6-TBP, and TPHP-$_{d15}$) in the samples ranged from 68 to 129%, except for $^{13}C_6$-2,4,6-TBP which was lower (51% and 57%) in two samples. $^{13}C_6$-2,4,6-TBP was probably affected by the evaporation step, as 2,4,6-TBP is somewhat more volatile compared to the other BFRs included in this study. All values are corrected for the recovery of the labeled standards.

2.6. FTIR analysis

The spectra of the 20 plastic products were acquired using a non-destructive Fourier Transform Infrared spectrometer (FTIR LUMOS, Bruker, Germany) operating in attenuated total reflectance mode, equipped with Germanium attenuated total reflection (ATR) crystal. All spectra were recorded in the 600–4000 cm−1 range, 4 cm−1 resolution and 256 scans. Prior to acquiring sample spectra, the ATR crystal was situated in ambient air, and a background spectrum was recorded. Between each sample the ATR crystal was cleaned using 2-propanol. The data processing was performed using the OPUS-IR software (Bruker, version 7.5). Each sample was analyzed twice and the results are shown in Fig. 3.

2.7. Data analysis

Total Br content in measured BFRs was calculated by multiplying the number of Br atoms in each BFR compound with the atomic mass of Br (79.904 amu) to find the molecular weight of Br in each compound. BFR levels detected (in mg/kg) were then converted to moles and multiplied by the percent of Br in each respective compound to determine the weight of Br in grams.

The association between Br levels measured by XRF and total Br content measured in BFRs was analyzed using simple linear regression (R v. 2023.09.1 + 494). Paired sample t-tests were used to conduct hypothesis testing (Excel v. 16.79.2) to compare FR levels found in electronics-associated polymers to non-electronics associated polymers, and products purchased from small local retailers to products purchased from large chain retailers. For compounds with detection frequencies of at least 50%, non-detects were substituted with LODs for descriptive statistics and statistical analyses.

3. Results and discussion

3.1. Product screening

Overall, screening indicated that about 10% of the products likely contained BFRs at levels to be expected from use of FR-containing recycled content. XRF screening of the 203 products identified 20 products with >50 ppm Br, ranging up to 18,600 ppm (Table 2). Sb was detected in 3 out of the 203 products, each of which contained Br levels >4000 ppm. These screening results suggest that at least for BFRs (as we were unable to screen for OPFRs), a minority of black plastic products are contaminated at levels >50 ppm Br.

3.2. FRs in black plastic household products

FR analysis found a high prevalence of both BFRs and OPFRs in products >50 ppm Br. A mixture of both classes was found in 65% of the products, while only BFRs were found in 20%. ∑FR levels ranged from ND–22,800 mg/kg (Table S6). The products containing multiple FRs included food serviceware, toys, kitchen utensils, and a hair accessory. None of these products have any apparent need for flame retardant properties. Further, the common presence of mixtures of multiple BFRs and OPFRs suggests that these products are probably contaminated with recycled waste electronic and electrical equipment (WEEE) content.

3.2.1. Detections of phased-out and replacement FRs

Five BFRs and three OPFRs were detected in ≥50% of samples. This included BFRs TBBPA (detected in 75% of samples), 2,4,6-TBP (70%), BDE-209 (70%), DBDPE (60%), and TTBP-TAZ (50%), and OPFRs BDP (60%), RDP (60%), and TPHP (55%) (see Table 3 for a summary of the results). Except for TBBPA, all of the most frequently detected compounds mirror those also detected in television casings in the U.S. (Schreder et al., 2017; Schreder and Uding, 2019). Explanations for the frequent detection and high median level (145 mg/kg) of TBBPA include its use in other electronics casings, other possible or past use in television casings, or that its use in printed circuit boards contaminates recycled content. TBBPA was also commonly detected in plastic toys purchased in the U.K. (Fatunsin et al., 2020), and in Mardi Gras-type beads purchased in the U.S. (Miller et al., 2016). Globally, it is one of the most produced and used BFRs, with production volumes estimated at over 120,000 tonnes per year (European Food Safety Authority, 2011). It has been detected in TV casings (Gallen et al., 2014), displays (Drage et al., 2022), and small electronics and appliances including a foot warming pad, car and battery chargers, and hair and clothing irons (van Bergen and Stone, 2021).

The other frequently detected compounds included BDE-209 and its replacements. The frequent detection and high median level (103 mg/kg) of BDE-209 indicate that despite phaseouts in the U.S., E.U., and China, the compound is still commonly present in recycled WEEE content. BDE-209 levels ranged from <2 to 11,900 mg/kg, with the highest maximum level found in the Sushi Tray. To our knowledge, this is the first indication of BFR contamination of food serviceware purchased in the U.S. (in this case, from a restaurant supply website based in the U.S.). BDE-209 levels detected in this study were higher than previously detected levels in foodware and kitchen utensils from the U.K. (median 1.15 mg/kg) (Kuang et al., 2018), Arabic and African countries (median 33.3 mg/kg) (Petrlik et al., 2022), and China, Indonesia, and Russia (median 92 mg/kg) (Jitka et al., 2022).

Replacements for deca-BDE were frequently detected in these products, including the BFRs DBDPE, TTBP-TAZ and the associated compound 2,4,6-TBP, although typical levels of these compounds were lower (medians 33, 7.0, and 9.0 mg/kg, respectively). DBDPE has been found at high frequency (59% of TVs tested) and concentrations in TV casings (Schreder et al., 2017; Schreder and Uding, 2019) and smaller electronic appliances (van Bergen and Stone, 2021). TTBP-TAZ has also been found in TV casings at high concentrations (Schreder et al., 2017; Schreder and Uding, 2019). TTBP-TAZ was detected in 50% of the products, while 2,4,6-TBP, an impurity and degradation product of TTBP-TAZ, was found in 70% of products (Table 3). The higher levels and more frequent detection compared to TTBP-TAZ suggests that degradation of TTBP-TAZ or other BFRs into 2,4,6-TBP may be occurring in the recycling process. Several other BFRs were detected less frequently but had fairly high maximum concentrations: TBBPA-BDBPE (30%, max. 850 mg/kg), BTBPE (5%, max. 390 mg/kg), and HBCD (5%, max. 290 mg/kg).

The OPFRs RDP, BDP, and TPHP, which are used in electronics as replacements for deca-BDE (Blum et al., 2019), were frequently detected in the collected products. Median levels were 158 mg/kg for RDP, 35 mg/kg for BDP, and 32 mg/kg for TPHP, concentrations similar to those of the BFRs. RDP and BDP have been recommended as safer alternatives to BFRs (TCO Certified, 2023) while TPHP has not, due to association with developmental toxicity and endocrine disruption (Ji et al., 2022).

The detection of these replacement FRs in household products indicates that alternatives to deca-BDE are already present in e-waste recycling and contaminating household products through the recycling process. At the same time, BDE-209 is still circulating in the economy despite its phaseout in some markets. It is possible that manufacturers outside the U.S. and E.U. have been intentionally applying BDE-209 in new products and that the polymer ultimately enters the U.S. market in products with recycled e-waste content. While not all 20 products analyzed in this study contained country of origin information, 12 were made in China, which only began restricting the use of deca-BDE in 2023 (SafeGuardS, 2023). In any case, BDE-209 is still making its way into the recycling stream and the result is frequent contamination of products at levels of concern. In all 14 of the products containing BDE-209, levels would exceed the 10 mg/kg unintentional trace contamination limit established for the E.U. (Official Journal of the European Union, 2019b).

The FRs detected are commonly used in electrical and electronic equipment, suggesting poorly controlled e-waste recycling is resulting in contamination of household items with toxic chemicals that do not serve any function in those items. As a consequence, people are exposed to FRs not only from products with intentional use but also when FRs contaminate products that do not require flame retardancy.

3.2.2. Association between Br content and XRF Br levels

Linear regression assessing the relationship between XRF Br content and the sum of Br content in BFRs detected found a strong and significant association between the two variables ($p < 0.01$, $R^2 = 0.91$) (Fig. 2). There was some inconsistency in this dataset, including the three samples that had no FR detections but had XRF Br levels of 69, 144, and 682 ppm. Additionally, one sample contained 136 ppm Br by XRF but was found to contain an unexpectedly high level of total Br content in measured BFRs of 2260 mg/kg. Previous studies have analyzed the relationship between XRF Br content and Br content in BFRs measured, generally finding a significant correlation, but that XRF analysis typically overpredicted Br content measured by GC-MS (Kuang et al., 2018; Stapleton et al., 2011). The weaker correlation found in Kuang et al. ($r = 0.493$, $p = 0.006$), where $\sum$BFRs were always lower than corresponding Br measurements, was attributed to a limited BFR analyte list and low extraction efficiency. Stapleton et al. also found a significant correlation ($p < 0.001$), yet in seven samples where XRF Br levels ranged from 1.4 to 3.4% by weight, GC-MS only detected OPFRs (Stapleton et al., 2011). The stronger correlation found in our data may be due to improved extraction methods and a larger analyte list.

a. Pirate Coin Medallion Beads b. Tabletop Pool c. Slotted Turner d. Sushi Tray

Fig. 1. Examples of products containing >50 ppm Br as determined by XRF Fig. 1aPirate Coin Medallion Beads Fig. 1b. Tabletop Pool Fig. 1c. Slotted Turner Fig. 1dSushi Tray.

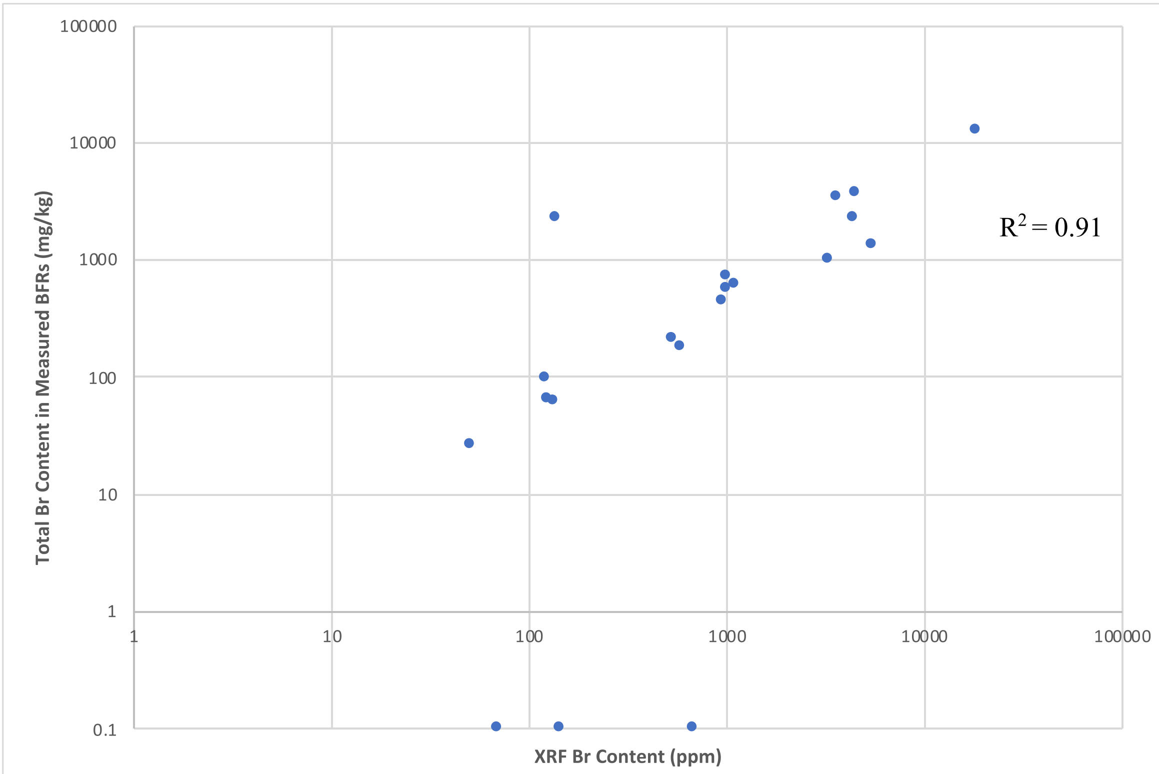

Fig. 2. Correlation between XRF Br levels and total Br content in BFRs measured.

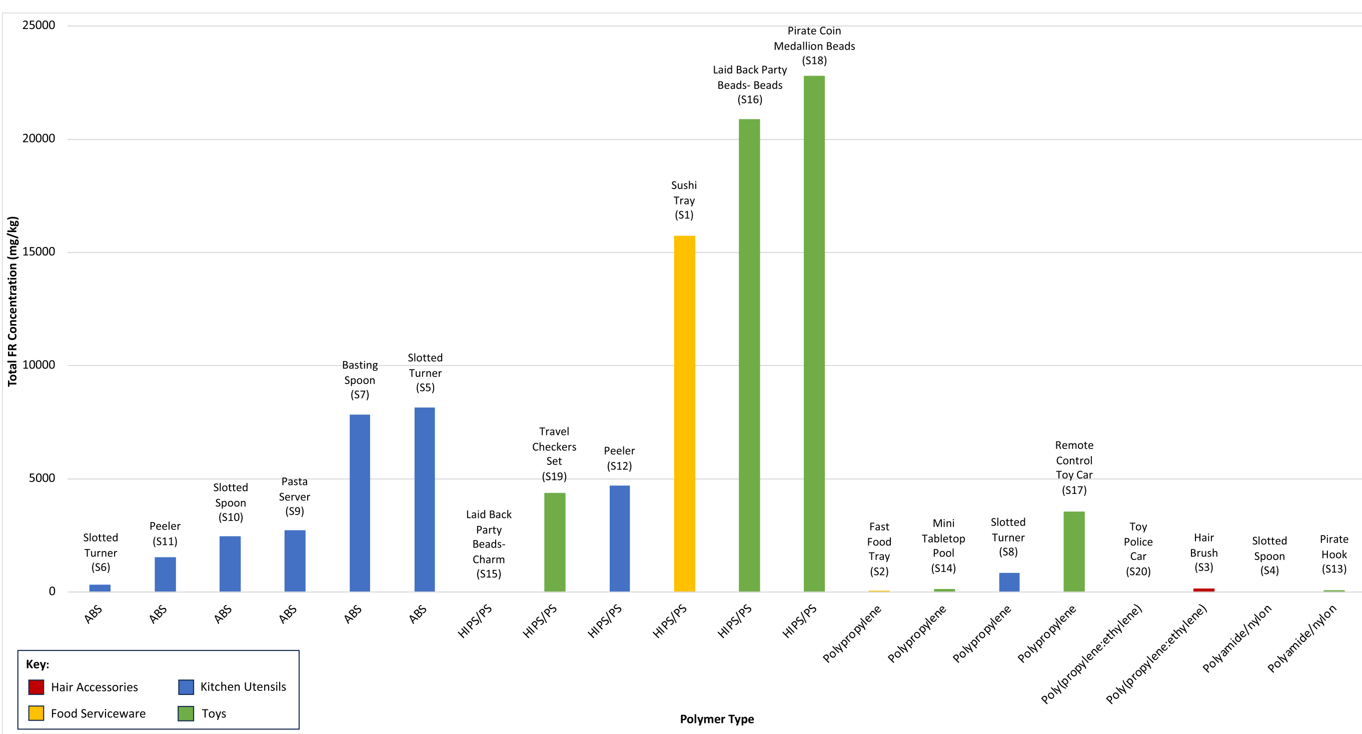

Fig. 3. Products are labeled with their sample numbers (S1, S2⋯S20), which can be referenced for more information on FR results in Tables S5 and S6.

3.3. Variation in FR detection by polymer type

Analysis for polymer type using FTIR of the 20 products that screened >50 ppm Br found that ABS and HIPS/PS were the dominant polymers (Fig. 2). ABS and HIPS/PS were found in 12 samples (60%), followed by polypropylene in 4 samples (20%). Less frequently detected polymers included poly(propylene:ethylene) (10%) and polyamide nylon (10%). Among these polymers, ABS and HIPS/PS are most associated with electronics (PINFA, 2017), and 11 out of 12 products identified as either ABS or HIPS/PS contained ∑FRs >310 mg/kg. Total FR content in ABS ranged from 310 to 8150 mg/kg, and from ND–22,800 mg/kg in HIPS/PS, which included the three products with the highest FR concentrations (all over 15,000 mg/kg).

Total FR levels in polymers more typically used in electronics (ABS and HIPS/PS) were compared with levels in polymers not generally associated with electronics (polypropylene and polyamide/nylon). Total FR levels in ABS and HIPS/PS, with a median of 4600 mg/kg, were significantly higher than those in polypropylene and polyamide/nylon, with a median of 150 mg/kg ($p < 0.01$) (see Fig. S1 for a visual comparison).

Most products screened and analyzed did not contain label information identifying plastic polymer composition. Only 5 out of the 20 products analyzed for FRs contained polymer information on their labels, and only the Sushi Tray (S1), labeled as polystyrene, matched with its FTIR result, HIPS/PS. The other four labeled products included a Slotted Turner (S6) and Peeler (S11) which were both labeled as polypropylene, and a Pasta Server (S9) and Slotted Spoon (S10) both labeled as nylon that were all identified as ABS by FTIR. The FTIR method used probed only the content of the top layer of a sample, so it is possible that the samples contain other plastic polymers, especially if they contain recycled content.

3.4. Association between FR levels and retailer type

Although we hypothesized that national retailers would be able to better control the level of FR contamination in household products, a slightly greater percentage of products purchased from small retailers (60%) screened as >50 ppm Br than from national retailers (40%). A summary of FR levels detected across retailer types can be found in Table S7. The median FR concentration in products purchased from large retailers was higher than that from small retailers (6150 vs. 885 mg/kg) (see Fig. S2 for a visual comparison), although the difference was not significant ($p = 0.07$). A number of large retailers have established chemical policies that restrict some chemicals of concern in some of the products they sell (Toxic-Free Future, 2019). However, to our knowledge these policies do not address chemicals in recycled content. Greater disclosure of ingredients and contaminants would be necessary for retailers to meaningfully act to prevent this type of contamination. Our results indicate that current policies have not successfully prevented the unintentional presence of FRs in household items that do not need flame retardancy.

While our results do not indicate that small retailers have a higher frequency of contaminated products, the presence of high FR concentrations in consumer products sold at small retailers catering to immigrant communities or specific ethnic groups is concerning. For example, a Peeler purchased at a retailer serving the Indian community contained >3500 mg/kg BDE-209, and a Pasta Server and Slotted Spoon purchased from a Middle Eastern grocery both contained BDE-209, DBDPE, and TBBPA. Products purchased from chain retailers are likely sold at other locations across the U.S., potentially exposing consumers nationally to FRs. Although our sample size was limited, the results appear to indicate that neither large nor small retailers have been able to meaningfully act to prevent this type of contamination in black plastic household products.

3.5. Health and exposure concerns

The presence of FRs in household products is concerning. In particular, deca-BDE has been the target of regulation to address concerns around carcinogenicity, endocrine disruption, neurotoxicity, and reproductive harm (Li et al., 2014). TBBPA, the compound found most frequently in our samples, has been found to cause cancer in laboratory studies (National Toxicology Program, 2014). Similarly, 2,4,6-TBP has been associated with thyroid disruption in humans and mice and has been detected in serum, breast milk, and placenta; TTBP-TAZ is converted to 2,4,6-TBP by human liver microsomes and may contribute to exposure (Butt et al., 2011; Hamers et al., 2006; Koch and Sures, 2018; Lee et al., 2016; Leonetti et al., 2016b; Schreder et al., 2023; Zheng et al., 2022). DBDPE is associated with reproductive and developmental toxicity, hepatotoxicity, thyrotoxicity, and neurotoxicity in laboratory animal studies (Shi et al., 2021; Xiangyang Li et al., 2021; Zhang et al., 2023), and exposure in workers has been associated with altered thyroid activity (Chen et al., 2019). These deca-BDE replacements have already been detected in electronics; dust from homes and electrical and electronic waste recycling facilities; and biota, including wildlife and people (Ballesteros-Gomez et al., 2014; Butt et al., 2016; Guo et al., 2018; Lippold et al., 2022; Vorkamp et al., 2019). In this study, most products containing FRs included a mixture of BFRs, raising concerns about additive or synergistic effects.

Migration of FRs from electronics has been documented, and the widespread presence of BDE-209 in indoor environments supports the conclusion that electronics are a key source of exposure (Rauert and Harrad, 2015). Exposure from products that do not need flame retardancy has not been extensively studied, but research indicates that FRs migrate from contaminated cooking utensils into food, and from toys into saliva (Brandsma et al., 2022; Kuang et al., 2018). Kuang et al. estimated daily exposure to BDE-209 from contaminated utensils after conducting migration experiments simulating the use of these utensils in hot oil. Applying the transfer rate derived in those experiments (11.7%) to the median concentration of BDE-209 in the cooking utensils in this study, we obtained an estimated daily intake of 34,700 ng/day from the use of contaminated utensils (see SI for methods). This compares to a $\sum$BDE intake in the U.S. of about 250 ng/day from home dust ingestion and about 50 ng/day from food (Besis and Samara, 2012) and would approach the U.S. BDE-209 reference dose of 7000 ng/kg bw/day (42, 000 ng/day for a 60 kg adult) (United States Environmental Protection Agency, 2008).

FRs have also been documented to migrate from toys into artificial saliva (Brandsma et al., 2022). With migration rates determined from subjecting contaminated toys to artificial saliva to mimic mouthing by children, Brandsma et al. estimated children's intake at up to 0.22 ng/kg bw/day for BDE-209, 0.88 ng/kg bw/day for 2,4,6-TBP, 0.03 ng/kg bw/day for TTBP-TAZ, and 2.28 ng/kg bw/day for TBBPA. These values were derived from the highest amount of migration detected, but may not represent the worst-case scenario, as Chen et al. (2019) detected greater migration using human volunteers.

4. Policy implications

The finding of multiple hazardous FRs in plastic consumer products points to the need for greater transparency in the supply chain, elimination of hazardous FR additives and a move to safer materials not requiring such additives. The detection of FRs in food serviceware, hair accessories, kitchen utensils, and toys, particularly in polymers associated with electronics enclosures, can logically be seen as e-waste-specific contamination, as FRs are not necessary in these products. The appearance of FRs in plastic consumer products is likely due to continuous use of hazardous FRs in plastics, lack of restrictions, and poorly controlled recycling practices.

While FRs continue to be widely produced and used, regulatory actions have started to target the most problematic compounds. According to reports submitted to the USEPA, U.S. manufacturers annually produce millions of kilograms of FRs currently used in electronics such as DBDPE, TBBPA, and RDP (United States Environmental Protection Agency, 2020; US Environmental Protection Agency, 2020) (see Table S8). Global consumption of FRs was greater than 2.25 million tonnes in 2017 and is forecast to grow by 3% a year, with electronics a major use (United Nations Environment Programme, 2019).

Within the last several years, regulatory bodies have begun to respond to concerns about the persistence, toxicity, and bioaccumulation of FRs, specifically organohalogen FRs, including brominated and chlorinated compounds. In 2019, the European Commission banned organohalogen FRs in electronic displays, including TVs and computer monitors (Official Journal of the European Union, 2019a). New York restricted the use of organohalogen FRs in electronic enclosures beginning in January 2024 (The New York State Senate, 2021), and Washington state's ban on the use of organohalogen FRs in enclosures for all indoor electric and electronic products goes into effect starting in 2025 (Washington State Department of Ecology, 2022).

While these regulatory actions target the most hazardous FRs, the high FR levels in household products found in this study further highlight the need for a regulatory system that moves manufacturers to the safest solutions. Unfortunately, when harmful FRs are used in TVs and other plastics, they may turn up where they are not expected, particularly when transparency across the supply chain is limited. To solve this problem, requirements are needed to disclose content of intentionally added chemicals and contaminants, restrict the intentional use of toxic FRs in products, prohibit hazardous chemicals in recycled content, and move manufacturers to the safest materials and chemicals.

5. Conclusion

These results show that when toxic additives are used in plastic, they can significantly contaminate products, made with recycled content, that do not require flame retardancy. Products found in this study to contain hazardous flame retardants included items with high exposure potential, including food-contact items as well as toys. Regulatory bodies have begun to address the use of certain classes of flame retardants (The New York State Senate, 2021; Washington State Department of Ecology, 2022), but more regulation is needed to end the use of hazardous additives and ensure that replacements are made with safer materials and chemicals.

CRediT authorship contribution statement

Megan Liu: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Sicco H. Brandsma:** Writing – review & editing, Resources, Methodology, Investigation, Formal analysis. **Erika Schreder:** Writing – review & editing, Visualization, Supervision, Methodology, Formal analysis, Conceptualization.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2024.143319.

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